ORSAY

n° d'ordre :

UNIVERSITÉ DE PARIS-SUD

CENTRE D'ORSAY

THÈSE

PRÉSENTÉE

POUR OBTENIR

Le GRADE de DOCTEUR ES-SCIENCES PHYSIQUES

PAR

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SUJET DE LA THÈSE : ETUDE THÉORIQUE ET MISE EN EVIDENCE EXPÉRIMENTALE DU PHÉNOMÈNE DE SUBRADIANCE

SOUTENUE LE 30 Mai 1986 DEVANT LA COMMISSION D'EXAMEN

M. S. Liberman, Président
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Ce travail a été réalisé au Laboratoire Aimé Cotton. Je tiens à exprimer toute ma reconnaissance à Sylvain Liberman, qui m'y a accueillie et m'a proposé le sujet de cette thèse. Il a ensuite dirigé l'évolution de celle-ci avec compétence, enthousiasme et bonne humeur; il m'a fait profiter à tout moment de sa grande sensibilité aux phénomènes physiques mais aussi aux petits problèmes de tous les jours. Je le remercie également d'avoir su, si souvent, m'écouter avec patience et gentillesse.

J'ai bénéficié pendant tout le déroulement de cette thèse de la compagnie stimulante d'Anne Crubellier: ce travail lui doit beaucoup. Je veux lui manifester ici mon admiration, qui n'a fait que s'accroître au cours de ces quatre ans de vie commune, pendant lesquels j'ai pu profiter de sa profonde compréhension de la physique, de sa grande culture, de son amour pour la précision scientifique, de sa disponibilité continue et de son amitié sincère qui m'a été si précieuse.

Je veux dire à Louis Cabaret combien sa présence a été importante pour moi: il m'a initiée aux mystères de la physique expérimentale, et c'est en bonne partie grâce à lui que l'expérience a pu être réalisée.

MM. Calvignac et Demarets ont assuré la clarté des dessins: je veux les remercier pour la rapidité et l'habileté qui caractérisent toujours leur travail.

Les membres de l'atelier mécanique m'ont bien souvent fourni, avant les pièces, l'étape préliminaire essentielle que constitue leur projet: qu'ils trouvent ici l'expression de mon estime sincère.

Je désire enfin manifester ma gratitude à tous les membres du laboratoire, chercheurs, enseignants, techniciens: ils ont suivi avec le sourire mes bégaiements (non seulement en ce qui concerne la langue française) et ne m'ont jamais refusé leur aide, qu'elle soit matérielle ou morale.

1. INTRODUCTION

1.1 L'émission spontanée coopérative

On considère l'expérience idéale d'un atome préparé, à un instant qui est pris comme origine des temps, dans un état excité, et laissé, à partir de cet instant, libre d'évoluer. La probabilité qu'il a d'émettre un photon décroît comme $e^{-\Gamma t}$, où Γ est la probabilité d'émission spontanée par unité de temps pour la transition considérée. Une telle expérience est évidemment difficile à réaliser avec un seul atome. Dans une expérience avec un grand nombre N d'atomes excités, pourvu que les atomes soient indépendants les uns des autres, l'émission globale est

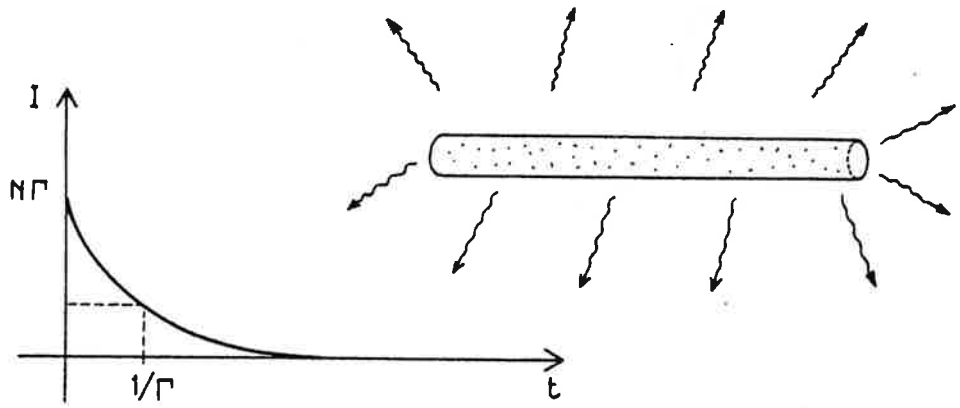

figure 1: émission spontanée non coopérative

la somme des émissions de chacun des N atomes, le signal détecté a l'allure d'une exponentielle décroissante de constante de temps $T_{SP} = 1/\Gamma$, et le rayonnement est essentiellement isotrope: c'est le phénomène d'émission spontanée ordinaire (figure 1). On observe généralement ces

caractéristiques dans l'émission des systèmes atomiques dilués.

Lorsque la densité d'émetteurs devient suffisamment importante, il est impossible de savoir lequel d'entre eux a émis un photon. Les atomes sont alors indiscernables vis–à–vis du champ rayonné, et ils interfèrent entre eux: l'émission spontanée devient coopérative. Cette propriété d'indiscernabilité va modifier sensiblement l'évolution globale du système. En particulier, la collection d'atomes va émettre son rayonnement par "bouffées", c'est à dire de manière beaucoup plus intense et rapide. L'émission présente une intensité maximale qui est

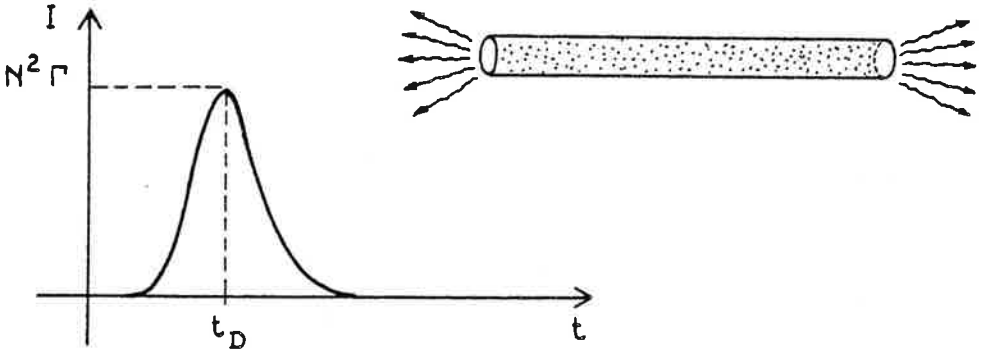

figure 2: émission spontanée coopérative

proportionnelle à N^2; elle est caractérisée par un temps plus court, $T_{SR} = 1/N\Gamma$ (figure 2), et elle a lieu dans des directions privilégiées qu'on appelle modes coopératifs. C'est le phénomène de superradiance, qui est, comme on va voir tout de suite dans le cas de deux atomes, le résultat d'une interférence interatomique constructive qui apparaît au cours de l'émission.

1.2 Superradiance et subradiance

Supposons qu'on ait préparé un système de deux atomes à deux niveaux de façon à avoir un atome dans chacun des deux niveaux (figure 3). Les deux atomes sont indiscernables vis-à-vis du rayonnement

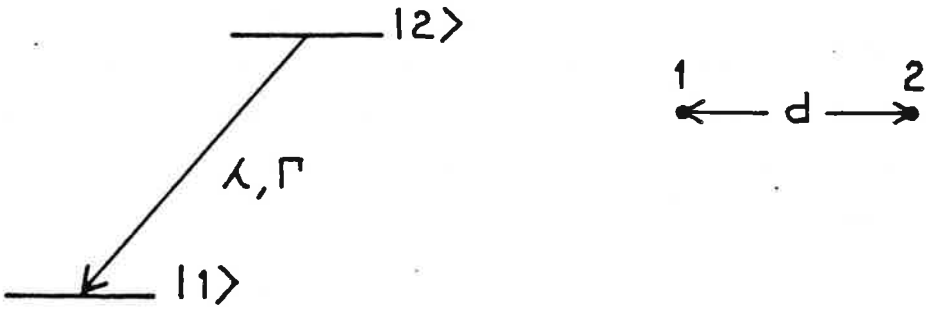

figure 3: cas de deux atomes à deux niveaux

qu'ils sont susceptibles d'émettre si on ne peut pas savoir lequel d'entre eux a émis un photon, et donc lequel se trouvait dans le niveau supérieur et lequel dans le niveau inférieur. La condition d'indiscernabilité est satisfaite quand la distance entre les deux atomes est suffisamment petite,

$$d \ll \lambda \tag{1}$$

où λ est la longueur d'onde de la transition. Dans ces conditions, ils évoluent en tant que couple d'atomes. L'état collectif est a priori représenté par une fonction d'onde à deux atomes,

$$|\psi_\epsilon >= 2^{-1/2}(|1,2> +\epsilon|2,1>) \tag{2}$$

où $|i,j>$ correspond à l'atome 1 dans l'état $|i>$ et l'atome 2 dans l'état $|j>$, et où $\epsilon = \pm 1$. Les états $|\psi_+>$ et $|\psi_->$ sont les états symétrique et antisymétrique qui correspondent à la transition $|2> \rightarrow |1>$. L'état

représentatif du système complètement désexcité est l'état symétrique $|1,1>$; comme l'évolution du système conserve la symétrie par rapport à l'échange des atomes, seul l'état $|\psi_+>$ peut évoluer vers cet état final. L'interférence entre les deux atomes est dans ce cas constructive et la probabilité d'émission par unité de temps est 2Γ, c'est à dire deux fois plus rapide qu'en émission spontanée ordinaire: il s'agit du processus élémentaire de superradiance. En revanche, l'interférence est destructive dans l'état antisymétrique et le taux d'émission est égal à zéro: les deux atomes sont bloqués dans un état qui comprend un atome dans chacun des deux niveaux, et il n'y a pas d'émission possible: l'on dit que l'état est subradiant.

1.3 Vers une mise en évidence de la subradiance

Lorsque fut prévu le phénomène de superradiance (Dicke 1954), l'éventualité de l'existence d'une interférence interatomique destructive fut aussi mentionnée, pour un système de deux particules à deux niveaux. Il fut montré que la nature, constructive ou bien destructive, de l'interférence dépend simplement des propriétés de symétrie de la fonction d'onde par rapport à l'échange des deux particules. La superradiance, c'est à dire l'idée que le rayonnement d'une collection d'atomes (ou de molécules) puisse avoir lieu par une impulsion d'intensité maximale bien plus importante que celle d'un même nombre d'atomes isolés les uns par rapport aux autres, a été, depuis sa proposition originale, le sujet d'un très grand nombre de discussions théoriques (voir à la fin du chapitre une liste des principales publications théoriques). L'autre aspect de l'émission spontanée coopérative , la subradiance, c'est à dire la diminution voire l'arrêt du rayonnement, fut en revanche un problème plus ou moins ignoré, même théoriquement. Du point de vue historique, les toutes premières observations d'effets d'émission coopérative remontent à l'expérience d'écho de spin (Hahn 1950). Il faut cependant attendre jusque 1973 la première observation de la superradiance, faite sur un gaz de HF (Skribanowitz et al 1973). La superradiance y était observée sur une transition entre deux niveaux rotationnels adjacents, dans le premier état vibrationnel excité correspondant au rayonnement infrarouge. La superradiance a ensuite été observée sur un très grand nombre de transitions et sur des longueurs d'onde qui varient entre le domaine du visible, du proche infrarouge, de l'infrarouge lointain et des micro–ondes (voir liste de références à la fin du chapitre).

Les premières études théoriques concernaient le cas des petits échantillons, c'est à dire le cas d'atomes contenus dans un volume petit par

rapport à la longueur d'onde de la transition considérée, et ignoraient l'interaction dipôle–dipôle. En fait, comme il a été montré plus tard (Friedberg et al 1972, Friedberg and Hartmann 1974a, b), cette interaction, qui donne lieu à un fort désaccord entre les différents atomes, empêche la formation d'un dipôle collectif dans les petits échantillons: on montre ainsi que, sauf pour des situations géométriques très particulières, le phénomène de superradiance est alors très sévèrement limité. Des études théoriques concernant des systèmes étendus se développèrent aussi assez rapidement (Ernst and Stehle 1968, Agarwal 1969, 1970, 1971, Arecchi and Courtens 1970, Rehler and Eberly 1971), et considérèrent le cas, expérimentalement réalisable à l'aide d'une excitation laser, par exemple, des échantillons cylindriques allongés. Bientôt, ces études prirent deux directions différentes, qui privilégient chacune l'un des deux aspects de l'émission spontanée coopérative. D'une part, on considère comme primordial l'aspect quantique, mais on se limite généralement à l'approximation du champ moyen (Bonifacio et al 1971, Banfi and Bonifacio 1974, 1975, Bonifacio and Lugiato 1975a, b). Il s'agit là d'un apport essentiel pour la compréhension du démarrage de la superradiance; cette approche ne peut toutefois pas décrire les détails de l'évolution coopérative, puis qu'elle ne considère pas la variation de l'enveloppe du champ le long de l'axe de l'échantillon. Ceci est en revanche le cas si on considére l'aspect semi–classique de l'émission spontanée coopérative (Skribanowitz et al 1973, McGillivray and Feld 1976). La validité de ce dernier traitement, excepté pour les tous premiers photons émis (Pillet 1977, Haroche 1978, Glauber and Haake 1978, Polder et al 1979, Haake et al 1979 a, b, Hopf 1979, Schuurmans et al 1981, Gross and Haroche 1982, Mostowski and Sobolewska 1983, Prasad and Glauber 1985), est maintenant bien établie. Des efforts

visant à réunir les deux points de vue (Haroche 1978, Ressayre and Tallet 1978a, b) doivent enfin être mentionnés.

Les études théoriques mentionnées ci–dessus ont exclusivement considéré le cas d'atomes à deux niveaux dans l'hypothèse d'une inversion de population initiale totale, c'est à dire quand seul le niveau supérieur de la transition est initialement peuplé. Comme on peut facilement le comprendre d'après le raisonnement simple fait dans le cas d'un système de deux atomes, cette situation initiale correspond forcément à un phénomène de superradiance, c'est à dire à une accélération et une augmentation de l'émission: l'état collectif est en effet symétrique par rapport aux permutations atomiques, et le système se désexcite complètement par une impulsion superradiante. La possibilité d'un phénomène de subradiance est ainsi exclue: seul un état antisymétrique peut éventuellement évoluer vers un état subradiant, et ceci n'est jamais le cas si seul le niveau supérieur est initialement peuplé.

La possibilité d'un phénomène de subradiance fut très tôt prise en compte dans des études concernant le cas de deux atomes (Stephen 1964, Hutchinson and Hameka 1964, Power 1967, Ernst and Stehle 1968, Arecchi and Courtens 1970, Lehmberg 1970, Steudel 1971, Varfolomeev 1971 Rayford 1974, Richter 1979, Leonardi and Vaglica 1982): on montra que la durée de vie du système est dans ce cas supérieure à celle d'un atome isolé, et tend vers l'infini lorsque la distance entre les deux atomes tend vers zéro. Un photon se trouve ainsi piégé par les deux atomes. Le phénomène de subradiance se manifeste également dans le processus de diffusion multiple cohérente qui consiste en un piégeage d'un petit nombre de photons par un grand nombre d'atomes (Barrat 1959, Ernst 1969, Ruschin and Ben Aryeh 1978). Il est en effet possible pour une collection de N atomes dont l'un est excité de piéger un photon. La

possibilité théorique d'un effet de subradiance dans le cas d'atomes à deux niveaux avait aussi été mentionnée (Freedhoff and Van Kranendonk 1967, Stroud et al 1972) mais l'idée d'un possible développement dans ce sens était aussitôt abandonnée. Une désexcitation incomplète avait également été prédite pour une collection de N atomes dans le cas des petits échantillons (Friedberg et al 1972, Friedberg and Hartmann 1974a, b); il a d'ailleur été très récemment montré qu'il s'agit là aussi d'un véritable effet de subradiance (Crubellier 1986). Mais si, d'une part, on n'étudia pas l'éventualité d'un phénomène de subradiance dans les systèmes macroscopiques, puisqu'on ne s'intéressa qu'au cas d'une inversion complète de population dans un système à deux niveaux, d'autre part aucune expérience impliquant un grand nombre d'atomes contenus dans un volume petit par rapport aux longueurs d'ondes des transitions en jeu ne fut réalisée. C'est ainsi qu'aucune expérience mettant en évidence un état subradiant n'avait finalement été proposée.

Une étude théorique de la subradiance ayant en particulier en vue la découverte de conditions expérimentales pour la mise en évidence du phénomène restait donc encore à faire: cela m'a paru mériter une analyse approfondie. C'est à la recherche théorique de systèmes macroscopiques et de configurations de niveaux où un phénomène de subradiance puisse se manifester que ce travail a été essentiellement consacré. Cette recherche a notamment abouti à la réalisation de la première mise en évidence de la subradiance.

Dans l'exemple simple de deux atomes à deux niveaux il apparaît que la symétrie de l'état collectif du système détermine le caractère constructif ou destructif de l'interférence qui exprime l'existence d'une coopération entre les atomes; le caractère de l'interférence est par aille-

urs responsable de l'évolution du système jusqu'à son état final. Malheureusement, une situation théoriquement simple comme celle de deux atomes à deux niveaux est mal adaptée aux exigences d'une expérience: mise à part la difficulté d'isoler deux atomes, il faudrait pouvoir préparer le système des deux atomes dans un état collectif qui possède une symétrie déterminée par rapport à leur permutation. Il s'est donc avéré opportun de rechercher des généralisations du cas simple de deux atomes à deux niveaux à des cas plus compliqués, mais qui soient réalisables expérimentalement. Nous avons été très naturellement amenés à étudier des situations où un grand nombre N d'atomes à plusieurs niveaux est impliqué dans le processus (Agarwal 1973a, b, 1974, Cho et al 1973, Makhviladze and Shelepin 1974, Gilmore et al 1975, Herman et al 1975, Senitzky 1975, Agarwal and Trivedi 1976, Abraham and Bullough 1980, Haake and Reibold 1982, 1984, Zverev 1983, Drummond 1984). Dans cette perspective, il a été tout à fait indispensable de disposer d'une théorie qui soit adaptée à ce cas général. Cette théorie devait en particulier pouvoir prédire l'apparition du phénomène de subradiance pour des systèmes plus complexes que le cas simple de deux atomes à deux niveaux; elle devait en outre donner les conditions initiales pour lesquelles on prévoit une évolution spontanée vers un état subradiant; il devait finalement être possible d'en extraire des cas expérimentalement réalisables.

Puisqu'un état subradiant consiste en un arrêt spontané de l'émission coopérative, il ne peut pas être observé simplement. Il s'agirait en effet d'accomplir une mesure simultanée du nombre de photons émis (ou, de façon équivalente, du nombre d'atomes désexcités ou non) et du nombre d'atomes initialement excités. Nous avons opté pour une autre façon de détecter un état subradiant, qui consiste en sa destruction; on

permet ainsi la désexcitation des atomes qui avaient précédemment été empêchés d'émettre, ce qui donne lieu à une impulsion superradiante qu'il suffit alors d'observer.

1.4 Plan du mémoire

Dans le chapitre 2 on commencera par décrire les points théoriques fondamentaux, hypothèses et concepts de base, utilisés dans ce travail. Ayant en vue une mise en évidence expérimentale, on a limité l'étude théorique au cas physiquement accessible d'échantillons cylindriques allongés, ce qui autorise l'emploi de l'approximation des ondes planes. Il est ainsi possible de dégager une bonne compréhension d'ensemble du phénomène dans les milieux macroscopiques, et d'opérer des simplifications intéressantes dans la description de l'évolution globale du système. On montrera ensuite le lien qui existe entre la condition de coopérativité des différents atomes et leur indiscernabilité vis–à–vis du champ rayonné. Cette étroite relation, qui nous suivra tout le long de ce travail, est tout à fait fondamentale dans une étude théorique des phénomènes coopératifs. On montrera enfin comment, pour un système macroscopique, la subradiance, aussi bien que la superradiance, est liée aux propriétés de symétrie de l'état collectif du système.

Dans le chapitre 3 on décrira brièvement les étapes du développement théorique qui a rendu possible la mise en évidence du phénomène de subradiance. Il s'agissait d'une part du problème posé par la création d'un état subradiant: on a pu dégager les conditions générales nécessaires à l'apparition d'une interférence interatomique destructive. D'autre part, il fallait trouver le moyen de mettre en évidence la subradiance, phénomène d'inhibition de l'émission spontanée: on donnera à la fin du chapitre le choix qu'on a opéré.

Le chapitre 4 est consacré à l'étude théorique de la subradiance due aux propriétés de symétrie de l'état collectif du système par rapport aux permutations atomiques. On étudiera en grand détail, essentiellement dans l'article intitulé "Superradiance and subradiance: I. Interatomic

interference and symmetry properties in three–level systems", la configuration simple en "V", qui permettrait en principe de rendre manifeste l'interférence destructive qui a lieu entre les atomes. Ceci nous donne les moyens d'examiner ensuite des configurations plus complexes, notamment le cas de transitions entre niveaux dégénérés, et d'en extraire celles, expérimentalement accessibles, pour lesquelles le même phénomène de subradiance est prévu, dans des conditions similaires.

L'étude des transitions entre niveaux dégénérés fait apparaître une deuxième sorte d'interférence entre les atomes: le chapitre 5, dont une grande partie est constitué par l'article intitulé "Superradiance and subradiance: II. Atomic systems with degenerate transitions", et par des extraits de l'article intitulé "Superradiance and subradiance: IV. Atomic cascades between degenerate levels", est consacré à ce type d'interférence, due à la présence de plusieurs transitions de même fréquence et même polarisation. Cette interférence aussi peut être destructive, et donner lieu à un phénomène de subradiance: on verra comment les deux types d'interférence influent sur l'état final d'un système, lorsqu'elles sont présentes simultanément.

La mise en évidence de la subradiance est à ce point envisageable: dans le chapitre 6, dont l'essentiel est constitué par l'article intitulé "Experimental evidence for subradiance", on donne le compte rendu de la première observation du phénomène, réalisée avec la configuration en cascade $4d_{3/2} \rightarrow 5p_{1/2} \rightarrow 5s_{1/2}$ du gallium. Le chapitre 7 est consacré à l'interprétation théorique des résultats obtenus; pour une grande partie le lecteur sera renvoyé, encore une fois, à l'article intitulé "Superradiance and subradiance: IV. Atomic cascades between degenerate levels", qui est présenté dans le chapitre 5. Le système étudié expérimentalement d'une cascade entre niveaux dégénérés est supposé

préparé dans un état collectif tel qu'une impulsion superradiante sur la première transition de la cascade ne vide pas complètement le niveau supérieur. On verra que l'existence de cet état subradiant entraîne la présence d'une deuxième impulsion superradiante sur la transition considérée.

Les calculs semi–classiques basés sur les équations de Bloch–Maxwell ont été faits dans le cas de l'expérience réalisée et aussi dans d'autres situations similaires. Ces calculs ont montré en particulier des effets remarquables concernant la polarisation de la lumière superradiante émise: on verra dans le chapitre 8 que ceci est tout à fait en accord avec une analyse des propriétés de symétrie.

Liste des principaux articles théoriques sur le problème de l'émission spontanée coopérative

Dicke 1954, 1964, Freedhoff and Van Kranendonk 1967, Ernst and Stehle 1968, Agarwal 1969, 1970, 1971, 1973a, b, 1974, Ernst 1969, Arecchi and Courtens 1970, De Giorgio and Ghielmetti 1971, De Giorgio 1971, Rehler and Eberly 1971, Bonifacio et al 1971, 1975a, b, Friedberg and Hartmann 1971, 1972, 1974a, b, 1976, Haake and Glauber 1972, Stroud et al 1972, Friedberg et al 1972, Cho et al 1973, Banfi and Bonifacio 1974, 1975 Herman et al 1974, Makhviladze and Shelepin 1974, Allen and Eberly 1975, Bonifacio and Lugiato 1975a, b, Herman et al 1975, Hopf and Meystre 1975, Lee 1975, 1976a, b, 1977a, b, Ressayre and Tallet 1975, 1976, 1977, 1978a, b, Senitzky 1975, Agarwal and Trivedi 1976, McGillivray and Feld 1976, Friedberg and Coffey 1976, Glauber and Haake 1976, 1978, Crubellier 1977, Ikeda 1977, Coffey and Friedberg 1978, Crubellier and Schweighofer 1978, Haroche 1978, Hopf 1979, Polder et al 1979, Haake et al 1979a, b, 1981a, b, Prakash and Chandra 1980, Abraham and Bullough 1980, Agarwal et al 1980, Crubellier et al 1980, Gross 1980, Schuurmans 1980, Mattar et al 1981, Haake and Reibold 1982, 1984, Pillet 1982, Mostowski and Sobolewska 1983, Zverev 1983, Drummond 1984, Prasad and Glauber 1985, Crubellier 1986, Crubellier et al 1986

Liste des articles de revue sur le sujet

Andreev et al 1980, Feld and McGillivray 1981, Vrehen and Gibbs 1981, Schuurmans et al 1981, Gross and Haroche 1982

Liste de références concernant les expériences de superradiance dans les différents domaines de longueurs d'onde:

visible Bréchignac et Cahuzac 1979, 1981, Cahuzac et al 1979, Florian et al 1982, 1984

proche infrarouge Flusberg et al 1976, Gross et al 1976, 1978, Gibbs et al 1977, Vrehen et al 1977a, b, Crubellier et al 1978, 1981, 1982, 1983a, b, 1984, Okada et al 1978a, b, Marek 1979, Vrehen 1979, Vrehen and Schuurmans 1979, Carlson et al 1980, Marek and Ryschka 1980, Ryschka and Marek 1981, Heinzen et al 1985

infrarouge lointain Skribanowitz et al 1973, Herman et al 1974, Gounand et al 1979, Rosenberger and DeTemple 1981

micro–ondes Gross et al 1979, Moi et al 1980, Raimond et al 1982, Goy et al 1983, Kaluzny et al 1983

2. HYPOTHESES ET CONCEPTS DE BASE

2.1 L'approximation des ondes planes

Les expériences réelles concernant des systèmes macroscopiques, qui impliquent une excitation laser initiale suivi d'une émission coopérative (figure 4), ont toutes été réalisées avec un milieu superradiant en forme de cylindre allongé. Deux méthodes ont été utilisées jusqu'ici. La première consiste à faire passer un faiseau laser qui se propage le long de l'axe Oz avec une section de faisceau a^2 dans une cellule de longueur $L \gg a$, dont l'axe de symétrie est parallèle à Oz. La seconde consiste en l'excitation d'un "multijet" atomique (voir par exemple Crubellier et al 1978) collimaté ou non et qui se propage le long d'une direction perpendiculaire à l'axe Oz par un faiseau laser de mêmes caractéristiques dirigé le long de Oz. Il est ainsi tout à fait naturel de limiter notre étude au cas des échantillons cylindriques allongés, de longueur L et section a^2, avec $L \gg a$, et composés d'un nombre N d'atomes à n niveaux non dégénérés. On appelle λ_{ij} la longueur d'onde correspondant à la transition permise qui relie les deux niveaux $|i>$ et $|j>$. L'hamiltonien général d'une collection de N atomes à n niveaux en interaction avec le champ peut être représenté par l'expression

$$H = H_A + H_F + H_I \tag{3}$$

où H_A est l'hamiltonien atomique, H_F l'hamiltonien du champ électromagnétique et H_I l'hamiltonien d'interaction dipolaire entre les atomes et le champ. Les hamiltoniens H_A et H_I peuvent être décrits par des sommes sur les N atomes,

$$H_A = \sum_{\alpha=1,N} h^\alpha \tag{4}$$

(5) $$H_I = \sum_{\alpha=1,N} (\mathbf{E}(\mathbf{z}^\alpha) \cdot \mathbf{d}^\alpha)$$

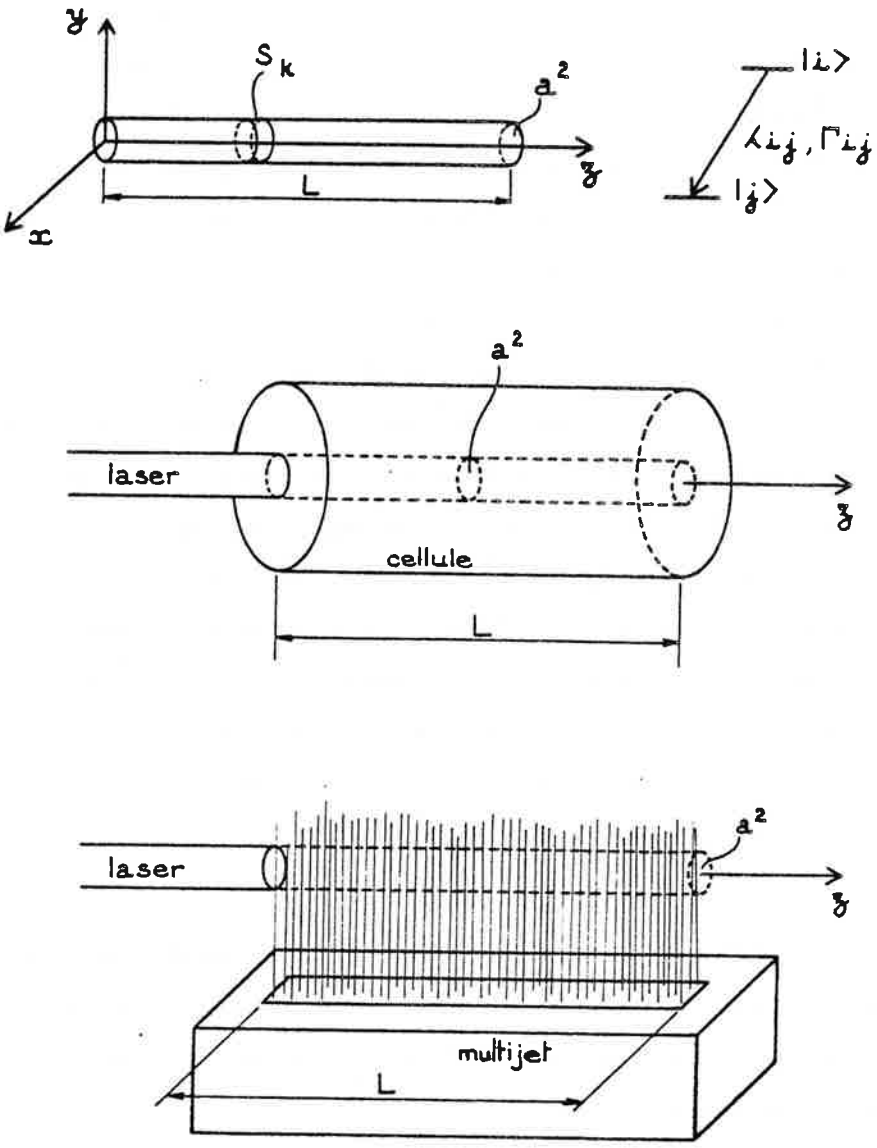

Figure 4

L'hamiltonien h^α d'un atome isolé est

$$h^\alpha = \sum_i \hbar\omega_i |i><i|_{\alpha\alpha} \tag{6}$$

où l'on somme sur les états monoatomiques $|i>$ d'énergie $\hbar\omega_i$, et l'opérateur dipolaire monoatomique est

$$\mathbf{d}^\alpha = \sum_{i,j(\omega_i>\omega_j)} (\mathbf{d}_{ij}|i><j|_{\alpha\alpha} + HC) \tag{7}$$

où l'on somme sur toutes les paires d'états monoatomiques qui sont connectées par des transitions dipolaires électriques, et où

$$\mathbf{d}_{ij} = {}_\alpha<i|\mathbf{d}^\alpha|j>_\alpha \tag{8}$$

$\mathbf{E}(\mathbf{z})$ est le champ électrique quantique et $\mathbf{z}^\alpha$ donne la position de l'atome α.

Il est bien connu dans la littérature consacrée à l'émission spontanée coopérative que le couplage entre les atomes a lieu par l'intermédiaire de modes du champ électromagnétique qui dépendent fortement de la géométrie de l'échantillon (Rehler and Eberly 1971, Herman et al 1974, Allen and Eberly 1975). Supposons que les conditions de coopérativité de l'émission spontanée (qu'on donnera dans le paragraphe suivant) soient remplies. On montre (Dicke 1964) que l'ensemble des N atomes excités, aléatoirement répartis avec une densité moyenne uniforme, émet spontanément de façon coopérative suivant les deux directions avant et arrière de l'axe Oz de symétrie du cylindre. Ces modes préférentiels de l'émission coopérative sont appelés modes coopératifs, de façon à les distinguer des autres modes du champ électromagnétique, où la désexcitation a lieu avec les caractéristiques de l'émission spontanée ordinaire. On peut alors, au moins en première approximation, considérer le champ électromagnétique comme une superposition de

deux paquets d'ondes planes qui se propagent le long de l'axe du cylindre dans les deux direction avant et arrière. Dans l'approximation des ondes planes (Rehler and Eberly 1971), le champ électromagnétique et donc l'hamiltonien H dépendent alors seulement des coordonnées z^α qui correspondent à l'axe du cylindre. On pourra d'autant mieux faire cette approximation que l'angle de diffraction λ_{ij}/a est du même ordre de grandeur que l'angle d'ouverture géométrique a/L, et cela pour toutes les transitions $|i> \rightarrow |j>$ du tfstème. Ceci est le cas quand le nombre de Fresnel $\mathcal{F}_{ij}$ associé à chaque transition $|i> \rightarrow |j>$, où $\mathcal{F}_{ij} \equiv a^2/\lambda_{ij}L$, est de l'ordre de l'unité (Gross 1980, Mattar et al 1981, Watson et al 1983), c'est à dire

$$\frac{a/L}{\lambda_{ij}/a} = \mathcal{F}_{ij} \simeq 1 \tag{9}$$

Du point de vue expérimental, il suffit en général de focaliser le faiseau laser opportunément pour que la condition (9) soit remplie.

2.2 Coopérativité de l'émission et indiscernabilité des atomes

On considère un échantillon de N atomes à n niveaux (n quelconque) dans l'approximation des ondes planes. Il s'agit alors de voir quand les N atomes dont le système est composé interfèrent les uns avec les autres. Dans le cas de deux atomes à deux niveaux, précédemment étudié, cette interférence existe dès que la distance entre les deux atomes est plus petite que la longueur d'onde de la transition (eq 1): lorsque les atomes sont suffisamment près l'un de l'autre, il sont indiscernables vis–à–vis du champ électromagnétique qu'ils rayonnent. Cette propriété d'indiscernabilité est bien sûr à l'origine de l'interférence interatomique qui caractérise l'émission coopérative. Il en sera naturellement de même pour un système macroscopique: la propriété d'indiscernabilité des atomes se trouve être équivalente à celle d'interférence interatomique. Dans l'approximation des ondes planes, cette condition est satisfaite pour les atomes qui sont au voisinage d'un même plan d'onde puisque, quand un photon s'échappe d'une telle région, on ne peut pas savoir quel atome l'a émis: les effets coopératifs se manifestent seulement quand le nombre d'atomes localement indiscernables est grand:

(10) $$\mathcal{N} \gg 1$$

où $\mathcal{N}$ est le nombre d'atomes contenus dans une tranche S_k du milieu émetteur dont l'épaisseur est de l'ordre des longueurs d'onde des transitions considérées. Avec la propriété (9), on peut écrire

$$\mathcal{N} \gg \mathcal{F}_{ij}$$

Mais puisque $\mathcal{N} = N\lambda_{ij}/L$,

$$N\lambda_{ij}/L \gg a^2/L\lambda_{ij}$$

et donc

$$N\lambda_{ij}^2/a^2 \gg 1$$

Le facteur de forme μ_{ij} relatif à chaque transition $|i> \rightarrow |j>$ pour un échantillon cylindrique allongé est donné par l' expression (Rehler and Eberly 1971, Allen and Eberly 1975)

$$\mu_{ij} = \frac{3}{8\pi}\frac{\lambda_{ij}^2}{a^2}; \tag{11}$$

on peut donc écrire, à un facteur $3/8\pi$ près

$$N\mu_{ij} \gg 1$$

et alors

$$T_{SR}(i,j) \equiv \frac{1}{N\Gamma_{ij}\mu_{ij}} \ll \frac{1}{\Gamma_{ij}} \equiv T_{SP}(i,j) \tag{12}$$

où $T_{SR}(i,j)$ est le temps caractéristique de l'émission superradiante et $T_{SP}(i,j)$ le temps caractéristique de l'émission spontanée ordinaire relatifs à la transition $|i> \rightarrow |j>$. On a ainsi rétabli la condition de coopérativité pour un milieu macroscopique qu'on obtient généralement en comparant directement les différents temps caractéristiques impliqués dans le processus (Skribanowitz 1973, Herman et al 1974, McGillivray and Feld 1976). Quand cette condition est satisfaite, l'émission spontanée du système des N atomes présente les caractéristiques d'une impulsion superradiante. L'émission est fortement anisotrope: quasiment tout le rayonnement est émis dans un angle solide petit autour de l'axe du cylindre. Un certain temps τ_D est nécessaire pour que l'intensité de l'émission atteigne son maximum, qui est proportionnel à N^2: cette sorte d'incubation est due au temps qui est nécessaire pour qu'un dipôle collectif puisse se former dans le milieu. Les impulsions fluctuent en forme et en retard: ceci a une origine quantique, en ce sens que ces

fluctuations correspondent à l'incertitude quantique de l'état initial du champ et du système atomique (Bonifacio et al 1971a,b, De Giorgio and Ghielmetti 1971, De Giorgio 1971, Haake and Glauber 1972, Agarwal 1974, Haake et al 1979a, b, Polder et al 1979, Schuurmans et al 1981, Gross and Haroche 1982).

Lorsqu'on ne peut plus négliger les effets déphasants (collisions, effet Doppler,...) qui sont susceptibles d'empêcher l'interférence interatomique, la condition (6) doit être modifiée et devient

$$T_{SR}(i,j) \ll T_2^*(i,j) \tag{13}$$

où $T_2^*(i,j)$ est le temps caractéristique des effets déphasants (McGillivray and Feld 1976); la condition de seuil pour le nombre N des atomes contenus dans l'échantillon s'écrit:

$$N \gg \frac{1}{\Gamma_{ij}\mu_{ij}T_2^*(i,j)} \equiv N_S(i,j) \tag{14}$$

Si l'on considère le volume actif comme étant formé de tranches successives S_k d'épaisseur plus petite que les longueurs d'ondes des différentes transitions, la propriété d'indiscernabilité locale des atomes vis–à–vis du champ se traduit par l'invariance de l'hamiltonien par rapport au group S_N des permutations locales des N atomes indiscernables de la tranche S_k (Kahan 1960, Messiah 1964, Racah 1965, Wybourne 1970, Judd 1973 Hermann 1974, Chisholm 1976). Ceci signifie qu'on ne modifie pas la forme de l'hamiltonien en échangeant deux indices atomiques quelconques à l'intérieur d'une tranche. Il est donc intéressant d'utiliser une base d'états collectifs ayant des propriétés de symétrie bien définies par rapport à ces permutations. Grâce à l'invariance de l'hamiltonien, les propriétés de symétrie de la fonction d'onde qui représente l'état collectif local des N atomes d'une tranche S_k vis-à-vis des permutations restent conservées.

Dans le paragraphe suivant on va voir que ces propriétés de symètrie de l'état collectif déterminent de plus la nature de l'interférence entre les atomes.

2.3 Nature de l'interférence et propriétés de symétrie

Si l'on veut connaître l'évolution coopérative d'un système, les propriétés de symétrie de son état collectif par rapport à l'échange des atomes constituent un point tout à fait essentiel. L'interférence interatomique, qui est la cause de la coopérativité de l'émission, peut avoir une nature constructive (superradiance) ou bien destructive (subradiance); cette nature est déterminée par la symétrie de l'état collectif. Compte tenu également de la propriété de conservation de cette symétrie, il est finalement naturel de prendre comme base d'états atomiques des produits d'états collectifs locaux, correspondant aux différentes tranches, qui possèdent des propriétés de symétrie bien définies par rapport au groupe S_N. Dans l'exemple simple de N atomes à deux niveaux on peut par exemple construire une base d'états collectifs avec les "états de Dicke" (Dicke 1954). On décrit chaque atome à deux niveaux de la tranche S_k comme une particule de spin 1/2 et on décrit la collection des atomes qui interfèrent à l'aide du moment angulaire total correspondant. Le composantes de ce "pseudo–moment angulaire" total sont les opérateurs collectifs de pseudo–spin de Dicke R_z, R_+ et R_-. On peut en particulier les écrire à l'aide des opérateurs P_{ij} qui sont impliqués dans l'hamiltonien d'interaction H_I (eq. 5) et qui peuvent être écrits comme sommes d'opérateurs agissant sur les différentes tranches

$$P_{ij} = \sum_k P_{ij}^{(k)}$$

avec

$$P_{ij}^{(k)} = \sum_{\alpha \in S_k} |i>_{\alpha\alpha}<j|, \tag{15}$$

où $|i>$ et $|j>$ correspondent à des états monoatomiques. On trouve:

$$R_z = (P_{22} - P_{11})/2$$

$$R_+ = P_{21} \tag{16}$$

$$R_- = P_{12},$$

où l'on a omis l'indice de la tranche. Donc

$$R^2 = [(P_{22} - P_{11})^2 + 2(P_{21}P_{12} + P_{12}P_{21})]/4 \tag{17}$$

Les états de Dicke ψ_{gmr} sont états propres de R_z et R^2 avec les valeurs propres m et $r(r+1)$, où $|m| \leq r$, $0 \leq r \leq \mathcal{N}/2$ si $\mathcal{N}$ est pair, et $1/2 \leq r \leq \mathcal{N}/2$ si $\mathcal{N}$ est impair, et où g indique qu'il existe plusieurs états de nombres quantiques r et m. Le nombre m caractérise l'état d'excitation du système, et donc son énergie $\left((\frac{\mathcal{N}}{2} + m)\right.$ atomes sont dans l'état excité$\left.\right)$. Le nombre r caractérise les états qui sont invariants par l'action de R_+ et R_-; on l'appelle aussi "nombre de coopération", puisqu'il caractérise les possibilités de coopération des atomes qui se trouvent dans l'état ψ_{gmr} (Dicke 1954, 1964). On trouve en particulier, comme on verra plus loin, que si r est différent de $\frac{\mathcal{N}}{2}$, les états sont partiellement antisymétriques. L'inconvénient des états de Dicke, très précieux dans l'étude des effets coopératifs pour des systèmes à deux niveaux, réside dans l'impossibilité qu'on a de les généraliser pour des systèmes à plusieurs niveaux. Ceci est le cas aussi pour le vecteur de Bloch (Bloch 1946) qu'on peut voir simplement comme l'analogue classique du pseudo–moment angulaire qui apparaît dans le modèle de Dicke. On rappelle la définition du vecteur de Bloch: sa projection dans la direction Oz représente l'inversion de population; sa projection dans le plan orthogonal représente la polarisation macroscopique. Lorsqu'on considère des configurations à plus de deux niveaux, on augmente le nombre de dimensions du pseudo–moment angulaire tout comme celui

du vecteur de Bloch: l'intérêt d'une représentation géométrique n'est alors plus évident.

C'est pour cette raison qu'on est amené, dans le cas des configurations à plus de deux niveaux, à construire une base d'états collectifs à l'aide de la théorie des tableaux d'Young (Racah 1965, Wybourne 1970, Judd 1973, Chisholm 1976). Les états collectifs sont alors caractérisés par des diagrammes d'Young qu'on appelle "types de symétrie" et qui correspondent à des représentations irréductibles $\{l_1, l_2, ...l_h\}$ de S_N $\left(l_1 \geq l_2 \geq ... \geq l_h \text{ , et } l_1 + l_2 + ... + l_h = N\right)$. Un type de symétrie est conservé par l'action de l'hamiltonien d'interaction atomes–rayonnement. En ce qui concerne l'exemple simple de N atomes à deux niveaux, un état collectif est alors caractérisé par un type de symétrie $\{l_1, l_2\}$. Il est en particulier état propre de P_{22} et P_{11}, avec les valeurs propres N_2 et N_1 qui sont les nombres d'atomes respectivement dans le niveau supérieur et inférieur de la tranche S_k considérée. Mais il est aussi état propre de l'opérateur de Casimir (Judd 1973)

$$W = P_{22}^2 + P_{11}^2 + P_{21}P_{12} + P_{12}P_{21} \tag{18}$$

qui est un invariant du groupe $S_N \otimes SU(2)$, avec valeur propre $[l_1(l_1+1) - l_2(l_2-1)]$. Puisque

$$R^2 = (W - N^2/2)/2, \tag{19}$$

il est aussi fonction propre de R_z et R^2 avec les valeurs propres $(N_2 - N_1)/2$ et $(l_1 - l_2)(l_1 - l_2 + 2)/4$, et donc identique à l'état de Dicke ψ_{gmr} avec $r = (l_1 - l_2)/2$ et $m = (N_2 - N_1)/2$.

Un état collectif correspondant à un tableau d'Young de type de symétrie $\{l_1, l_2\}$ donné (Crubellier et al 1980, Pillet 1982) est le produit de

—l_2 fonctions d'onde à deux atomes antisymétriques,

—une fonction d'onde symétrique qui contient $\left(l_1 - l_2\right)$ états monoatomiques.

Une telle fonction d'onde est égale à zéro quand le même état monoatomique apparaît plus d'une fois dans une fonction d'onde antisymétrique donnée; sauf si $l_2 = 0$, elle ne peut pas décrire un état où tous les atomes sont désexcités. En fait, le nombre d'atomes excités est nécessairement $\geq l_2$. Les états avec l_2 $\left(l_2 \neq 0\right)$ atomes excités sont appelés états subradiants et dans ces états l'émission est coopérativement inhibée. Comme dans le cas de deux atomes à deux niveaux, on trouve une relation fondamentale entre propriétés de symétrie de l'état collectif et subradiance. Si l'état collectif est partiellement antisymétrique par rapport à l'échange des atomes, on doit prévoir son évolution spontanée vers un état où une certaine proportion d'atomes est empêchée d'émettre: un état subradiant.

2.4 Vers les systèmes à plusieurs niveaux

Si on savait préparer un système à deux niveaux non dégénérés dans un état collectif avec $l_2 \neq 0$, on saurait créer un état subradiant. Il est en fait difficile de préparer le système dans un tel état collectif initial qui implique un peuplement simultané des deux niveaux. Toutes les études expérimentales sur les phénomènes coopératifs concernent le cas où tous les atomes sont initialement excités; dans le cas des systèmes à deux niveaux seuls les états de type de symétrie complètement symétrique $\{N\}$ sont alors accessibles, et la subradiance n'est donc pas prévue: l'évolution prend fin quand tous les atomes sont dans l'état inférieur de la transition concernée. Le système émet une impulsion superradiante et toute l'énergie atomique disponible est transformée en rayonnement. Ce type d'évolution est peut être plus immédiatement compréhensible à l'aide de la théorie du vecteur de Bloch: tant qu'il y a des atomes dans l'état supérieur de la transition, il y a nécessairement un dipôle optique différent de zéro dans le milieu, à cause de la conservation de la longueur du vecteur. Ce dipôle rayonne, et le système atomique continue de perdre son énergie. Le seul état stable est celui dans lequel tous les atomes sont désexcités: le système atomique arrête le rayonnement quand toute son énergie a été émise.

Lorsqu'on veut essayer de mettre en évidence le phénomène de subradiance, il est intéressant, pour ne pas dire pratiquement indispensable, d'entreprendre une étude des systèmes à plusieurs niveaux. Cette étude va en particulier permettre l'analyse des configurations à niveaux dégénérés, où l'on peut alors dégager les possibilités de préparation expérimentale d'un système physique qui évolue spontanément vers un état subradiant. Dans les chapitres suivants, où l'on considère ces systèmes plus compliqués, on verra plus manifestement l'intérêt de

l'utilisation de la théorie des tableaux d'Young. En fait cette théorie est pratiquement la seule capable de prédire l'état final d'une configuration impliquant plusieurs niveaux, et d'en expliquer clairement les raisons physiques.

Dans le chapitre 3 on donne les étapes de notre développement théorique. Le premier pas consiste en la recherche des conditions initiales nécessaires à l'apparition d'un phénomène de subradiance pour un système macroscopique d'atomes à trois niveaux. On considère ensuite les processus impliquant deux niveaux dégénérés: on verra que deux sortes d'interférence interatomique sont impliquées, et peuvent être responsables d'un phénomène de subradiance. Finalement, il faut rendre manifeste la subradiance. C'est vers ce but qu'est orientée la fin du chapitre, dans laquelle on donne le choix qu'on a fait pour mettre en évidence le phénomène.

3. ETAPES DU DEVELOPPEMENT THEORIQUE

3.1 N atomes à trois niveaux dans la configuration en "V"

Comme on a vu dans le cas d'atomes à deux niveaux, un phénomène de subradiance est possible lorsque l'état collectif est partiellement antisymétrique par échange des atomes. Pour la configuration de N atomes à trois niveaux dans la configuration "V", le même phénomène est prévu et apparaît dans les mêmes conditions: la subradiance se manifeste quand l'état collectif est partiellement antisymétrique par rapport aux permutations atomiques. Nous avons pu établir les conditions initiales nécessaires pour que le système évolue effectivement vers un état subradiant grâce à une relation quelque peu inattendue que nous avons su trouver entre les propriétés de symétrie et la matrice densité. Les proprietés de symétrie de l'état initial, qui établissent complètement la nature constructive ou destructive de l'interférence entre les divers atomes, ne sont en effet pas accessibles directement dans une situation réaliste. Par ailleurs, l'état initial d'atomes non correlés, qui est ce qu'on prépare habituellement, est décrit par une matrice densité factorisable. L'existence d'une relation entre deux caractéristiques de l'état collectif apparemment étrangères l'une à l'autre que sont la symétrie et le mélange statistique, nous a permis de montrer, comme on verra dans le chapitre 4, que l'émission coopérative est spontanément inhibée, à la suite d'une impulsion superradiante, lorsque, tout simplement, le système a été initialement préparé dans un état collectif qui est un mélange statistique des deux niveaux supérieurs de la configuration considérée. On montre en effet, à l'aide de la théorie des tableaux d'Young, que la proportion de mélange statistique de l'état atomique fixe les propriétés de symétrie de cet état, établissant ainsi une relation entre le processus d'excitation qui est non coopératif d'une part et le

processus de désexcitation coopérative de l'autre.

Néanmoins, on s'aperçoit qu'une telle configuration est difficilement utilisable physiquement. Il faudrait en effet disposer d'une excitation simultanée des deux niveaux supérieurs, qui est en général assez délicate à préparer. On a donc décidé d'étudier des configurations plus compliquées et, en particulier, le cas des transitions $j \rightarrow j'$ entre niveaux dégénérés. Toutefois, pour ces transitions, un deuxième type d'interférence est alors impliqué dans l'évolution globale du système: à cause de la dégénéréscence des niveaux, plusieurs transitions de même fréquence et même polarisation sont en effet simultanément concernées.

3.2 Transitions dégénérées: un deuxième type d'interférence

Lorsqu'on considère un système atomique avec une transition dégénérée, une deuxième sorte d'interférence, cette fois entre différentes transitions de même fréquence et même polarisation, apparaît entre les atomes. De la même façon que pour l'interférence interatomique sur chaque transition séparément, et on le verra en détail dans le chapitre 5, l'indiscernabilité des atomes par rapport au champ qu'ils rayonnent est aussi responsable de ce deuxième type d'interférence entre les atomes. Cette interférence, encore une fois, peut être constructive ou bien destructive, et donne lieu soit à une augmentation de l'émission coopérative, soit à une inhibition de cette émission, et donc finalement, dans ce deuxième cas, à un état subradiant. Les deux interférences, causées toutes les deux par l'indiscernabilité des atomes, et donc intrinsèques à l'émission coopérative, ont lieu, bien évidemment, dans les mêmes conditions et avec les mêmes éventuelles limitations.

Cependant, puisque l'interférence implique cette fois des transitions différentes, sa nature constructive ou destructive ne dépend plus des propriétés de symétrie vis–à–vis des permutations atomiques. Pour le cas simple d'un système à quatre niveaux avec une transition dégénérée, on trouve en particulier que le taux de subradiance dépend de façon compliquée des populations initiales et des probabilités de transition.

On montre brièvement dans le paragraphe suivant que les deux types d'interférence interatomique, sur chaque transition séparément et entre les différentes transitions de même fréquence et même polarisation existent simultanément dans le cas d'une transition dipolaire entre deux niveaux dégénérés.

3.3 Transition entre niveaux dégénérés

Dans l'intention de se rapprocher d'une éventuelle expérience de subradiance, on s'intéresse au cas physiquement accessible de transitions dipolaires entre niveaux dégénérés. Un phénomène de subradiance dû aux propriétés de symétrie de l'état collectif du système est en effet prévu pour une transition entre niveaux dégénérés de moments angulaires j et $j-1$: on verra en détail dans le chapitre 4 que, pour cette configuration, le système évolue vers un état subradiant quand il est préparé dans le niveau excité j avec une proportion suffisamment importante de mélange statistique. Encore une fois, donc, apparaît la relation, tout à fait essentielle dans la perspective d'une mise en évidence expérimentale, qui relie mélange statistique initial et propriétés de symétrie de l'état collectif. Grâce à ce lien, on est capable de définir les conditions initiales favorables à la manifestation d'un phénomène de subradiance,

Par ailleurs, le deuxième type d'interférence interatomique, cette fois entre différentes transitions ayant même fréquence et même polarisations, va aussi jouer un rôle dans l'évolution d'un système à niveaux dégénérés. On verra dans le chapitre 5 que bien souvent les deux sortes d'interférences, quand elles coexistent, somment simplement leurs contributions à la subradiance.

On peut très facilement imaginer de préparer expérimentalement un système macroscopique dans un mélange statistique des sous-niveaux du niveau supérieur j d'une transition $j \to j-1$, par exemple avec une excitation laser linéairement polarisée qui a lieu à partir d'un troisième niveau de moment angulaire égal ou supérieur à j. On peut donc obtenir un système qui évolue spontanément vers un état où l'émission est coopérativement inhibée. Le dernier pas vers la réalisation d'une

expérience de subradiance est alors la mise en évidence de cet état collectif du système, où une certaine proportion d'atomes est restée dans l'état excité de la transition considérée.

3.4 Cascade entre niveaux dégénérés

Le problème qui se pose, une fois qu'on sait créer un état qui évolue vers un état subradiant, est celui de la mise en évidence d'un phénomène qui consiste en une inhibition de l'émission. On peut imaginer de détruire l'état subradiant préalablement créé, et de permettre ainsi de nouveau l'émission sur la transition considérée. On peut parvenir à cela, par exemple, à l'aide d'une émission superradiante qui démarre du niveau inférieur de la transition vers d'autres niveaux d'énergie encore plus basse. Si, donc, outre la transition entre les niveaux dégénérés j et $j-1$, la configuration atomique comporte une transition partant du niveau $j-1$ vers un troisième niveau de moment angulaire j', le système, supposé initialement dans un mélange statistique d'états du niveau supérieur j, va schématiquement évoluer comme suit. Après une première impulsion superradiante sur la transition $j \rightarrow j-1$, un état subradiant est créé par rapport à cette transition. Ceci n'empêche pas l'émission d'une impulsion superradiante sur la transition $j-1 \rightarrow j'$, qui va vider le niveau $j-1$ intermédiaire. Alors, la fonction d'onde représentative de l'état collectif du système comprendra seulement les deux niveaux supérieur et inférieur, qui ne sont pas reliés par une transition atomique permise. L'état, bien que partiellement antisymétrique, n'est donc pas subradiant. Une deuxième impulsion superradiante sur la première transition va donc pouvoir avoir lieu, vidant ainsi le niveau supérieur de la cascade. Cette deuxième impulsion, qu'on appelle "écho de subradiance", implique directement l'existence d'un état subradiant dans lequel une certaine proportion d'atomes était empêchée de se désexciter.

4. SUBRADIANCE PAR ANTISYMETRIE

4.1 Configuration à trois niveaux

On étudie le cas d'un système de N atomes à trois niveaux dans la configuration "V", c'est à dire une configuration composée de deux niveaux supérieurs $|2>$ et $|3>$ reliés, par l'intermédiaire de deux transitions dipolaires électriques, à un troisième niveau inférieur $|1>$ (figure 5). On suppose que la condition (eq. 12) de coopérativité de l'émission

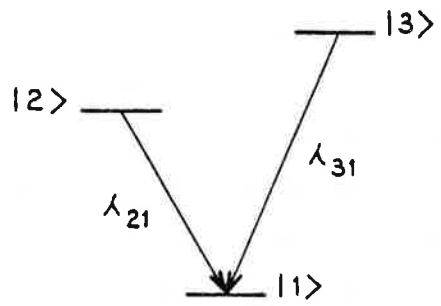

figure 5: configuration à 3 niveaux en "V"

spontanée du système est satisfaite. On considère ici trois cas particuliers différents correspondant à trois états collectifs initiaux.

4.1.1 Evolution dans trois cas simples:

influence du mélange statistique

a) Dans le premier cas, on suppose que tous les atomes sont portés initialement dans un seul des deux niveaux supérieurs. En raison de l'interférence qui existe entre les atomes et qui, dans ce cas, est constructive, ils se désexciteront par une impulsion superradiante qui vide complètement les niveaux supérieurs, suivant la même évolution que

celle d'un système à deux niveaux: le niveau qui n'avait pas été peuplé initialement agit comme simple "spectateur" dans l'évolution globale du système, et l'on peut considérer ce problème comme équivalent au problème à deux niveaux précédemment étudié.

b) Le deuxième cas concerne une excitation qui peuple également et simultanément les deux niveaux supérieurs de façon à obtenir une cohérence initiale complète entre eux: il s'agit donc d'un cas pur. Un raisonnement simple nous permet de prévoir l'état final du système, en fonction de la cohérence existant entre les deux niveaux initialement peuplés. Dans le modèle semi–classique, le mélange statistique est conservé. Ceci équivaut à dire que σ^2, la trace du carré de la matrice densité monoatomique, est conservée. Puisque σ^2 est initialement égale à un, le système reste dans un état pur tout le long de l'évolution; l'évolution s'arrête quand tous les atomes sont dans le niveau inférieur de la configuration "V", c'est à dire quand tous les atomes sont désexcités.

c) Considérons, en revanche, le cas où la cohérence entre les deux niveaux supérieurs est initialement nulle. Ce cas correspond à un mélange statistique complet à poids égaux de ces deux niveaux. Le système va alors rester dans un mélange statistique d'états durant toute l'évolution. En particulier, l'état final sera un mélange statistique d'états, et ne pourra pas être l'état complètement désexcité qui correspond à un cas pur. L'état final du système dans le cas où la cohérence initiale est nulle est tel que la moitié des atomes reste dans les deux niveaux supérieurs du système en configuration "V"; l'émission coopérative est alors inhibée.

4.1.2 Interprétation physique des trois cas: lien entre mélange statistique et symétrie

A la différence du cas à deux niveaux, une interférence interatomique

destructive, qui donne lieu à un état subradiant, peut être ainsi prévue pour cette configuration "V" même quand tous les atomes sont initialement excités. Le raisonnement simple qui nous a permis de prévoir un effet de subradiance n'est cependant pas capable de nous fournir une compréhension physique du phénomène. Ceci sera en revanche possible, comme c'était déjà le cas pour les situations plus simples, en considérant les propriétés de symétrie de l'état collectif des atomes.

Regardons d'abord brièvement le cas de deux atomes en configuration "V". Initialement les deux atomes sont supposés être dans les deux niveaux supérieurs du système, et l'état collectif est représenté par une fonction d'onde construite avec les deux états supérieurs, qui peut être, comme dans le cas à deux niveaux (eq. 2) symétrique ou antisymétrique par échange des atomes. Après l'émission d'un photon, un atome est resté dans un des deux niveaux supérieurs et l'autre se trouve dans le niveau inférieur sans que la symétrie ait changé. Dans le cas des états antisymétriques, en particulier, l'interférence entre les deux atomes est alors destructive et l'émission est inhibée: il s'agit du processus élémentaire de subradiance.

Forts de ce résultat, on étudie alors l'évolution d'un système macroscopique à N atomes dans la configuration "V". Le nœud du traitement théorique est de connaître le type de symétrie du système dans son état initial. Si l'état initial du système est défini seulement de façon statistique, c'est à dire par une matrice densité, on pourra connaître les propriétés du système uniquement de manière probabiliste. On peut calculer (voir paragraphe 4.1.3) la probabilité pour une matrice densité factorisable et spatialement homogène que l'état collectif de la tranche S_k ait un type de symétrie $\{l_1, l_2\}$. On montre que le type de symètrie est en fait bien déterminé, et que l'on a $l_1 = N\rho_{aa}$ et

$I_2 = \mathcal{N}\rho_{bb}$, où ρ_{aa} et ρ_{bb} sont les valeurs propres de la matrice densité monoatomique $\left(\text{avec } \rho_{aa} \geq \rho_{bb}\right)$. Par conséquent, un état pur $\left(\sigma^2 = 1\right)$ correspond au type de symétrie $\{\mathcal{N}\}$ complètement symétrique. En revanche, un mélange statistique initial à poids égaux des deux niveaux supérieurs $\left(\sigma^2 = 1/2\right)$ possède un type de symétrie $\{\frac{\mathcal{N}}{2}, \frac{\mathcal{N}}{2}\}$, qui correspond à des états partiellement antisymétriques par rapport aux permutations des $\mathcal{N}$ atomes de la tranche. Les cas a) et b) correspondent au type de symétrie complètement symétrique: leur état final est l'état complètement désexcité, et il sera atteint après émission d'une impulsion superradiante qui vide les niveaux supérieurs. En revanche, le cas c) a le type de symétrie "le plus antisymétrique" $\{\frac{\mathcal{N}}{2}, \frac{\mathcal{N}}{2}\}$. Pour ces conditions initiales, on peut visualiser l'état collectif comme un ensemble de paires d'atomes dans un état antisymétrique. On peut alors associer à chaque "paire antisymétrique" une fonction antisymétrique à deux atomes $|i,j>_{AS}$ construite sur deux états monoatomiques i et j. Initialement les deux états i et j sont les deux états excités. L'évolution de chaque paire prend fin quand un état supérieur est remplacé, dans la fonction associée à la paire atomique, par l'état inférieur. Cet effet de subradiance est, bien sûr, dû à l'antisymétrie de la fonction d'onde par rapport à l'échange des atomes, mais il est aussi étroitement lié au choix qu'on a fait pour la configuration des niveaux: c'est l'unicité du niveau inférieur qui rend possible l'inhibition de l'émission spontanée. Il apparaît donc clairement que le phénomène de subradiance nécessite, pour pouvoir se manifester, des configurations atomiques particulières.

4.1.3 Interférence interatomique et propriétés de symétrie dans les systèmes à trois niveaux

L'article qui est présenté ci–après ("Superradiance and subradiance: I. Interatomic interference and symmetry properties in three–level sys-

tems") est essentiellement orienté vers l'étude de l'interférence interatomique destructive responsable de la subradiance. On y étudie en grand détail l'exemple typique d'un échantillon cylindrique allongé d'atomes dans la configuration "V", pour lequel on montre que l'on peut atteindre spontanément un état subradiant. Cela se produit pour des atomes initialement non corrélés quand l'état initial est un mélange statistique des deux niveaux supérieurs de la configuration. La clef de voûte est donc donnée par les propriétés de symétrie de l'état collectif par rapport aux permutations atomiques. Le résultat fondamental de l'article est la démonstration du lien existant entre les propriétés de symétrie, non accessibles directement, et la matrice densité initiale, expérimentalement disponible. Pour la description de l'évolution du système on a souvent utilisé une approche semi–classique: on peut par exemple se convaincre rapidement de l'existence de la subradiance à travers des considérations simples impliquant la conservation du mélange statistique (§ 3); par ailleurs, on a utilisé les équations de Bloch–Maxwell pour la description de l'évolution du système (§ 7.1). Une approche quantique a été en revanche utilisée quand il s'agit de la compréhension physique du phénomène: le cas simple de deux atomes est, par exemple, étudié avec un traitement markovien (§ 2). On a trouvé, à l'aide de la théorie des tableaux d'Young, la valeur minimum du taux de subradiance (§ 8), valable pour une matrice densité factorisable et homogène. Ceci montre que, à moins que l'état initial ne soit un état pur, il faut toujours, pour cette configuration, s'attendre à un phénomène de subradiance. En examinant les résultats trouvés, on pourrait imaginer que les atomes qui interfèrent destructivement entre eux vont rester indéfiniment dans un état subradiant. On montre à la fin de l'article (§ 9) que toute restriction à l'indiscernabilité lo-

cale des atomes perturbe le phénomène de subradiance. L'emploi de l'approximation des ondes planes est autorisé parce que on peut prouver que l'émission dans les modes transversaux est négligeable pendant l'émission superradiante. Mais, ensuite, seule l'émission dans les modes longitudinaux est inhibée: l'émission spontanée ordinaire dans les autres modes peut avoir lieu même dans un état subradiant, et les atomes peuvent ainsi se désexciter complètement. Un état subradiant ne peut donc exister que pendant une période de temps inférieure à ou de l'ordre de T_{SP}.

J. Phys. B: At. Mol. Phys. 18 (1985) 3811-3833. Printed in Great Britain

Superradiance and subradiance: I. Interatomic interference and symmetry properties in three-level systems

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Received 18 March 1985

Abstract. This paper is the first of a sequence of four papers devoted to the theoretical study of subradiance, i.e. of the cooperative inhibition of spontaneous emission, a phenomenon which has been recently observed. It is shown, in the typical example of a pencil-shaped sample of three-level atoms in the 'V' configuration (two transitions sharing a common lower level), that the system can spontaneously reach a state in which the emission in the so-called 'end-fire' superradiant field modes is cooperatively inhibited; this occurs for initially uncorrelated atoms when the initial state is a statistical mixing of the two upper states. The whole study is centred on the concept of destructive interatomic interference and it is therefore based on the symmetry properties of the collective atomic state with respect to the permutations of locally indistinguishable atoms. Semiclassical and fully quantum-mechanical models are used in turn. The obstacles to the interatomic interference and thus to subradiance are finally discussed.

1. Introduction

When predicting superradiance Dicke (1954) had already mentioned the possibility of a pair of two-level systems (two neutrons in a uniform magnetic field) trapping one photon. In the limit of fully cooperative emission, a pair of two-level systems is indeed unable to radiate when it is in the antisymmetrical state, the so-called subradiant state, which still contains one excited system. This quenching of the emission is due to destructive interference between the two systems. As a simple generalisation of Dicke's results, one can thus expect that the cooperative effects either accelerate and enhance or slow down and inhibit the spontaneous emission of a collection of atoms, according to either the constructive or destructive nature of the interatomic interference. The first case corresponds to the well known superradiant emission, observed for the first time in 1973 (Skribanowitz *et al* 1973). The inhibition of spontaneous emission by destructive interference, while the atoms are not all de-excited, is called subradiance. The first experimental evidence for this phenomenon has been obtained only very recently (Crubellier *et al* 1985, Pavolini *et al* 1985). The existence of many-atom subradiant states could have been accounted for by most of the theoretical treatments of two-level atom superradiance (or superfluorescence) (see the review papers on the subject: Feld and McGillivray 1981, Vrehen and Gibbs 1981, Schuurmans *et al* 1981, Gross and Haroche 1982 and references therein). The phenomenon has, however, been mentioned in very few papers (Freedhof and Van Kranendonk 1967, Stroud *et al* 1972). In fact, these subradiant states, which can be simply described as containing

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0022-3700/85/183811+23$02.25

pairs of atoms in antisymmetrical states, are neither easy to create nor to observe. This is probably the reason why the subradiance phenomenon, although it should seem to be fundamentally as important as its counterpart, the superradiance, has been more or less ignored, theoretically as well as experimentally, for quite a long time. With the exception of the two-atom case (Stephen 1964, Hutchinson and Hameka 1964, Ernst and Stehle 1968, Lehmberg 1970, Varfolomeev 1971) and the coherent multiple scattering (Barrat 1959, Ernst 1969, Ruschin and Ben Arych 1978) (in which many atoms trap very few photons), the earliest case of subradiance which has been studied in the literature concerns the case of small samples. As a matter of fact the 'limited superradiance' which is predicted in this case (Friedberg *et al* 1972, Friedberg and Hartmann 1974, Coffey and Friedberg 1978) is due to destructive interatomic interference: this will be shown in detail in a following paper (III). Being interested in an experimental study of subradiance, we have first dealt with the realistic case of extended pencil-shaped samples. Recent studies of level degeneracy effects (Crubellier 1977, Crubellier and Schweighofer 1978) and of polarisation properties (Crubellier *et al* 1978, 1979, 1981, 1984) have led us to consider the case of many-level atoms, which opens new possibilities for the observation of subradiance. In a previous paper (Crubellier *et al* 1980), we have analysed in this context the typical case of three-level atoms in the 'V' configuration (two transitions sharing a common lower level), and we have now developed a general theory (Pillet 1982, Crubellier *et al* 1983b) which has allowed us to interpret our experimental results (Crubellier *et al* 1985, Pavolini *et al* 1985). 'Limited superradiance' has also been studied, for extended samples, in several particular cases of many-level atoms and has already been interpreted as due to destructive interference (Gross and Haroche 1982); as will be shown in a following paper (II), this effect could in any case be called subradiance too. In the present sequence of four papers (I–IV), we develop a general theoretical study of subradiance. The concept of destructive interatomic interference provides the physical understanding of the phenomenon, as well as the basis of our mathematical analysis of the problem.

In the present paper, the first of the sequence, we study in much greater detail than in our previous paper (Crubellier *et al* 1980) the case of three-level atoms in the 'V' configuration. For this simple atomic configuration, a group theoretical approach shows that fully excited partially antisymmetrical states spontaneously evolve, through cooperative emission, to subradiant states. In a realistic situation, however, the symmetry properties of the initial state are not always directly available. As one usually prepares uncorrelated atoms, their initial state is indeed described by a factorisable density matrix. In order to elaborate an experiment for the observation of subradiance, a link between the permutation symmetry properties and the density matrix point of view is thus required. This link is in fact the main result of the present theoretical study. In this study, both semiclassical and quantum-mechanical approaches are used in turn. A Markovian treatment, for example, is used in a brief introductory study of the two-atom case (§ 2), whereas one can be quickly convinced of the existence of subradiance by using the statistical mixing conservation as it applies in the semiclassical model (§ 3). However central to the analysis is the notion of symmetry properties with respect to the atomic permutations, which allows one to understand the physical origin of subradiance and which can be studied regardless of the model choice. These symmetry properties are shown to be conserved in the plane-wave approximation (§ 4) and collective states with well defined symmetry properties are described (§ 5): the number of atoms which cannot de-excite appears then to depend on the symmetry properties of the initial state only (§ 6). The evolution of the system is most easily

calculated in the semiclassical model, whereas the physical meaning of the results of a Markovian quantum-mechanical treatment appears more clearly (§ 7). It is then demonstrated using the Young tableaux theory that, in the semiclassical model, the amount of statistical mixing of the atomic state determines in fact the symmetry properties of this state, so that both approaches are unified; this provides the above mentioned link between the non-cooperative excitation process and the cooperative de-excitation (§ 8). Finally the obstacles to subradiance are discussed (§ 9).

2. Two-atom case

The 'V' level configuration is represented in figure 1. All levels are non-degenerate. The two upper levels, $|1\rangle$ and $|2\rangle$, are assumed to be connected by electric dipole transitions to the lower level, $|0\rangle$. The two transitions are assumed to be resonant with different modes of the electromagnetic field. The matrix elements of the atomic dipole operator, $\boldsymbol{d}$, are

$$\begin{aligned} \boldsymbol{d}_{10} &= \langle 1|\boldsymbol{d}|0\rangle \\ \boldsymbol{d}_{20} &= \langle 2|\boldsymbol{d}|0\rangle \end{aligned} \tag{2.1}$$

and the transition probabilities per unit time are Γ_1 and Γ_2.

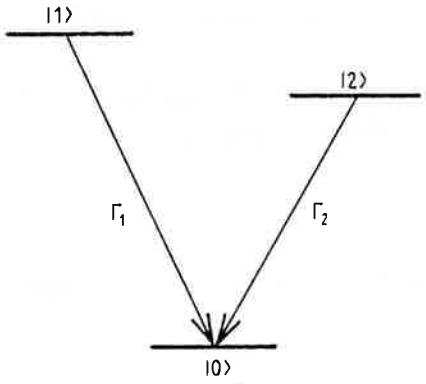


Figure 1. Three-level 'V' configuration.

We consider now two such three-level atoms. The interatomic distance is assumed to be much smaller than the wavelengths of the two atomic transitions and therefore the emission is cooperative (Stephen 1964). The initial two-atom state is assumed to be of the form

$$|\psi_\varepsilon\rangle = 2^{-1/2}(|1, 2\rangle + \varepsilon|2, 1\rangle) \tag{2.2}$$

where $|i, j\rangle = |i\rangle_1|j\rangle_2$ corresponds to atom 1 in state $|i\rangle$ and atom 2 in state $|j\rangle$ and where ε is either 1 or -1. After the emission of a first photon, the system reaches either the state

$$|\psi_{1\varepsilon}\rangle = 2^{-1/2}(|0, 2\rangle + \varepsilon|2, 0\rangle) \tag{2.3}$$

if the photon has been emitted on transition $1 \to 0$, or the state

$$|\psi_{2\varepsilon}\rangle = 2^{-1/2}(|1, 0\rangle + \varepsilon|0, 1\rangle) \tag{2.4}$$

if the photon has been emitted on $2 \rightarrow 0$. The states $|\psi_{1+}\rangle$ and $|\psi_{1-}\rangle$ (respectively $|\psi_{2+}\rangle$ and $|\psi_{2-}\rangle$) are the symmetrical and antisymmetrical states corresponding to the $1 \rightarrow 0$ (respectively $2 \rightarrow 0$) transition. In the symmetrical states ($\varepsilon = 1$), the interatomic interference is constructive and the emission rate, $2\Gamma_1$ (respectively $2\Gamma_2$), is twice as large as in non-cooperative emission: this is the elementary process for superradiance. In the antisymmetrical states ($\varepsilon = -1$), on the contrary, the interference is fully destructive and the emission rate is zero; one photon is therefore trapped by the atoms and these states are subradiant. In addition, one can notice that the symmetry properties of the wavefunction with respect to the exchange of the atoms are conserved and that the de-excitation of the system in its final state is completely determined by the symmetry of the initial state.

In the following sections, we shall study cooperative spontaneous emission of N three-level atoms. We first demonstrate the existence of subradiance, by using the semiclassical model. As in the two-atom case, the constructive or destructive nature of the interatomic interference will finally be shown to be fixed by conservative symmetry properties of the multi-atom wavefunction with respect to atomic permutations.

3. Statistical mixing conservation and subradiance

Existence of subradiance for three-level atoms in the 'V' configuration (figure 1) can be predicted using simple considerations based on the conservation, in the semiclassical model, of statistical mixing, i.e. on the conservation of the squared density matrix trace (see also Crubellier *et al* 1980).

Let us assume that the N three-level atoms are initially uncorrelated, so that the initial atomic state is represented by a factorisable density matrix

$$\rho(0) = \prod_{\alpha=1,N} \rho^{\alpha}(0). \tag{3.1}$$

If all atoms are initially excited, either in state $|1\rangle$ or in state $|2\rangle$, one has

$$\rho^{\alpha}(0) = \sum_{i,j} \rho_{ij}^{\alpha}(0) |i\rangle_{\alpha\alpha}\langle j| \tag{3.2}$$

the sums on i and j running over 1 and 2. In the semiclassical model, the evolution of the atomic system is ruled by a Hamiltonian which, since there is no direct interaction between the atoms, is a sum of monatomic Hamiltonians. The density matrix remains therefore factorisable and equation (3.1) holds at any time. In addition the statistical mixing does not change during the evolution and thus

$$\mathrm{Tr}[(\rho^{\alpha}(t))^2] = \sum_{i,j} \rho_{ij}^{\alpha}(t)\rho_{ji}^{\alpha}(t) = (\sigma^{\alpha})^2 \tag{3.3}$$

with constant $(\sigma^{\alpha})^2$. At the beginning of the evolution one has $\frac{1}{2} \leqslant (\sigma^{\alpha})^2 \leqslant 1$; the case $(\sigma^{\alpha})^2 = 1$ corresponds to pure states and the case $(\sigma^{\alpha})^2 = \frac{1}{2}$ to full statistical mixing with equal populations. If, at the end of the evolution, the atoms were completely de-excited, i.e. in state $|0\rangle$, one would necessarily have $(\sigma^{\alpha})^2 = 1$. Therefore the squared density matrix trace conservation implies that the de-excitation of the atoms cannot be complete except if the initial atomic state is a pure state. Otherwise the atoms cannot be all de-excited at the end of the evolution and the final state of the system is expected to be subradiant. We shall see in the following that this effect is basically

due, as in the two-atom case, to destructive interatomic interference and that the study of the symmetry properties of the collective atomic states will provide a good understanding of the phenomenon.

4. Invariance properties of the atom–field Hamiltonian in the plane-wave approximation

4.1. Atom–field Hamiltonian

We consider the general Hamiltonian of a collection of N atoms with n levels interacting via electric dipole interaction with the quantised electromagnetic field,

$$H = H_A + H_F + H_I \tag{4.1}$$

where H_A is the atomic Hamiltonian, H_F is the Hamiltonian of the electromagnetic field and H_I is the interaction Hamiltonian. The Hamiltonians H_A and H_I can be written as sums over the N atoms,

$$H_A = \sum_{\alpha=1,N} h^\alpha \tag{4.2}$$

$$H_I = \sum_{\alpha=1,N} (\boldsymbol{E}(z^\alpha) \cdot \boldsymbol{d}^\alpha) \tag{4.3}$$

The Hamiltonian h^α of one isolated atom is

$$h^\alpha = \sum_i \hbar\omega_i |i\rangle_{\alpha\alpha}\langle i| \tag{4.4}$$

where the sum runs over the monatomic states $|i\rangle$ of energy $\hbar\omega_i$, and the monatomic dipole operator $\boldsymbol{d}^\alpha$ can be written as

$$\boldsymbol{d}^\alpha = \sum_{i,j(\omega_i > \omega_j)} (\boldsymbol{d}_{ij} |i\rangle_{\alpha\alpha}\langle j| + \text{HC}) \tag{4.5}$$

where the sum runs over all pairs of monatomic states which are connected by electric dipole transitions and where

$$\boldsymbol{d}_{ij} = {}_\alpha\langle i|\boldsymbol{d}^\alpha|j\rangle_\alpha. \tag{4.6}$$

$\boldsymbol{E}(z)$ is the quantised electric field and z^α gives the position of atom α.

4.2. Plane wave approximation

We assume here that the atoms which are affected by cooperative spontaneous emission are homogeneously and randomly distributed inside a pencil-shaped volume of section a^2 and length L. In this case the emitted field is approximately the superposition of two plane-wave packets travelling forward and backward along the cylinder axis (Dicke 1964, Herman *et al* 1974). It is recalled that the plane-wave approximation best applies if the Fresnel numbers associated with the emitted field, $F_{ij} = a^2/L\lambda_{ij}$, are of the order of one (Rehler and Eberly 1971). For a Fresnel number smaller than one, the diffraction angle λ_{ij}/a is larger than the geometrical angle a/L and diffraction effects are important (Le Berre-Rousseau *et al* 1979). For a Fresnel number larger than one, the field is emitted independently inside several diffraction lobes contained in the geometrical angle (Mattar *et al* 1981, Drummond and Eberly 1982) and the plane wave

approximation is also difficult to justify. For Fresnel numbers of the order of one, semi-classical calculations taking into account transverse effects (Mattar *et al* 1981, Watson *et al* 1983) are in rather good agreement with the results obtained using the plane-wave approximation.

4.3. Indistinguishability of the atoms

In the plane-wave approximation, the electromagnetic field depends only on the z coordinate which corresponds to the cylinder axis. Therefore all atoms α contained in a given slice S_k defined by $z_k \leqslant z^\alpha < z_k + \Delta z$, with Δz of the order of the smallest λ_{ij}, are indistinguishable for the field they can radiate. It is impossible to know which atom of the slice has emitted a photon escaping from this slice. This local indistinguishability of the atoms is at the origin of the interatomic interference which characterises cooperative emission. As previously shown (Crubellier *et al* 1980), the necessary condition for cooperative spontaneous emission, which can be written as $T_{SR} \ll T_{sp}$, where T_{sp} and T_{SR} are respectively the characteristic times of non-cooperative emission and of superradiant emission, implies the number of (locally) indistinguishable atoms to be much larger than one.

If one considers the whole active volume as formed by successive slices S_k of thickness Δz, the atom–field Hamiltonian is invariant under any permutation of atoms inside a given slice S_k, i.e. under the corresponding permutation groups S_N where N is the number of atoms contained in a slice,

$$N = N_t \Delta z / L$$

N_t being the number of atoms in the whole volume. Therefore if one uses, as atomic basis states, products of collective states corresponding to the successive slices, the symmetry properties of these latter states with respect to the S_N groups will be conserved during the evolution. More precisely the states of any slice can be labelled according to the irreducible representations of S_N. These representations will be called in the following 'symmetry types' and they are characterised by partitions of N, written $\{l_1 l_2, \ldots, l_h\}$, with $l_1 + l_2 + \ldots + l_h = N$ and $l_1 \geqslant l_2 \geqslant \ldots \geqslant l_h$. The local indistinguishability of the atoms implies the conservation of the irreducible representations of the groups S_N, i.e. of the symmetry types of the local atomic wavefunctions.

5. Collective states of N indistinguishable atoms

5.1. General description

As in the two-atom case, the conserved symmetry properties of the multi-atom wavefunction with respect to local atomic permutations determine the constructive or destructive nature of the interatomic interference. The description of the cooperative evolution (superradiance as well as subradiance) requires therefore a basis of collective states having well defined symmetry types, i.e. a basis labelled according to S_N irreducible representations. However this labelling does not completely define the collective states, even for two-level atoms. In this latter case, as well known, Dicke's description of collective states (Dicke 1954) uses the pseudo-angular momentum properties of two-level atoms. This can be generalised for n-level atoms. A completely defined basis of collective states of the N atoms of a slice S_k is obtained using a

non-invariance property of the atom-field Hamiltonian (4.1) (Crubellier 1977). The atomic operators which are involved in this Hamiltonian are all of the type

$$P_{ij} = \sum_{\alpha=1,N} |i\rangle_{\alpha\alpha}\langle j| \tag{5.1}$$

i and j corresponding to monatomic states. They can be written as sums of operators acting on the different slices S_k:

$$P_{ij} = \sum_k P_{ij}^{(k)} \tag{5.2}$$

with

$$P_{ij}^{(k)} = \sum_{\alpha \in S_k} |i\rangle_{\alpha\alpha}\langle j|. \tag{5.3}$$

For i and j running from 1 to n, these latter operators are the infinitesimal operators of a SU(n) group which is the generalisation of Dicke's SU(2) cooperation group. This SU(n) group commutes with the S_N group corresponding to the same slice and the collective states of each slice can be labelled according to the irreducible representations of the direct product of two groups, $S_N \times SU(n)$. For two-level atoms it leads to Dicke's basis; ψ_{gmr} states with given r and g correspond to an irreducible representation of $S_N \times SU(2)$. For the N indistinguishable n-level atoms of a slice S_k, since the Hamiltonian is invariant under the permutation group S_N and contains, as atomic operators, infinitesimal operators of SU(n) only, the evolution of the system will conserve the irreducible representations (irreps) of $S_N \times SU(n)$. In particular the evolution of the irrep-diagonal density matrix elements is not related to the evolution of the irrep-off-diagonal ones and the trace of the atomic density matrix over the space of an irreducible representation is constant.

The explicit construction of collective basis states having well defined symmetry properties, i.e. of basis states for the irreducible representations of $S_N \times SU(n)$ is most easily realised using Young tableaux theory (see, for example, Judd 1973) which will be outlined hereafter. In this theory, one first defines N-cell diagrams with h successive rows of $l_1, l_2, \ldots, l_h$ cells (with $l_1 \geqslant l_2 \geqslant \ldots \geqslant l_h$ and $l_1 + l_2 + \ldots + l_h = N$), called Young diagrams, which correspond to partitions of N. Standard Young tableaux are then defined by numbering in a specific way the N cells of the diagram. N-atom wavefunctions are finally associated with each standard Young tableau after mapping of an ensemble of N monatomic states on the tableau cells, i.e. on the atomic indices. The product of the monatomic states corresponding to each row of the tableau is first constructed and then symmetrised with respect to the atomic indices concerned. The product of the functions corresponding to the different rows is finally written and the N-atom wavefunction obtained is antisymmetrised with respect to the atomic indices of each column. Such a wavefunction is characterised

(i) by a Young diagram χ which corresponds to an irreducible representation $\{l_1, l_2, \ldots, l_h\}$ of S_N and which is what we called a symmetry type (Crubellier *et al* 1980);

(ii) by a standard Young tableau η associated to the Young diagram χ, which characterises an irreducible representation of $S_N \times SU(n)$;

(iii) by the mapping of an ensemble of N monatomic states on the tableau cells.

As a general result of Young tableaux theory, the wavefunctions constructed in such a way from different standard Young tableaux are linearly independent. The wavefunctions corresponding to a given standard Young tableau span the space of the corresponding irreducible representation of $S_N \times SU(n)$ and the wavefunctions associated

to all different standard Young tableaux span the whole space of collective states of the N n-level atoms we are dealing with. However the number of functions associated to a standard Young tableau is generally larger than the dimension of the corresponding representation space and the constructed functions do not form a basis for this space. A wavefunction associated to a standard Young tableau can be written as the product of:

(i) a symmetrised product of l_h fully antisymmetrical h-atom wavefunctions associated to the h-cell columns of the Young tableau; an example of such a function for $h=3$ is

$$|i\rangle_\alpha|j\rangle_\beta|k\rangle_\gamma+|j\rangle_\alpha|k\rangle_\beta|i\rangle_\gamma+|k\rangle_\alpha|i\rangle_\beta|j\rangle_\gamma-|i\rangle_\alpha|k\rangle_\beta|j\rangle_\gamma-|k\rangle_\alpha|j\rangle_\beta|i\rangle_\gamma-|j\rangle_\alpha|i\rangle_\beta|k\rangle_\gamma$$

where i, j and k are monatomic states (corresponding to a given 3-cell column of the tableau) and α, β and γ are atomic indices

(ii) a symmetrised product of $(l_{h-1}-l_h)$ fully antisymmetrical $(h-1)$-atom wavefunctions associated to $(h-1)$-cell columns of the tableau;

(iii) ...;

(iv) a symmetrised product of (l_2-l_3) antisymmetrical two-atom wavefunctions associated to the two-cell columns of the tableau;

(v) a fully symmetrical (l_1-l_2)-atom wavefunction associated to the single-cell columns of the tableau.

It is therefore clear that the wavefunction associated to a standard Young tableau cannot contain a given monatomic state more than once in a given column; in particular, the Young diagram cannot have more than n rows ($h\leqslant n$).

An invariant of the $S_N\times SU(n)$ group of the slice S_k is the Casimir operator (Judd 1973) $W^{(k)}$ defined by

$$W^{(k)}=\sum_{i,j}P_{ij}^{(k)}P_{ji}^{(k)} \tag{5.4}$$

where the sums run over all monatomic states. This operator characterises the symmetry properties of the atomic system under permutations of the atoms of the slice S_k in the sense that its eigenvalues depend on the symmetry type $\{l_1, l_2, \ldots, l_k\}$ of the wavefunction only. More precisely the previously constructed wavefunctions are eigenstates of $W^{(k)}$ with eigenvalues

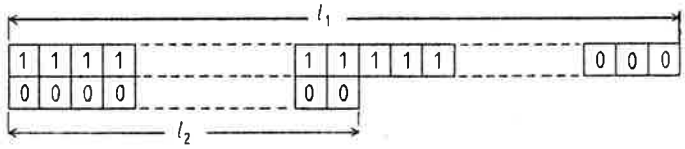
$$W^{(k)}[\{l_1, l_2, \ldots, l_k\}]=\sum_{i=1,h}l_i(l_i+n+1-2i). \tag{5.5}$$

As already mentioned, for two-level atoms the collective state basis which is described here is very close to Dicke's basis: a detailed comparison is given in the next section.

5.2. Two-level atoms and Dicke's states

Let us consider N atoms with two levels, $|1\rangle$ (upper level) and $|0\rangle$ (lower level), connected by an electric dipole transition. A collective state constructed with the general method described in § 5.1 is characterised by a symmetry type $\{l_1, l_2\}$ (with $l_1\geqslant l_2$ and $l_1+l_2=N$), by a standard Young tableau and by an ensemble of monatomic states associated to the tableau cells. The most general form for such a wavefunction

is given by the following tableau



where the numbers placed in the cells refer to monatomic states (the numbering of the standard Young tableau is omitted). Let N_1 and N_0 be respectively the number of atoms in states $|1\rangle$ and $|0\rangle$; one has clearly $N_1+N_0=N$ and $l_2 \leqslant N_i \leqslant l_1$ $(i=0,1)$. The states corresponding to the different values of N_1 and N_0 are linearly independent. They provide thus a basis of collective states which are characterised by l_1, l_2, a given standard Young tableau, N_1 and N_0.

In order to compare these collective states with the well known Dicke's states, we write Dicke's pseudo-spin collective operators, R_z, R_+ and R_-, in terms of P_{ij} operators (see equation (5.3); the index of the slice is omitted here)

$$\begin{aligned} R_z &= (P_{11}-P_{00})/2 \\ R_+ &= P_{10} \\ R_- &= P_{01}. \end{aligned} \tag{5.6}$$

One has therefore

$$R^2=[(P_{11}-P_{00})^2+2(P_{10}P_{01}+P_{01}P_{10})]/4. \tag{5.7}$$

As is well known, Dicke's states are eigenstates of R_z and R^2 with eigenvalues m and $r(r+1)(|m| \leqslant r,\ 0 \leqslant r \leqslant N/2$ for even N, $\frac{1}{2} \leqslant r \leqslant N/2$ for odd $N)$. On the other hand, the previous collective states are eigenstates of P_{11} and P_{00} with eigenvalues N_1 and N_0. They are also eigenstates of the Casimir operator (see equation (5.4)),

$$W=P_{11}^2+P_{00}^2+P_{10}P_{01}+P_{01}P_{10} \tag{5.8}$$

with eigenvalues $[l_1(l_1+1)-l_2(l_2-1)]$; since, from equation (5.7),

$$R^2=(W-N^2/2)/2 \tag{5.9}$$

they are finally eigenfunctions of R_z and R^2 with eigenvalues $(N_1-N_0)/2$ and $(l_1-l_2)(l_1-l_2+2)/4$, so that they are identical to Dicke's states ψ_{gmr} with $r=(l_1-l_2)/2$ and $m=(N_1-N_0)/2$. The index g represents in our description the numbering of a given standard Young tableau.

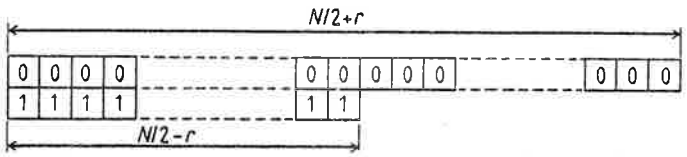
In the following part of this paper, it will be shown how these results allow one to predict the existence of subradiance and to calculate the amount of finally non-de-excited atoms, which will be called the subradiance rate. This will be made first for two-level atoms, and then for three-level atoms in the 'V' configuration.

6. Symmetry properties and subradiance rate

6.1. Two-level atoms

For a given slice S_k, Dicke's collective states ψ_{g-rr} have the smallest energy compatible with their symmetry type, $\{N/2+r,\ N/2-r\}$. If r is different from $N/2$, they are

subradiant: in effect the indistinguishability of the atoms, and, therefore, the previously discussed conservation of symmetry type prevent them from radiating. Such a state is represented by a Young tableau of the type

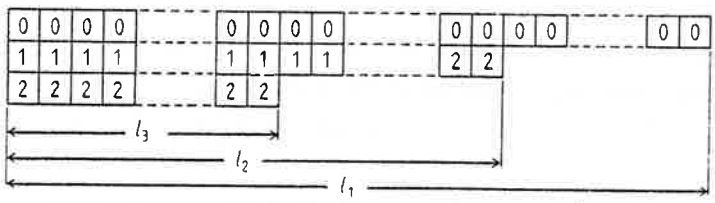


The number of atoms in the excited state, $N/2-r$, depends on the symmetry type only. Following the description of § 5.1, this state can be considered as formed by l_1-l_2 atoms in the ground state and by l_2 pairs of atoms in a two-atom antisymmetrical state. As in the two-atom case, destructive interference occurs between the atoms of each pair, which explains the subradiance of the N-atom state. A complete atomic state which is the product of subradiant states corresponding to the different slices is obviously subradiant: in effect the different slices are independent from each other when they do not radiate.

Because of the symmetry type conservation (see § 4) such a state can be reached by the atomic system only if the initial state has the same symmetry type, i.e. $\{N/2+r, N/2-r\}$ with $r \neq N/2$. In the superradiance experiments which have been realised up to now with non-degenerate two-level atoms (Gibbs *et al* 1977), the system was prepared in a complete population inversion, i.e. in a fully symmetrical state, so that no subradiance was predicted. To our knowledge no excitation process leading to a state with another symmetry type has ever been proposed.

6.2. Three-level atoms

A collective state of three-level atoms in the 'V' configuration which has the smallest energy compatible with its symmetry type can be represented by a Young tableau of the type



The number of de-excited atoms is equal to l_1 and the number of atoms which are excited either in state $|1\rangle$ or in state $|2\rangle$ is l_2+l_3. According to the description of §5.1, such a state can be considered as formed by l_1-l_2 de-excited atoms, l_2-l_3 pairs of atoms in antisymmetrical states, which are subradiant either for transition $1\rightarrow 0$ or for transition $2\rightarrow 0$, and by l_3 trios of atoms in fully antisymmetrical states in which destructive interference appears for both transitions. This state is therefore subradiant except if $l_2=l_3=0$. As for two-level atoms, a collective state of the whole active volume which is the product of subradiant states corresponding to the different slices is subradiant. The amount of atoms which are in an excited state, i.e. the subradiance rate, is $\tau=(l_2+l_3)/N$, with $0<\tau\leqslant\frac{2}{3}$.

Because of the local symmetry-type conservation, a subradiant state with subradiance rate $\tau=(l_2+l_3)/N$ can be spontaneously reached by the atoms of a slice S_k only if the corresponding initial state has the same symmetry type $\{l_1, l_2, l_3\}$. One can assert in this latter case that the subradiance rate of the slice S_k is equal to or larger than $(l_2+l_3)/N$. Quite generally the knowledge of the initial density matrix and of its expansion over the spaces of given symmetry type allows one to predict a minimal value for the subradiance rate τ of the system. One has

$$\tau \geqslant \sum_k \sum_{\{l_1,l_2,l_3\}} f_k(\{l_1, l_2, l_3\}) \frac{l_2+l_3}{N} \tag{6.1}$$

where the sums run over the indices of slices and over the symmetry types $\{l_1, l_2, l_3\}$; f_k $(\{l_1+l_2, l_3\})$ is a partial trace of the density matrix, taken over the space of wavefunctions of the slice S_k having the symmetry type $\{l_1, l_2, l_3\}$. In the particular case where state $|0\rangle$ is initially empty, the only symmetry types which are of concern are $\{l_1, l_2\}$ and one has

$$\tau \geqslant \sum_k \sum_{\{l_1,l_2\}} f_k(\{l_1, l_2\}) \frac{l_2}{N}. \tag{6.2}$$

Contrary to the case of two-level atoms, a number of initial conditions leading to non-zero subradiance rates can be thought of. These conditions will be discussed in the following, together with a brief study of the evolution, through cooperative spontaneous emission, of the collection of three-level atoms.

7. Cooperative spontaneous emission of three-level atoms and subradiance

In this section, we shall briefly study the superradiant emission of three-level atoms in the 'V' configuration, assumed to be initially in the excited levels $|1\rangle$ and $|2\rangle$. Semiclassical results corresponding to the initial pure state and to full initial statistical mixing (diagonal density matrix) will first be discussed. Analogous effects will then be predicted in the mean-field Markovian master equation treatment (Bonifacio and Lugiato 1975a, b), for a symmetrical initial state and 'most antisymmetrical' initial state. Propagation effects, which can be taken into account in a quantum-mechanical treatment (Ressayre and Tallet 1978), are ignored here, because we are only interested in a qualitative comparison with the semiclassical treatment. Pure states and symmetrical states are easily identified. The correspondence which appears between statistical mixing and antisymmetry will be explained in the next section.

7.1. Semi-classical model

Bloch-Maxwell equations describing the evolution, in the semiclassical model of a collection of N atoms with three levels in the 'V' configuration (figure 1) are given by

$$\frac{\partial}{\partial t'} Q_{11} = \frac{\mathrm{i} d_1}{\hbar} (\varepsilon_1^* Q_{10} - \varepsilon_1 Q_{01})$$

$$\frac{\partial}{\partial t'} Q_{22} = \frac{\mathrm{i} d_2}{\hbar} (\varepsilon_2^* Q_{20} - \varepsilon_2 Q_{02})$$

$$
\begin{aligned}
&\frac{\partial}{\partial t'} Q_{00} = -\frac{\partial}{\partial t'}(Q_{11} + Q_{22}) \\
&\frac{\partial}{\partial t'} Q_{12} = -\frac{\mathrm{i}}{\hbar}(d_1 \varepsilon_1 Q_{02} - d_2 \varepsilon_2^* Q_{10}) \\
&\frac{\partial}{\partial t'} Q_{10} = \frac{i}{\hbar}[d_1 \varepsilon_1 (Q_{11} - Q_{00}) + d_2 \varepsilon_2 Q_{12}] \\
&\frac{\partial}{\partial t'} Q_{20} = \frac{i}{\hbar}[d_2 \varepsilon_2 (Q_{22} - Q_{00}) + d_1 \varepsilon_1 Q_{21}] \\
&\frac{\partial}{\partial z} \varepsilon_1 = -\frac{\mathrm{i} k_1 d_1}{2\varepsilon_0} Q_{10} \\
&\frac{\partial}{\partial z} \varepsilon_2 = -\frac{\mathrm{i} k_2 d_2}{2\varepsilon_0} Q_{20}.
\end{aligned}
\tag{7.1}
$$

In these equations, ε_1 and ε_2 are the slowly varying envelopes of the electromagnetic fields associated with transitions $1 \leftrightarrow 0$ and $2 \leftrightarrow 0$. Q_{00}, Q_{11} and Q_{22} are the populations of levels $|0\rangle$, $|1\rangle$ and $|2\rangle$, Q_{12} is the slowly varying envelope of the coherence between levels $|1\rangle$ and $|2\rangle$ and Q_{10} and Q_{20} are those of the optical coherences corresponding to transitions $1 \leftrightarrow 0$ and $2 \leftrightarrow 0$. The time t' is the retarded time $t' = t - z/c$. For the sake of simplicity equations (7.1) are written only for the waves travelling forward along the cylinder axis direction, but the numerical calculations take into account both forward and backward directions.

Examples of semiclassical results are given in figures 2, 3 and 4. Figures 2 and 3 concern the same atomic level system, with $\Gamma_1 \mu_1 = 1.44 \Gamma_2 \mu_2$ and the initial populations of levels $|1\rangle$ and $|2\rangle$ are equal ($Q_{11}(0) = Q_{22}(0) = 0.5$). Figure 2 corresponds to an atomic initial state which is a pure state ($Q_{12}(0) = 0.5$), whereas figure 3 corresponds to a full initial statistical mixing ($Q_{12}(0) = 0$). In figure 4 the results one would obtain if the two transitions were not coupled by their common lower level are represented.

7.1.1. Initial pure state. Initiation effect. For an initial pure state (figure 2), the coherence Q_{12} remains maximum ($|Q_{12}|^2 = Q_{11} Q_{22}$) during the whole evolution, the state remains a pure one and levels $|1\rangle$ and $|2\rangle$ de-excite simultaneously. Emission occurs simultaneously on both transitions and both emissions are accelerated and enhanced (compare figures 2 and 4), especially the emission on the less probable transition ($2 \leftrightarrow 0$), which is triggered or 'initiated' (Crubellier 1977, Crubellier and Schweighofer 1978) by the emission on the other transition.

7.1.2. Initial statistical mixing. Inhibition effect and subradiance. If the coherence Q_{12} is initially zero, excited levels $|1\rangle$ and $|2\rangle$ are generally not empty at the end of the evolution (Raimond 1979, Crubellier *et al* 1980, Gross 1980, Pillet 1982). In the case of equal initial populations of these levels, the total population of excited levels tends to $\frac{1}{2}$ (figure 3), whereas coherence Q_{12} tends to be maximum. Figure 3 shows that superradiant emission appears preferentially on transition $1 \rightarrow 0$ and that the emission on $2 \rightarrow 0$ is strongly quenched or 'inhibited' (Crubellier 1977, Crubellier and Schweighofer 1978). Of course, the total number of emitted photons corresponding to figure 3 is half the total number of emitted photons corresponding to figures 2 and 4: this can be checked by comparing the pulse areas in the three cases.

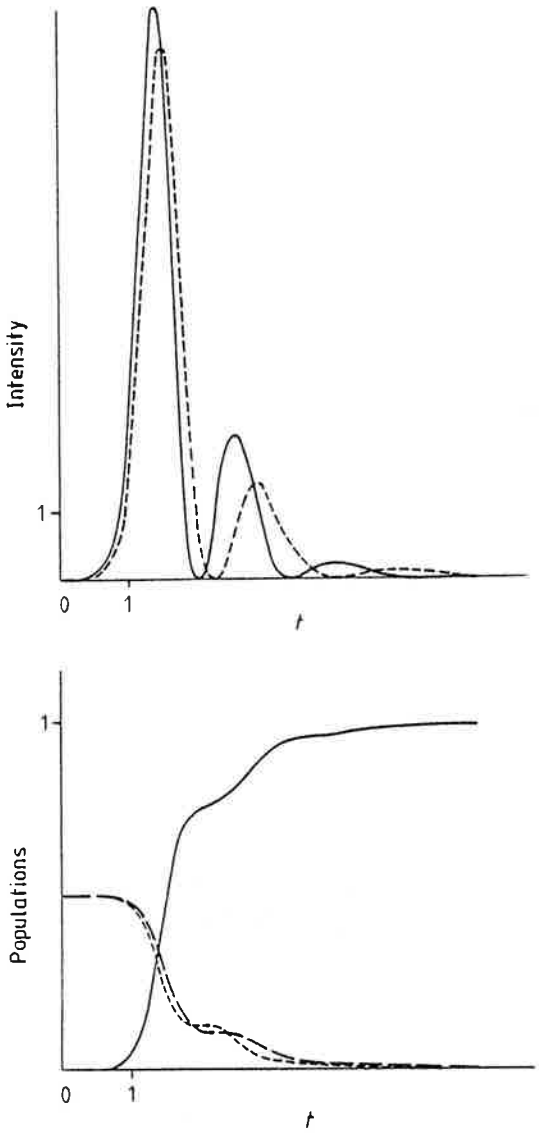


Figure 2. Semiclassical results for an initial pure state ($Q_{11} = Q_{22} = |Q_{12}| = 0.5$) in the case $\Gamma_1\mu_1 = 1.44\Gamma_2\mu_2$. The upper curves show the time evolution, in arbitrary units, of the intensity radiated on the transition $1 \to 0$ (——) and on the transition $2 \to 0$ (— — —); the lower curves show the time evolution of the population of level 1 (— — —), level 2 (- - - -) and level 0 (——).

As predicted in § 2 from statistical mixing conservation, subradiance is obtained when the atomic initial state (in which the atoms are implicitly assumed to be uncorrelated) is not a pure state. It is shown here that this effect is accompanied by a quenching of the emission on the less probable transition. However, the reason why initial statistical mixing causes both effects is not obvious and will be analysed in § 8.

7.2. Markovian master equation

Trying to explain, at least qualitatively, the different effects which appear in the semiclassical results, we shall consider a mean-field Markovian quantum-mechanical treatment. The evolution of N indistinguishable atoms is then given by a master equation (Crubellier 1977, equation (1)) and the system is simply cascading spontaneously among its collective states. Two extreme cases corresponding to N atoms initially excited either in state $|1\rangle$ or in state $|2\rangle$ will be considered here: the case of a fully symmetrical initial collective state and the case of an initial state with the 'most antisymmetrical' symmetry type, $\{N/2, N/2\}$.

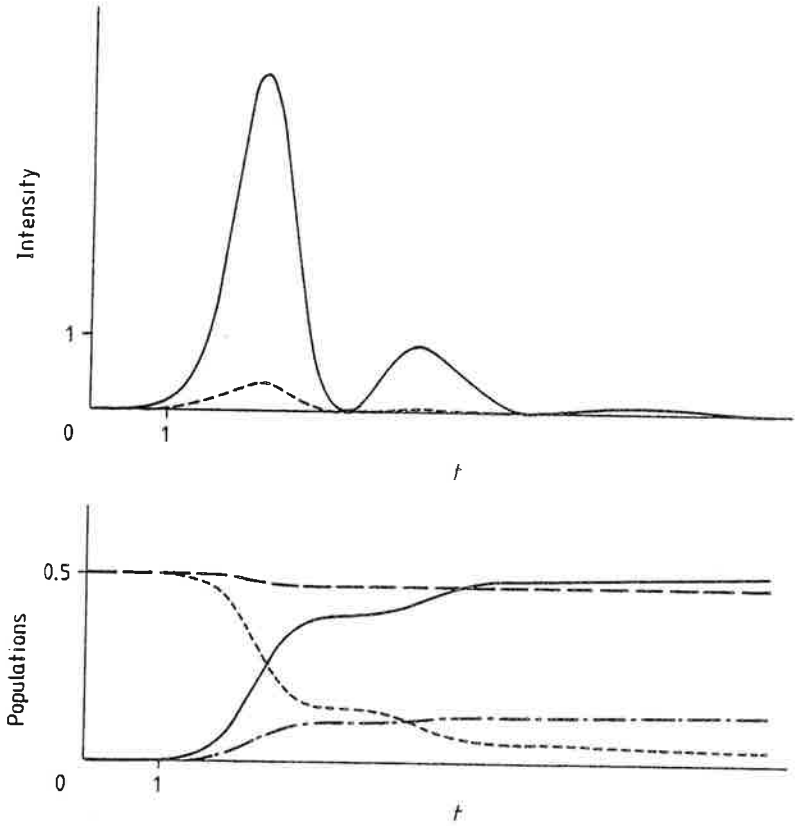


Figure 3. Same as figure 2 for an initial full statistical mixing ($Q_{11} = Q_{22} = 0.5$, $Q_{12} = 0$). The time evolution of the coherence $|Q_{12}|$ is also shown (— · —) in the lower curves. The arbitrary units are the same as in figure 2.

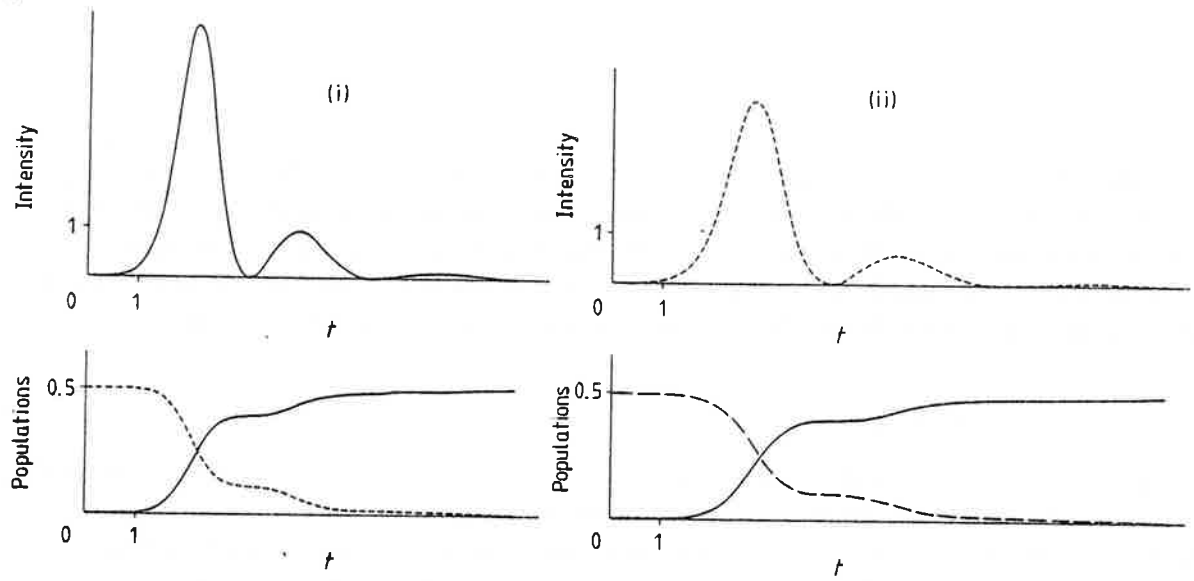


Figure 4. Semiclassical results for the transition $1 \rightarrow 0$ (case (i)) and for the transition $2 \rightarrow 0$ (case (ii)) when the coupling between the transitions is suppressed; the calculations are performed in the same conditions as in figures 2 and 3 except that $\Gamma_2 = 0$ in case (i) and $\Gamma_1 = 0$ in case (ii). The different curves correspond to the same quantities as in figures 2 and 3 and the arbitrary units are the same as in these figures.

7.2.1. Symmetrical initial state. The first case, with symmetrical initial state, has been already discussed (Crubellier and Schweighofer 1978, case (b)). The initiation of the emission on the less probable transition was predicted, i.e. the same effect as the one obtained in the semiclassical calculations when the initial state is a pure state. This convergence is not surprising since the two initial situations are actually identical. Our semiclassical calculations assume a homogeneous factorisable initial density matrix which is a pure state and can be written as

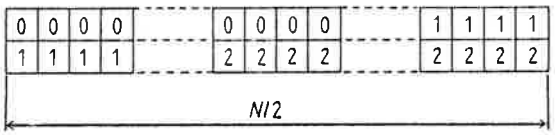
$$\rho(0) = |S\rangle\langle S|$$

where $|S\rangle$ is a fully symmetrical collective wavefunction,

$$|S\rangle = \prod_{\alpha=1,N} |\phi\rangle_\alpha \tag{7.2}$$

$|\phi\rangle$ being a linear combination of states $|1\rangle$ and $|2\rangle$.

7.2.2. 'Most antisymmetrical' initial state. The case of the 'most antisymmetrical' initial state can be discussed in the same way as the case of symmetrical initial state. Let us consider N indistinguishable three-level atoms with an initial state of symmetry type $\{N/2, N/2\}$ (N is assumed to be even, for the sake of simplicity). For this particular symmetry type, the wavefunctions are of the type



and they are fully characterised by the numbers of columns containing a given pair of states, n_{01}, n_{02} and n_{12}. One has $\Sigma_{i<j} n_{ij} = N/2$ and

$$\begin{aligned} N_0 &= n_{01} + n_{02} \\ N_1 &= n_{01} + n_{12} \\ N_2 &= n_{02} + n_{12}. \end{aligned} \tag{7.3}$$

The result of the action of the energy-decreasing operator corresponding to the transition $1 \leftrightarrow 0$, for example, which is given by

$$R_1^- = \sum_{\alpha=1,N} |0\rangle_{\alpha\alpha}\langle 1| \tag{7.4}$$

is the sum of all wavefunctions which are obtained by replacing, in the above tableau, one of the indices 1 by 0 (this leads of course to the zero wavefunction when the index 1 belongs to a subradiant pair of indices). The probability per unit time of the emission on the transition $1 \to 0$ (respectively $2 \to 0$) is equal to $\Gamma_1 \mu_1 n_{12}\ (n_{01}+1)$ (respectively $\Gamma_2 \mu_2 n_{12}\ (N_{02}+1)$). The collective energy level diagram corresponding to all states associated with a standard Young tableau is given in figure 5, together with the values of the collective transition probabilities. The lowest points of the diagram correspond to states with $n_{12} = 0$, i.e. with $N_0 = N_1 + N_2 = N/2$ and they are subradiant. The branching ratio between the probability of the emission on $1 \to 0$ and $2 \to 0$ at a point of the diagram is $\Gamma_1 \mu_1\ (n_{02}+1)/\Gamma_2 \mu_2\ (n_{01}+1)$. If $\Gamma_1 > \Gamma_2$ and if the system starts from the top of the diagram, this ratio is always larger than one and the emission of each photon on $1 \to 0$ makes it even larger (since n_{02} increases and n_{01} decreases by one

unit). The emission occurs therefore essentially on $1 \rightarrow 0$ and the emission on $2 \rightarrow 0$ is expected to be almost completely quenched. The same inhibition effect on the emission on the less probable transition has been already predicted (Pillet 1977, Crubellier and Schweighofer 1978) for three-level atoms in the 'Λ' configuration (two transitions sharing a common upper level), and the similitude of the two cases appears when comparing figure 5 in the present text and figure 1(a) in the above quoted reference. In the present case, subradiance is an unavoidable consequence of the symmetry properties of the collective state and its occurs whatever the ratio Γ_1/Γ_2 is. If $\Gamma_1 \neq \Gamma_2$, in addition, the population remains essentially in one of the excited levels, due to the quenching effect. As previously shown, quenching and subradiance are both obtained by semiclassical calculations too, in the case of initial statistical mixing. The reason for such a similitude is not obvious, since symmetry and statistical mixing are quite different physical properties. We shall elucidate this point in the next section, by studying the statistical symmetry properties of the initial states used in our semiclassical calculations.

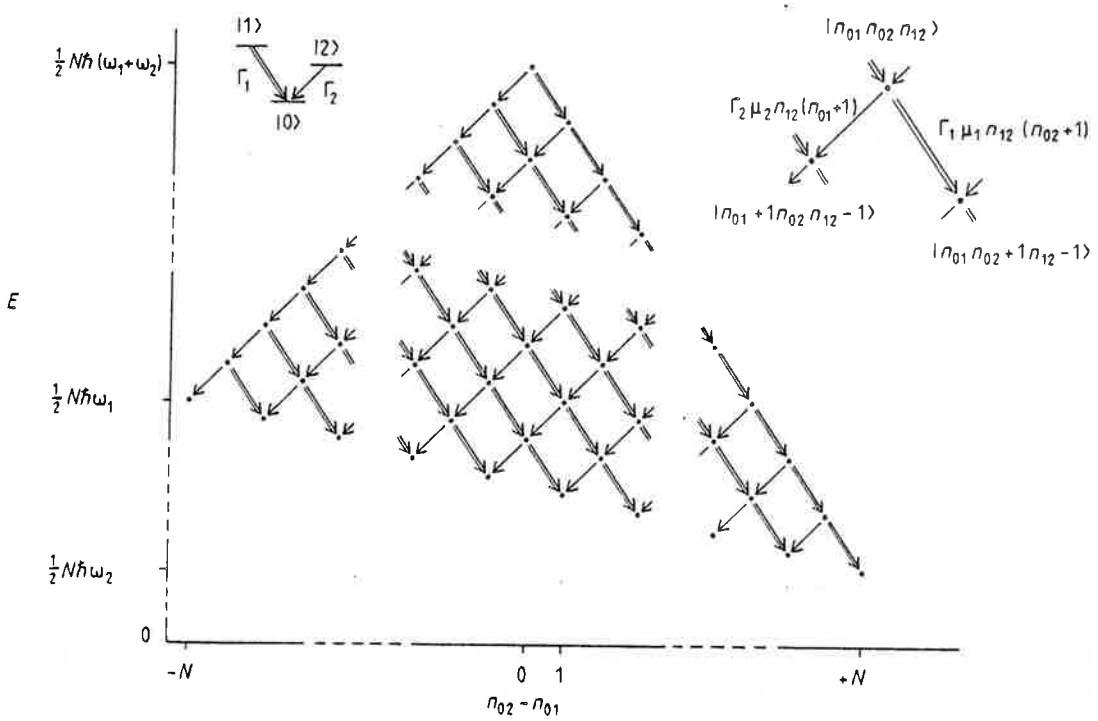


Figure 5. Subradiance and inhibition effect for three-level atoms with a 'most symmetrical' initial state. The figure includes three diagrams. First, the energy level diagram of one single atom. Second, a diagram representing the cascading emission between the collective states of N such atoms; each point represents a 'most antisymmetrical' collective state, $|n_{01}n_{02}n_{12}\rangle$, which contains n_{0i} (respectively n_{02}, n_{12}) pairs of atoms in antisymmetrical states constructed with the monatomic states $|0\rangle$ and $|1\rangle$ (respectively $|0\rangle$ and $|2\rangle$, $|1\rangle$ and $|2\rangle$); the double and single arrows correspond respectively to the emission of one photon on the transitions $1 \rightarrow 0$ and $2 \rightarrow 0$. Third the general expression for the collective transition probabilities. The bottom states of the diagram correspond to $n_{12} = 0$ and contain therefore subradiant pairs only.

8. Statistical mixing and symmetry properties

The constructive or destructive nature of the interatomic interference is determined by the symmetry properties of the collective atomic states of the slices S_k under

permutations of atoms inside S_k. If the atomic state is represented by a density matrix, interatomic interference properties are only statistically defined. They are characterised by the partial traces of the reduced density matrices of the slices S_k, the traces being taken over the spaces of wavefunctions of S_k corresponding to a given symmetry type. Assuming that all atoms are either in state $|1\rangle$ or in state $|2\rangle$ and that the density matrix is factorisable and homogeneous, one has

$$\rho = \prod_k \rho^{(k)} \tag{8.1}$$

where the product runs over successive slices S_k and where

$$\rho^{(k)} = \prod_{\alpha \in S_k} \rho^{\alpha} \tag{8.2}$$

(the product running over all the atoms of the slice). We shall consider the diagonal form of the monatomic density matrix,

$$\rho^{\alpha} = \rho_{aa} |a\rangle_{\alpha\alpha}\langle a| + \rho_{bb} |b\rangle_{\alpha\alpha}\langle b|. \tag{8.3}$$

We now define collective states of slice S_k which are of the form

$$|\psi\rangle_k = \prod_{\alpha \in S_k} |i_\alpha\rangle_\alpha \tag{8.4}$$

where $|i_\alpha\rangle$ is either $|a\rangle$ or $|b\rangle$, the product containing $N_a|a\rangle$ terms and $N_b|b\rangle$ terms. The density matrix $\rho^{(k)}$ is diagonal in N_a and N_b and

$$\rho^{(k)}|\psi\rangle_k = (\rho_{aa})^{N_a}(\rho_{bb})^{N_b}|\psi\rangle_k. \tag{8.5}$$

The same relation holds for any linear combination of states of the form (8.4) with given N_a and N_b values, in particular for any state having a symmetry type compatible with these values. Since all atoms are either in state $|a\rangle$ or in state $|b\rangle$, a non-orthogonal basis of collective states of slice S_k is provided by states which are characterised by: a symmetry type $\{l_1, l_2\}$, a standard Young tableau η corresponding to this symmetry type; and a couple of N_a and N_b values (with $N_a + N_b = N$). The $(l_1 - l_2 + 1)$ possible values of N_a and N_b are given by

$$l_2 \leqslant N_i \leqslant l_1 \qquad i = a, b.$$

The partial trace of the density matrix $\rho^{(k)}$ over a space of symmetry type $\chi = \{l_1, l_2\}$ is given by

$$f_k(\chi) = \sum_{\eta, \eta', N_a, N_b} (\chi\eta N_a N_b|\rho|\chi\eta' N_a N_b) g^{\chi\eta,\chi\eta'} \tag{8.6}$$

where the sum runs over all standard Young tableaux η and η' and over all N_a and N_b values compatible with the symmetry type χ; $g^{\chi\eta,\chi\eta'}$ is the metric tensor, which is diagonal in χ, N_a and N_b and does not depend on N_a and N_b,

$$(\chi\eta N_a N_b|\chi'\eta' N'_a N'_b) = \delta_{\chi\chi'} \delta_{N_a N'_a} \delta_{N_b N'_b} g_{\chi\eta,\chi\eta'}. \tag{8.7}$$

From equation (8.5), one has

$$(\chi\eta N_a N_b|\rho|\chi\eta' N_a N_b) = (\rho_{aa})^{N_a}(\rho_{bb})^{N_b} g_{\chi\eta,\chi\eta'}. \tag{8.8}$$

The sum over η and η' in equation (8.6) is easily carried out, since

$$\sum_{\eta,\eta'} g_{\chi\eta,\chi\eta'} g^{\chi\eta,\chi\eta'} = Y(\chi)$$

$Y(\chi)$ being the number of standard Young tableaux associated with the symmetry type χ, i.e. the dimension of the corresponding irreducible representation of S_N (see for example Wybourne 1970),

$$Y(\{l_1, l_2\}) = \frac{(l_1 - l_2 + 1)N!}{(l_1+1)!l_2!}. \tag{8.9}$$

A straightforward calculation gives then

$$\sum_{N_a, N_b (N_a+N_b=N, l_2 \leq N_a, N_b \leq l_1)} (\rho_{aa})^{N_a} (\rho_{bb})^{N_b} = \frac{(\rho_{aa})^{l_1+1} - (\rho_{bb})^{l_2+1}}{\rho_{aa} - \rho_{bb}}. \tag{8.10}$$

The eigenvalues ρ_{aa} and ρ_{bb} of the monatomic density matrix are completely determined by the trace of its square, σ^2:

$$\begin{aligned} \rho_{aa} &= (1+\omega)/2 \\ \rho_{bb} &= (1-\omega)/2 \end{aligned} \tag{8.11}$$

with $\omega = (2\sigma^2 - 1)^{1/2}$. This allows one to finally write the partial trace (8.6) in terms of l_1, l_2 and σ^2 only:

$$f_k(\{l_1, l_2\}) = \frac{(l_1 - l_2 + 1)N!}{(l_1+1)!l_2!} \frac{(1-\omega^2)^{l_2}}{2^{N+1}\omega} [(1+\omega)^{l_1 - l_2 + 1} - (1-\omega)^{l_1 - l_2 + 1}]. \tag{8.12}$$

The number N, which is the number of locally indistinguishable atoms, is assumed to be large. As a function of $l_1 - l_2$, the partial trace (8.12) has a sharp maximum at about $l_1 - l_2 = N\omega$, with a width of the order of $N^{1/2}$. Therefore the probability for the collective state of slice S_k to have a symmetry type given by:

$$\begin{aligned} l_1 &= N/2(1+\omega) = N\rho_{aa} \\ l_2 &= N/2(1-\omega) = N\rho_{bb} \end{aligned} \tag{8.13}$$

is close to unity. We find again, in this way, that a pure state ($\sigma^2 = 1$) corresponds to the fully symmetrical symmetry type $\{N\}$. We find also that a full statistical mixing with equal weights of states $|1\rangle$ and $|2\rangle$ ($\sigma^2 = \frac{1}{2}$) has an almost fully determined symmetry type, $\{N/2, N/2\}$. The quenching and subradiance effects obtained semiclassically can therefore also be explained by destructive interatomic interference. In addition, a minimum value for the subradiance rate can be evaluated for any homogeneous factorisable initial density matrix. It is given by

$$\tau \geq (1-\omega)/2 = \rho_{bb} \tag{8.14}$$

and this shows again that subradiance is expected except for initial pure states.

A somewhat surprising link between the amount of statistical mixing and the symmetry properties of the atomic state under the permutations of indistinguishable atoms has been pointed out here. Even more surprisingly, the factorisable density matrix ρ, which obviously describes uncorrelated atoms, appears to be equivalent, as far as the interatomic interference is concerned, to collective states with well defined symmetry properties, which are characterised by their atom-atom correlations. This is possible because of the very large number of such collective states, which allows

the correlations to statistically cancel in the expansion of ρ over these states. In the study of cooperative spontaneous emission, this result provides a valuable link between the usually non-cooperative excitation process, which prepares uncorrelated atoms, and the cooperative de-excitation. For this latter process, the symmetry properties are indeed essential since they allow one to predict directly the constructive or destructive nature of the interference.

9. Obstacles to subradiance

According to the preceding results, a collection of three-level atoms in the 'V' configuration could seem to remain indefinitely in a subradiant state once it had reached it. In fact, as the photon trapping is due to a destructive interatomic interference, any restriction of the local indistinguishability of the atoms will disturb the subradiance phenomenon, either by modifying the evolution of the atoms toward a subradiant state or by allowing the decay of these states. In this section, the assumptions and approximations which have been made in the preceeding sections are discussed. Extended pencil-shaped samples are still considered and the atomic density is assumed to be smaller than λ^{-3} so that the dipole-dipole interaction can be ignored; departure from the plane-wave approximation and 'dephasing' effects only are studied.

9.1. Plane-wave approximation

As the plane-wave approximation supports the local indistinguishability property, any departure from this approximation will fundamentally disturb the cooperative behaviour of the system and it can possibly modify the features of superradiant emission as well as those of subradiance. In the plane-wave approximation, any emission in the transverse modes of the emitting volume is neglected. Ordinary spontaneous emission in these modes cannot disturb appreciably the superradiant emission, because of its much longer characteristic time. On the other hand, cooperative spontaneous emission in the transverse modes has been proved to be negligible during the whole superradiant emission period, provided that the Fresnel number is of the order of one (Mattar *et al* 1981). In this case, the evolution of the atomic system is satisfactorily described in the plane-wave approximation, whether the final state is fully de-excited or subradiant. However this approximation does not hold anymore when the system does not radiate cooperatively, i.e. at the beginning of the emission and, when subradiance is expected, at the end of the superradiant emission. Because of the shortness of the time interval which corresponds to the beginning of superradiant emission (a few T_{SR} (Vrehen and Schuurmans 1979, Carlson *et al* 1980, Schuurmans *et al* 1981, Crubellier *et al* 1984)), the emission in the transverse modes has been found to be of small importance during this period, too. Concerning the end of the emission, the inhibition of spontaneous emission is predicted for the end-fire superradiant emission modes only. Ordinary spontaneous emission in the transverse modes is expected even in a subradiant state. This emission de-excites the atoms and therefore destroys the subradiant state in a time interval of the order of the characteristic time T_{sp} for non-cooperative spontaneous emission. A subradiant state can thus exist only after the emission, with a delay time T_D, of the superradiant light pulse which creates it and before its destruction by ordinary spontaneous emission: its duration cannot be longer than T_{sp}.

9.2. Dephasing effects

Dephasing effects due to atomic collisions or to the Doppler effect, for example, are known to be able to prevent superradiant emission. The threshold condition depends upon the characteristic dephasing time T_2^* and can be written (McGillivray and Feld 1976)

$$T_{SR} < T_2^*. \tag{9.1}$$

This condition means that the dephasing effects completely disturb the atomic local indistinguishability in a time of the order of T_2^* and that the interatomic interference disappears when T_2^* is smaller than the characteristic time T_{SR} for superradiant emission. In this latter case the emission is non-cooperative and neither superradiance nor subradiance may occur. When the threshold condition (9.1) holds, it is known for two-level superradiance that dephasing effects still delay and widen the emitted superradiant pulse, at least near threshold (Pillet 1977). The same result is expected for multilevel superradiance. In addition, dephasing effects can also change the features of the subradiance phenomenon. Two possible modifications are examined in the following: first, a modification of the superradiant emission which leads to a subradiant state and, in particular, of the subradiance rate of the state which is actually reached by the system; secondly, a modification of the decay of the subradiant state which, in the absence of dephasing effects, is due to non-cooperative emission. The influence of both atomic collisions and atomic motion are successively discussed.

9.2.1. Atomic collisions. If the average collision duration is small compared with T_{SR} (impact approximation; see also Crubellier *et al* 1983a) the average effect of atomic collisions is in general an exponential decay of the off-diagonal monatomic density matrix elements. As a consequence, the trace σ^2 of the squared monatomic density matrix decreases during the evolution of the system and the 'amount' of statistical mixing increases: the atomic de-excitation is thus expected to be even more severely limited than in the absence of collisions. This additional limitation of superradiance can also be explained by the symmetry properties of the collective atomic state. A general correspondence between statistical mixing and symmetry can indeed be obtained as follows. Let us consider a factorisable density matrix of N indistinguishable n-level atoms, with σ^2 as the trace of the squared monatomic density matrix. The mean value of the Casimir operator W of the $S_N \times SU(n)$ cooperation group, which is defined by equation (5.4), is related to σ^2 by

$$\langle W \rangle = nN + N(N+1)\sigma^2 \sim N^2\sigma^2. \tag{9.2}$$

The operator W is diagonal with respect to the symmetry type and its eigenvalues are decreasing when the symmetry 'decreases', i.e. when one or several cells of the corresponding Young diagram are removed from a row to a lower one. This operator provides thus a 'scale' for the symmetry with respect to the local atomic permutations (by analogy with a temperature scale) and, because of equation (9.2), σ^2 characterises the symmetry of the collective atomic state, at least on average. Any decrease of σ^2 implies a decrease in symmetry and thus, sooner or later, an enhancement of the destructive interatomic interference. The subradiance rate is therefore predicted to be increased by the atomic collision.

Once a subradiant state has been reached by the collection of atoms, the atomic collisions can still only diminish σ^2, and therefore the 'mean' symmetry of the collective state: they are of course unable to force the atoms to resume the cooperative emission.

The decay of the subradiant state occurs thus through ordinary spontaneous emission, as without collisions.

9.2.2. Doppler effect. The threshold condition associated with the Doppler effect has a particularly simple physical meaning. The characteristic time associated with the Doppler dephasing effect is, for an atomic vapour at temperature T,

$$T_2^* = 1/4\bar{v}(\ln 2)^{1/2} \tag{9.3}$$

where $\bar{v} = (2kT/m)^{1/2}$ is the quadratic mean value of the atomic velocity. The threshold condition (9.1) can thus be written as

$$\lambda/\bar{v} > T_{SR} \tag{9.4}$$

and it clearly means that the emission is cooperative provided that the N atoms of a slice S_k stay in this slice, i.e. remain indistinguishable, for a time interval which is on average larger than T_{SR}. Otherwise the interatomic interference cannot be efficient.

When the threshold condition (9.4) holds, semiclassical calculations including the Doppler effect (Crubellier *et al* 1980) show that the subradiance rate is the same as in the absence of atomic motion, even when the superradiant pulse is appreciably delayed and widened. This result agrees with the fact that the trace of the squared monatomic density matrix is still conserved when the Doppler effect is taken into account. The atomic motion has indeed two consequences on the cooperative evolution of the atoms. The first one is a dephasing effect, which can be understood as being due to a change of the atomic internal frequencies; this effect can obviously change the amplitude of the radiated field but it does not change the local invariance properties of the atom-field Hamiltonian. The second consequence of the atomic motion from slice to slice is to disturb the atomic local indistinguishability by changing the ensembles of indistinguishable atoms. However this motion does not change the local symmetry properties when, as is the case in the semiclassical model, the atomic state is represented by a factorisable density matrix which is initially spatially homogeneous. In any case the atomic motion from slice to slice could only diminish the useful atomic correlations. As for atomic collisions, the Doppler effect can only increase the subradiance rate and it does not accelerate the decay of the subradiant state, which is in both cases due to ordinary spontaneous emission.

10. Conclusion

Aiming essentially to define the conditions of the experimental observation of cooperative inhibition of spontaneous emission (Crubellier *et al* 1985, Pavolini *et al* 1985), the present theoretical study can be summarised as follows. Starting from the elementary case of a 'subradiant pair', one easily finds that spontaneous emission can be cooperatively inhibited in extended pencil-shaped samples too, in the end-fire cooperative field modes at least. A careful study of the atomic collective states shows then that the many-atom subradiant states can be defined as containing subradiant pairs, i.e. pairs of atoms in antisymmetrical states constructed with the upper and lower states of an atomic transition. Such subradiant states appear to be very difficult to prepare directly in an experiment. However, in the three-level 'V' configuration, the atoms can spontaneously reach subradiant states, starting from collective states which contain pairs of atoms in fully excited antisymmetrical states. In addition, the preparation of these collective states does not require a very special excitation process: one has simply

to prepare uncorrelated atoms in a statistical mixing of the two upper states. We have indeed shown that, when uncorrelated atoms are involved in a cooperative process, their evolution depends essentially on the 'amount' of statistical mixing, which determines the symmetry properties of the collective atomic states with respect to the permutations of cooperating atoms. The last step to a realistic situation is the generalisation of the present results to the case of any $j \rightarrow j-1$ transition between degenerate levels (Pillet 1982): this will be described in a following paper (IV), together with the excitation process and the other conditions required for the observation of subradiance. We have already mentioned here that experimental situations in which subradiance could be observed are not so easy to find. A detailed analysis of all the superradiance experiments which are described in the literature (Gross *et al* 1976, 1978, 1979, Flusberg *et al* 1976, Vrehen *et al* 1977, Rosenberger *et al* 1977, Crubellier *et al* 1978, 1982, Okada *et al* 1978, Marek 1979, Bréchignac and Cahuzac 1979, Cahuzac *et al* 1979, Gounand *et al* 1979) shows that the symmetry properties of the initial state never allowed subradiance, except in the HF experiments (Skribanowitz *et al* 1973), when the excitation used the molecular P branch: this latter case will be analysed in paper IV.

The three-level configuration provides a typical example in which subradiance is due both to the partially antisymmetrical nature and to the conservation of the symmetry properties of the collective atomic state. Subradiance can also occur in quite different ways. First, as already mentioned (Crubellier 1977), a supplementary interatomic interference appears when the level system contains a degenerate transition (i.e. several atomic transitions which are resonant with the same mode of the electromagnetic field). When destructive, this interference inhibits the spontaneous emission in the cooperative field modes. This type of subradiance, which has already been discussed (Gross and Haroche 1982) will be studied in the paper II. Its features are found to be substantially different from those described in the present paper. The 'limited superradiance' which is predicted for small samples (Friedberg *et al* 1972, Friedberg and Hartmann 1974a, b) will be analysed in terms of symmetry properties in paper III. It appears to be a third type of subradiance, in which the destructive interatomic interference is due to a sudden spontaneous symmetry breaking. The last paper of the sequence will be mainly devoted to the realistic case of a $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ atomic cascade. It will give an illustration of the general results of papers I and II and will also provide an interpretation of our experimental results (Crubellier *et al* 1985, Pavolini *et al* 1985), in which the first experimental evidence for subradiance has been obtained on a $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ cascade of atomic gallium.

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4.2 Configuration à niveaux dégénérés

Dans une étude qui veut aboutir à une mise en évidence de la subradiance, on est très naturellement amené à étudier le cas des configurations à niveaux dégénérés et à chercher les conditions expérimentales nécessaires pour que l'évolution coopérative prenne fin dans un état collectif subradiant.

4.2.1 Transitions entre deux niveaux dégénérés

On s'intéresse ici au cas d'une transition dipolaire électrique entre deux niveaux de moments angulaires j et j' quelconques, et à l'éventualité d'un phénomène de subradiance dû aux propriétés de symétrie de l'état collectif. On peut montrer, et ce sera fait dans le chapitre 5, que, à un état excité initial qui est un mélange statistique complet à poids égaux de tous les sous-niveaux du niveau supérieur j correspond un type de symétrie $\{\frac{N}{2j+1}, \frac{N}{2j+1}, ..., \frac{N}{2j+1}\}$. L'état collectif est alors représenté, comme on a vu dans le paragraphe précédent, par un tableau d'Young composé de $N/(2j+1)$ lignes et $(2j+1)$ colonnes. Par analogie avec les "paires antisymétriques" que l'on considère dans le cas de la configuration à trois niveaux quand les deux niveaux supérieurs ont été également peuplés initialement dans un mélange statistique, on peut visualiser l'état collectif en lui associant $N/(2j+1)$ fonctions complètement antisymétriques $|l, m, ..., n>_{AS}$ de $2j+1$ atomes dans $2j+1$ états monoatomiques $l, m, ..., n$. Au début de l'évolution, chaque fonction contient les $2j+1$ états supérieurs, c'est à dire les $2j+1$ sous-niveaux Zeeman du niveau supérieur. Si le niveau j' possède un moment angulaire égal ou plus grand que j, à la fin de l'évolution les $2j+1$ sous-niveaux du niveau supérieur seront remplacés par $2j+1$ parmi les $2j'+1 \geq 2j+1$ sous-niveaux du niveau inférieur. Si, en revanche, le niveau inférieur

a un moment angulaire plus petit que celui du niveau supérieur, c'est à dire, pour une transition dipolaire électrique, si $j' = j - 1$, alors la fonction contiendra les $2j' + 1 = 2j - 1$ états qui sont les sous–niveaux du niveau inférieur $j - 1$, mais aussi deux états qui sont encore des sous–niveaux du niveau supérieur j. Ceci équivaut à dire que deux atomes de l'ensemble des $2j + 1$ atomes sont restés dans l'état excité: l'état final est subradiant. On calcule facilement le taux minimal de subradiance correspondant: il vaut $\tau = \frac{2}{2j+1}$.

On peut encore simplifier la description qualitative de l'évolution coopérative du système lorsqu'on choisit comme axe de quantification l'axe de symétrie du cylindre. L'émission superradiante, qui a lieu dans la même direction, est alors polarisée σ_+ et σ_-. A l'instant pris comme origine des temps, c'est à dire l'instant où tous les $\mathcal{N}$ atomes se trouvent dans le niveau supérieur de la transition, $\mathcal{N}_a$ atomes sont dans les sous–niveaux du sous–système a, et $\mathcal{N}_b$ atomes dans les sous–niveaux du sous–système b. $\mathcal{N}_a$ et $\mathcal{N}_b$ sont évidemment conservés par l'action de l'hamiltonien, ce qui revient à dire que le nombre d'atomes dans chaque sous–système est conservé durant l'évolution. Suivant qu'ils sont initialement dans le sous–système a ou bien dans le sous–système b, les $\mathcal{N}$ atomes de chaque tranche évoluent alors à l'intérieur des deux classes, a et b, qui sont en général définies seulement de façon statistique. Les propriétés de symétrie de l'état collectif vis–à–vis du groupe $\mathcal{S}_{\mathcal{N}} = \mathcal{S}_{\mathcal{N}_a+\mathcal{N}_b}$ sont conservées. Cependant, on peut montrer (Pavolini et al 1985, Crubellier et al 1986) que les propriétés de symétrie significatives concernent en fait les groupes $\mathcal{S}_{\mathcal{N}_a}$ et $\mathcal{S}_{\mathcal{N}_b}$ qui sont associés aux deux classes d'atomes. De plus, les deux sous–systèmes sont, en première approximation, indépendants l'un de l'autre, et l'on pourra donc analyser séparément l'évolution dans les deux.

Les deux sous–systèmes sont en réalité couplés par une interférence entre les transitions Zeeman de même polarisation, interférence dont on va étudier dans le chapitre suivant l'influence sur l'évolution collective du système.

4.2.2 Cascades entre niveaux dégénérés

Si le système atomique est composé seulement des trois niveaux, le niveau à partir duquel a lieu l'excitation initiale et les deux niveaux j et $j-1$ liés par la transition dipolaire sur laquelle on veut étudier la subradiance, l'observation du phénomène est assez délicate à réaliser. Il s'agit en effet d'accomplir une mesure simultanée du nombre d'atomes initialement excités et du nombre de photons émis (ou du nombre d'atomes desexcités). C'est ainsi qu'on est amené à compliquer davantage la configuration de niveaux et, plus précisement, à étudier le cas de configurations en cascade $j \to j-1 \to j'$. La présence du niveau dégénéré j' que, pour simplifier l'exposition du problème, on suppose ici supérieur ou égal à $j-1$, c'est à dire soit $j' = j$, soit $j' = j-1$, bouleverse complètement la situation, lorsqu'un phénomène de subradiance est prévu sur la transition $j \to j-1$ supérieure de la cascade. En effet, supposons que, à cause de l'antisymétrie partielle de l'état collectif, l'émission d'une impulsion superradiante sur la transition $j \to j-1$ ait amené spontanément le système dans un état subradiant, c'est à dire un état où l'émission sur cette transition ne peut plus continuer alors qu'une certaine proportion d'atomes se trouve encore dans le niveau supérieur j. La présence du niveau inférieur j' permet néanmoins de continuer l'évolution par l'émission d'une impulsion sur la deuxième transition $j-1 \to j'$, émission qui vide complètement le niveau intermédiaire $j-1$ puisqu'on a choisit $j' \geq j-1$. L'état collectif contient alors seulement des états monoatomiques qui sont sous–niveaux Zeeman des deux niveaux

j et j', niveaux qui ne sont pas reliés par une transition permise. Cet état, bien que partiellement antisymétrique, n'est pas subradiant, et l'évolution peut continuer par l'émission d'une deuxième impulsion superradiante sur la première transition $j \rightarrow j-1$. Cette impulsion, que nous appellons écho de subradiance, est fabriquée par les photons émis lors de la désexcitation des atomes qui étaient restés dans le niveau j après la première impulsion. Elle prouve ainsi l'existence d'un état intermédiaire du système où l'émission sur la transition $j \rightarrow j-1$ était coopérativement inhibée. L'observation expérimentale de deux impulsions successives sur la même transition, la deuxième d'entre elles étant émise après une impulsion sur la transition inférieure, témoigne alors de l'état subradiant par lequel le système est forcément passé.

5. INTERFERENCE INTERATOMIQUE DUE AUX TRANSITIONS DEGENEREES

Les transitions entre niveaux dégénérés sont le cas habituel dans les expériences réelles. En émission spontanée coopérative ces transitions présentent en général un deuxième type d'interférence. En effet, si on choisit l'axe de quantification le long de la direction de propagation de la lumière, l'émission superradiante, qui a lieu dans la même direction, ne peut pas être polarisée π. Alors une transition entre deux niveaux dégénérés de moments angulaires respectifs j et j' comprend $(j+j')$ transitions correspondant aux deux polarisations σ_+ et σ_- suivant lesquelles a lieu l'émission. Dans ce chapitre on étudie l'influence sur l'état final du système de l'interférence due à la présence des deux transitions dégénérées correspondant aux deux polarisations, et notamment la possibilité que cette interférence soit destructive et que l'état final soit donc subradiant.

5.1: Configurations impliquant une transition dégénérée

Dans l'article présenté ci–après, "Superradiance and subradiance:II. Atomic systems with degenerate transitions", le cas d'atomes à quatre niveaux avec une transition dégénérée, qui représente un exemple typique simple, est étudié en premier (§ 2). On considère d'abord le cas de deux atomes qui se trouvent initialement dans chacun des deux états excités, en supposant satisfaite la condition de coopérativité (eq. 1). Les deux atomes sont alors indiscernables vis–à–vis du champ émis et interfèrent entre eux, alors qu'ils seraient complètement indépendants si la transition n'était pas dégénérée. Dans le cas général de N atomes, l'interférence entre les atomes a lieu à la fois sur chaque transition séparément et entre deux transitions. Celle qui a lieu entre deux tran-

sitions différentes est soumise aux mêmes conditions (eq. 12) et limitations que celle qui a lieu sur une même transition, et qui est étudiée dans le chapitre 4. Son caractère peut par ailleurs être constructif ou bien destructif: la probabilité que le système atteigne un état où l'émission est coopérativement inhibée est, en général, non nulle, sauf si les deux probabilités de transition sont égales. On appelle subradiance "Zeeman" la subradiance due à cette sorte d'interférence. L'équation pilote qu'on peut écrire pour le système macroscopique de N atomes devient toutefois trop compliquée à résoudre dès que N depasse quelques unités, et l'on trouve des résultats quantitatifs dans le modèle semi–classique (§ 2.4). On définit les deux angles de Bloch correspondant aux deux transitions. Ces deux angles, à cause de la dégénérescence de la transition, restent proportionnels l'un à l'autre (eq.2.19). De plus, on peut exprimer à travers une équation (eq.2.21) la condition que l'évolution s'achève soit quand toutes les cohérences optiques sont nulles, ce qui implique une désexcitation complète des atomes, soit quand il y a une interférence complètement destructive entre les champs émis sur les deux transitions. On peut ainsi calculer sans ambiguité l'état final du système. En particulier, si les deux probabilités de transition sont différentes, les atomes ne peuvent pas être tous désexcités et l'état final est subradiant. A l'aide des équations trouvées pour les deux angles de Bloch, on peut calculer le taux de subradiance pour les différentes valeurs des probabilités de transition et des populations initiales. Les resolutions numériques des équations de Bloch–Maxwell faites pour plusieurs valeurs différentes de ces grandeurs sont en accord avec ces résultats théoriques.

Dans l'article présenté ci–après on considère ensuite les configurations possibles lorsque les deux transitions dégénérées ont, en plus, un

niveau commun (§ 3). Les deux configurations correspondant à un niveau supérieur (configuration "Λ") ou inférieur (configuration "V") commun entre les deux transitions sont quelque peu fictives: elles pourraient être réalisées seulement par un système atomique dont un niveau aurait une structure fine ou hyperfine qui soit en même temps petite par rapport à l'inverse du temps caractéristique de l'émission superradiante, et grande par rapport à la largeur spectrale du processus d'excitation. De plus, pour ces deux cas on trouve (§ 3.2) que le problème se simplifie en utilisant une base opportune d'états: l'évolution de la collection d'atomes est alors calculée de la même façon que pour un système à deux niveaux. Ces deux configurations nous permettent néanmoins de montrer comment l'interférence, aussi bien interatomique que intratomique, entre les deux transitions influe sur l'émission coopérative et, pour la configuration "V", en particulier, sur le taux de subradiance d'une telle collection d'atomes à trois niveaux. Cependant, ceci est un peu artificiel dans la mesure où un changement de base fait disparaître une transition, si bien que cette interférence n'existe plus dans la nouvelle base.

Ceci n'est pas le cas pour la troisième configuration étudiée, où le niveau commun est, cette fois, le niveau intermédiaire entre les deux transitions dégénérées: le cas de la configuration en "cascade dégénérée" (§3.3), qui revêt un intérêt tout à fait remarquable. Une telle configuration est envisageable expérimentalement si, par exemple, le système atomique est mis en présence d'un champ externe qui modifie convenablement les fréquences atomiques. Contrairement aux deux cas précedents, le problème de l'émission spontanée coopérative ne se réduit pas simplement à un problème à deux niveaux. Cependant l'existence d'un invariant (eq.3.26) implique une condition restrictive spécifique

sur l'état final et, par conséquent, éventuellement, un type spécifique de subradiance. On examine en détail deux cas limites, où tous les atomes sont initialement soit dans le niveau supérieur (§ 3.3.3), soit dans le niveau intermédiaire (§ 3.3.4). On montre en particulier que les deux transitions de la cascade peuvent "battre" l'une avec l'autre, et donnent alors lieu à un nouveau type de battement superradiant. On trouve aussi que, si les atomes sont excités dans le niveau intermédiaire de la cascade, la superradiance à partir de ce niveau est inhibée de façon résonnante quand les fréquences des deux transitions deviennent égales; l'inhibition est complète et les atomes restent dans l'état intermédiaire si la transition la plus probable est celle qui va vers le niveau supérieur; autrement les atomes sont transférés aux états extrêmes de la cascade par émission d'une impulsion superradiante d'aire proportionnelle à la différence entre les deux probabilités de transition.

J. Phys. B: At. Mol. Phys. 19 (1986) 000–000. Printed in Great Britain

Superradiance and subradiance: II. Atomic systems with degenerate transitions

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Received 2 August 1985, in final form 24 October 1985

Abstract. This paper is the second of a theoretical study of subradiance, i.e. of the cooperative inhibition of spontaneous emission by a destructive interatomic interference, a phenomenon which has recently been observed. It is devoted to the particular case of atomic systems with degenerate transitions, i.e. with several transitions of the same frequency and the same polarisation. The interatomic interference which then occurs between the different transitions is responsible, when destructive, for a new sort of subradiance. The phenomenon is studied for a few typical level configurations, using both quantum-mechanical and semiclassical models. The first example concerns a four-level configuration with two transitions of the same frequency and polarisation. The other examples concern three-level configurations with a degenerate transition; in the case of the 'degenerate' cascade, new superradiant beats and subradiance effects are predicted.

1. Introduction

This paper is the second of a sequence for four papers devoted to the theoretical study of subradiance, i.e. the cooperative inhibition of spontaneous emission by a destructive interatomic interference, a phenomenon which has recently been observed (Pavolini *et al* 1985). In the first paper of the sequence (Crubellier *et al* 1985 to be referred to as I) we studied in great detail the cooperative spontaneous emission of three-level atoms in the 'V' configuration (two transitions sharing a common lower level). It was shown that in this typical case incomplete de-excitation is predicted for some specific initial collective states. This subradiance phenomenon was shown to be due to a destructive interatomic interference, which is allowed by the indistinguishability of the atoms with respect to the field they radiate, a property which is quite fundamental for cooperative effects: for pencil-shaped atomic samples, for example, the conditions for a local atomic indistinguishability are exactly identical to the cooperativity conditions. The constructive or destructive nature of the interatomic interference was then found to simply depend upon the symmetry properties with respect to the permutations of locally indistinguishable atoms of the collective atomic state. These latter properties were finally shown to depend, for uncorrelated atoms, on the amount of statistical mixing of the atomic state. These results allow one to qualitatively predict the evolution of the system through cooperative spontaneous emission whatever the initial state is and, in particular, to calculate the subradiance rate, i.e. the proportion of atoms which will remain in the two upper levels.

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0022-3700/86/000000+00$02.50

For other level configurations and, in particular, for the case of a $j \rightarrow j'$ transition between degenerate levels, an additional interatomic interference is expected when there is a degenerate transition (Crubellier 1977), i.e. several transitions with the same frequency and polarisation. If a photon corresponding to the degenerate transition is emitted, one can know neither which atom has emitted it nor on which particular transition it has actually been emitted. If the transition is only nearly degenerate, this type of interference explains the existence of superradiant beats (Vrehen *et al* 1977a, b, 1978, Marek 1979, Marek and Ryschka 1980, Ryschka and Marek 1981) which are of quite a different nature than usual 'single-atom' quantum beats and which exhibit frequencies corresponding to both initial- and final-level splitting. Doppler superradiant beats have also been observed (Gross *et al* 1978), providing clear experimental evidence for the interatomic inteference. A theoretical description of these superradiant beats has now been given, using various approaches (Raimond 1979, Gross 1980, Abraham and Bullough 1980, Agarwal *et al* 1980, Leonardi and Vaglica 1981, 1985, Leonardi *et al* 1982, Gross and Haroche 1982). In the fully degenerate case, which we consider here, the interference between the different transitions corresponding to the same degenerate transition can be either constructive or destructive. A constructive interference will be responsible for the triggering of the emission on the other transitions by the emission on the must probable one. A destructive interference will lead to a new sort of subradiance, the specific features of which are found to be rather different from those previously studied (I). The new kind of interatomic interference is still allowed by the indistinguishability of the atoms, as for two- and three-non-degenerate-level atoms. As a consequence, the conditions required for the existence of the interference and also the obstacles to the subradiance phenomenon are exactly the same as in I: they will not be discussed again here. However, since the interference concerns different transitions, its constructive or destructive nature no longer depends upon the symmetry properties of the collective atomic state with respect to the atomic permutations. The typical case of four-level atoms with a degenerate transition has already been discussed, in the semiclassical model (Hermann and Bullough 1978, Gross and Haroche 1982); the 'limited superradiance' effect which was predicted and interpreted by the latter authors as being due to a negative interference between the two transitions can of course be called subradiance too.

In this paper, we give a detailed theoretical study of the cooperative spontaneous emission of such four-level atoms (§ 2). This simple configuration allows one to point out the specific influence of the interatomic interference between different transitions and the properties of the corresponding type of subradiance. Both fully quantum-mechanical and semiclassical models are used and the role of the symmetry properties under atomic permutations of the collective state is thoroughly discussed. These results provide a first approach to the problem of the 'Zeeman subradiance' which is expected in $j \rightarrow j'$ transitions between degenerate levels because all Zeeman components of the same polarisation correspond to the same degenerate transition (Pillet 1982). The different cases of three-level configurations ('Λ' configuration, 'V' configuration and cascade configuration) are also studied assuming that the two transitions which are already coupled by the common level they share (Crubellier and Schweighofer 1978) correspond to the same degenerate transition. In the case of the 'degenerate' cascade, cooperative spontaneous emission is expected to exhibit new and striking subradiance effects. It is also briefly shown, for the different level configurations under study, how subradiance disappears and superradiant beats appear when the degeneracy of the transition is slightly removed.

2. Four-level atoms with a degenerate transition

2.1. Two-atom case

The four-level configuration under study is represented in figure 1. The two upper states, $|1\rangle$ and $|2\rangle$ are respectively connected to the lower states $|1'\rangle$ and $|2'\rangle$ by two transitions corresponding to a single degenerate transition, i.e. two transitions with the same frequency ω_0 and the same polarisation e. The matrix elements of the atomic dipole operator $\mathbf{d}$ are given by

$$\begin{aligned} d_{11'} &= \langle 1|\mathbf{d}\cdot e|1'\rangle = \alpha_1 d \\ d_{22'} &= \langle 2|\mathbf{d}\cdot e|2'\rangle = \alpha_2 d \end{aligned} \tag{2.1}$$

where α_1 and α_2 are dimensionless coefficients which are taken to be real and which verify $\alpha_1^2+\alpha_2^2=1$. The transition probabilities per unit time, Γ_1 and Γ_2, can then be written as

$$\begin{aligned} \Gamma_1 &= \alpha_1^2\Gamma \\ \Gamma_2 &= \alpha_2^2\Gamma \end{aligned} \tag{2.2}$$

with $\Gamma=\Gamma_1+\Gamma_2$.

We now consider two such four-level atoms, initially in the state

$$|\Psi(0)\rangle = |1,2\rangle = |1\rangle_1|2\rangle_2 \tag{2.3}$$

which corresponds to atom 1 in state $|1\rangle$ and atom 2 in state $|2\rangle$. We assume that the interatomic distance is much smaller than the wavelength of the emitted field. When a photon is emitted, it is thus impossible to know which atom has emitted it; this means that interatomic interference occurs and that spontaneous emission is cooperative (Stephen 1964). One notices that this is uniquely due to the degeneracy of the transition and that the two atoms would radiate quite independently if the degeneracy was removed. The master equation describing this evolution is given by (Crubellier 1977 equation (1'))

$$\dot{\rho}(t) = \tfrac{1}{2}\Gamma[(R^-, \rho(t)R^+) + \text{HC}] \tag{2.4}$$

where

$$R^+ = \sum_{i=1,2} (\alpha_1|1\rangle_{ii}\langle 1'| + \alpha_2|2\rangle_{ii}\langle 2'|) \tag{2.5}$$

and R^- is the Hermitian conjugate of R^+. The system evolves inside the state space spanned by the basis set

$$\{|1,2\rangle, |\text{SR}\rangle, |\text{sr}\rangle, |1',2'\rangle\}$$

with

$$\begin{aligned} |\text{SR}\rangle &= \alpha_1|1,2'\rangle + \alpha_2|1',2\rangle \\ |\text{sr}\rangle &= \alpha_2|1,2'\rangle - \alpha_1|1',2\rangle \end{aligned} \tag{2.6}$$

which diagonalises the R^+R^- operator. One has in particular

$$\begin{aligned} R^+R^-|\text{SR}\rangle &= |\text{SR}\rangle \\ R^+R^-|\text{sr}\rangle &= 0. \end{aligned} \tag{2.7}$$

The value of the decay rate of the system in the state $|\text{SR}\rangle$, $\Gamma=\Gamma_1+\Gamma_2$, is due to a fully constructive interference. On the contrary, the interatomic interference is fully destructive in the state $|\text{sr}\rangle$, which is subradiant and has a zero decay rate.

Superradiant and subradiant states do not simply correspond to symmetrical and antisymmetrical states, as was the case for two two-level atoms or for two three-level atoms in the 'V' configuration (I). Contrary to these latter cases, the interatomic interference occurs indeed on two different transitions, which have different probabilities. The evolution of the density matrix elements is ruled by (see figure 2)

$$\begin{aligned}
&\langle 1,2|\dot{\rho}(t)|1,2\rangle = -\Gamma\langle 1,2|\rho(t)|1,2\rangle \\
&\langle \mathrm{SR}|\dot{\rho}(t)|\mathrm{SR}\rangle = -\Gamma\langle \mathrm{SR}|\rho(t)|\mathrm{SR}\rangle + (4\Gamma_1\Gamma_2/\Gamma)\langle 1,2|\rho(t)|1,2\rangle \\
&\langle \mathrm{sr}|\dot{\rho}(t)|\mathrm{sr}\rangle = [(\Gamma_1-\Gamma_2)^2/\Gamma]\langle 1,2|\rho(t)|1,2\rangle \\
&\langle 1',2'|\dot{\rho}(t)|1',2'\rangle = \Gamma\langle \mathrm{SR}|\rho(t)|\mathrm{SR}\rangle.
\end{aligned} \tag{2.8}$$

The probability of the system being in the subradiant state $|\mathrm{sr}\rangle$ at the end of the evolution is found to be equal to $(\Gamma_1-\Gamma_2)^2/\Gamma^2$; it is different from zero provided that $\Gamma_1 \neq \Gamma_2$. It is to be noted that the chosen initial state (2.3) is neither symmetrical nor antisymmetrical. The same results would in fact be obtained in both latter cases; the evolution of the system does not depend on the symmetry of the initial state, but rather on the ratio of the two transition probabilities.

The solution of the master equation (2.4) becomes quickly inextricable as the number of atoms increases. For N indistinguishable atoms, two general results can, however, be obtained. Firstly, one can show that, contrary to the case of the 'V' level configuration (I), the evolution starting from a fully excited collective state does not depend on the symmetry properties of this state. Secondly, an explicit expression of the multi-atom subradiant states can be exhibited and subradiance is shown to be generally predicted.

2.2. Subradiance and symmetry properties

In the plane-wave approximation, a pencil-shaped sample of four-level atoms (figure 1) can be considered as formed by successive slices with thickness smaller than the wavelength of the atomic transitions. The N atoms of a slice are indistinguishable since the field they radiate and the atom-field Hamiltonian are invariant under the local atomic permutations (I). As the two transitions do not share any common level, the N atoms of a slice form two classes, one corresponding to the atoms which are in states $|1\rangle$ and $|1'\rangle$, the other one corresponding to the atoms which are in states $|2\rangle$ and $|2'\rangle$. No atom exchange between the two classes is allowed during the evolution of the

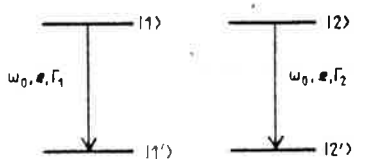

Figure 1. Four-level configuration with a degenerate transition.

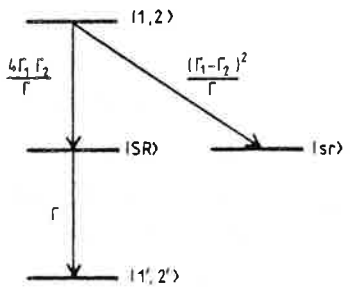

Figure 2. Collective energy level diagram for two four-level atoms with a degenerate transition, showing the collective transition probabilities.

system; the class each atom belongs to is fixed (at least statistically) by the initial atomic state, and the numbers of locally indistinguishable atoms in the two classes, N_1 and N_2 (with $N_1+N_2=N$) remain constant. Because of the degeneracy of the transition, the two classes of atoms do not evolve independently. The interatomic interference occurs indeed not only among atoms of the same class but also between atoms of different classes. However, as in the two-atom case, the constructive or destructive nature of the 'interclass' interference is not simply fixed by the symmetry properties of the collective atomic state under the atomic permutations between the two classes. Moreover, as it will be shown hereafter, these latter symmetry properties have little physical interest and the only relevant symmetry properties are those with respect to the atomic permutations inside each class.

The existence of the two classes is related to the invariance of the atom-field Hamiltonian under the direct product of the local permutation groups S_{N_1} and S_{N_2} associated with the two classes. The two corresponding local symmetry types (I) of the collective atomic state, χ_1 and χ_2, are thus conserved during the whole evolution of the system. The evolution of the physical quantities which are diagonal with respect to χ_1 and χ_2, such as, for example, the populations of the different levels and the optical coherences associated with the two transitions, is not coupled with the evolution of the off-diagonal quantities. These latter ones are also off-diagonal with respect to the two two-level systems 1 and 2 and they do not influence the cooperative spontaneous emission. The symmetry properties under the interclass permutations, which are irrelevant to the χ_1 and χ_2 diagonal quantities, can thus be ignored. In particular, if all atoms are initially in the upper states $|1\rangle$ and $|2\rangle$, the cooperative emission of the atoms is fully determined by the S_{N_1} and S_{N_2} symmetry types, which are necessarily, in this particular case, the fully symmetrical ones, $\{N_1\}$ and $\{N_2\}$.

The symmetry properties of the initial collective state with respect to the local permutation groups S_N influence only the evolution of the coherences between the two two-level systems. In particular, the subradiance rate does not depend on these symmetry properties but only on the two numbers N_1 and N_2.

2.3. Many-atom subradiant states

The existence of subradiance in the case of four-level atoms can be demonstrated by giving a general explicit expression for the subradiant states of N such indistinguishable atoms. The master equation describing the evolution of these atoms is given by equations (2.4) and (2.5), except that the sum of equation (2.5) runs now from 1 to N. According to the $S_{N_1}\times S_{N_2}$ invariance of the atom-field Hamiltonian, a suitable basis set of collective states is provided by products of two Dicke's states $|grm\rangle$ (Dicke 1954) corresponding to the two two-level systems. In the particular case of atoms initially in states $|1\rangle$ and $|2\rangle$ only, one has just to consider products of fully symmetrical Dicke's states which can be written $|N_1/2m_1\rangle\otimes|N_2/2m_2\rangle$, where the index g, which is useless in this case, is omitted and where m_1 (or m_2) is the half difference between the populations of levels $|1\rangle$ and $|1'\rangle$ (or $|2\rangle$ and $|2'\rangle$). We now consider the following non-normalised collective states

$$|s(m)\rangle=\sum_{m_1,m_2}-1^{(N_1+N_2)/2+m_1}\alpha_1^{N_2/2+m_2}\alpha_2^{N_1/2+m_1}\times\left(\frac{(N_1/2-m_1)!(N_2/2-m_2)!}{(N_1/2+m_1)!(N_2/2+m_2)!}\right)^{1/2}|N_1/2m_1\rangle\otimes|N_2/2m_2\rangle \tag{2.9}$$

where the sum runs over the m_1 and m_2 values verifying $m_1 + m_2 = m$, with $-N/2 \leqslant m \leqslant -|(N_1 - N_2)/2|$. These states, whose energy is given by $m\hbar\omega_0$, verify

$$R^-|s(m)\rangle = 0 \tag{2.10}$$

and they are therefore subradiant, except if $m = -N/2$, which corresponds to the fully de-excited collective state. Any state with all atoms in states $|1\rangle$ and $2\rangle$ is connected to the $|s(m)\rangle$ states by a chain of transition matrix elements which are generally different from zero, unless $\Gamma_1 = \Gamma_2$ (1). At the end of the evolution, these metastable collective states are thus populated and the subradiance rate is given by

$$\tau = \frac{1}{N}\sum (N/2 - m)\langle s(m)|\rho(\infty)|s(m)\rangle \tag{2.11}$$

where the sum runs over the values of m such that $-N/2 \leqslant m \leqslant -|N_1 - N_2|/2$. It is expected to be non-zero, except if $\Gamma_1 = \Gamma_2$. However, for N larger than a few units, the solution of the master equation (2.4) becomes inextricable and, in order to find quantitative results, we will now make use of the semiclassical model.

2.4. Semiclassical model

In the semi-classical model, using the plane-wave approximation (McGillivray and Feld 1976), the evolution of a pencil-shaped sample of four-level atoms (figure 1) is given by the following Bloch–Maxwell equations (Raimond 1979, Abraham and Bullough 1980, Pillet 1982, Gross and Haroche 1982)

$$\begin{aligned}
&\frac{\partial}{\partial t'}(Q_{11} - Q_{1'1'}) = \frac{2id\alpha_1}{\hbar}(\varepsilon^* Q_{11'} - \varepsilon Q_{11'}^*)\\
&\frac{\partial}{\partial t'}(Q_{22} - Q_{2'2'}) = \frac{2id\alpha_2}{\hbar}(\varepsilon^* Q_{22'} - \varepsilon Q_{22'}^*)\\
&\frac{\partial}{\partial t'}Q_{11'} = \frac{id\alpha_1}{\hbar}\varepsilon(Q_{11} - Q_{1'1'})\\
&\frac{\partial}{\partial t'}Q_{22'} = \frac{id\alpha_2}{\hbar}\varepsilon(Q_{22} - Q_{2'2'})\\
&\frac{\partial}{\partial z}\varepsilon = -\frac{ikd}{2\varepsilon_0}(\alpha_1 Q_{11'} + \alpha_2 Q_{22'}).
\end{aligned} \tag{2.12}$$

In these equations, the Q_{ii} (with $i = 1, 1', 2, 2'$) are the populations of the different levels, whereas ε, $Q_{11'}$ and $Q_{22'}$ are the slowly varying envelopes of the field and of the optical coherences associated with the two transitions. Time t' is the retarded time, $t' = t - z/c$ and, for the sake of simplicity, equations (2.12) are written only for the wave packet travelling in the forward direction of the cylinder axis. One notices that equations (2.12) do not contain all the atomic coherences of the system. The omitted coherences are off-diagonal with respect to the local $S_{N_1} \times S_{N_2}$ invariance groups of the Hamiltonian (see § 2.2) and, as expected, their evolution equations are not coupled with equations (2.12). As these coherences do not influence any interesting physical quantity, they will be ignored.

Initially, the atoms are excited either in state $|1\rangle$ or in state $|2\rangle$ and they are assumed to be uncorrelated, with ρ_{11} and ρ_{22} as diagonal monatomic density matrix elements

(as shown before, the initial value of the coherence ρ_{12} does not need to be precise here). The initial conditions for equations (2.12) are then

$$\begin{aligned} &Q_{11}(z,0) = n\rho_{11} \\ &Q_{22}(z,0) = n\rho_{22} \\ &Q_{11'}(z,0) = 0 \\ &Q_{22'}(z,0) = 0 \\ &\varepsilon(0,0) = \varepsilon_0 \end{aligned} \tag{2.13}$$

where n is the atomic density and ε_0 a small but non-zero value of the electromagnetic field simulating the vacuum fluctuations which allow the superradiant emission to start (Pillet 1977, Haake *et al* 1979, Polder *et al* 1979).

Two Bloch vectors can be associated with the two transitions; both lengths are shown to be separately conserved:

$$\begin{aligned} &(Q_{11} - Q_{11'})^2 + 4|Q_{11'}|^2 = n^2\rho_{11}^2 \\ &(Q_{22} - Q_{2'2'})^2 + 4|Q_{22'}|^2 = n^2\rho_{22}^2. \end{aligned} \tag{2.14}$$

This conservation property is closely related to the local $S_{N_1} \times S_{N_2}$ invariance of the Hamiltonian. Using the notation of I, the Casimir invariant operator (Judd 1973) of a local group $SU(2) \times S_{N_1}$, for example, can be written

$$\begin{aligned} W_1 &= P_{11}^2 + P_{1'1'}^2 + P_{11'}P_{11'} + P_{11'}P_{11'} \\ &= \tfrac{1}{2}(P_{11} + P_{1'1'})^2 + \tfrac{1}{2}(P_{11} - P_{1'1'})^2 + P_{11'}P_{11'} + P_{11'}P_{11'} \end{aligned} \tag{2.15}$$

where all operators correspond to the considered atomic slice and where

$$P_{ij} = \sum_{\alpha=1,N_1} |i\rangle_{\alpha\alpha}\langle j|. \tag{2.16}$$

Equations (2.14) thus appear to be the semiclassical analogues of the conservation equations for the mean values of the Casimir invariants W_1 and W_2 of the local $SU(2) \times S_{N_1}$ and $SU(2) \times S_{N_2}$ invariance groups. These equations allow one to introduce two Block angles, $\theta_1(z, t')$ and $\theta_2(z, t')$, which are defined by

$$\begin{aligned} &Q_{11} - Q_{1'1'} = n\rho_{11} \cos\theta_1 \\ &Q_{22} - Q_{2'2'} = n\rho_{22} \cos\theta_2 \\ &Q_{11'} = \mathrm{i}n\rho_{11} \sin\theta_1/2 \\ &Q'_{22} = \mathrm{i}n\rho_{22} \sin\theta_2/2. \end{aligned} \tag{2.17}$$

These angles obey the following equations

$$\begin{aligned} &\frac{\partial}{\partial t'}\theta_1 = \frac{2d\alpha_1}{\hbar}\varepsilon \\ &\frac{\partial}{\partial t'}\theta_1 = \frac{2d\alpha_2}{\hbar}\varepsilon \end{aligned} \tag{2.18}$$

where the field ε is taken to be real, without any loss of generality. Since, from equations (2.17) and (2.13), one has $\theta_1(z, 0) = \theta_2(z, 0) = 0$, θ_1 and θ_2 remain proportional everywhere and at any time:

$$\frac{\theta_1(z, t')}{\alpha_1} = \frac{\theta_2(z, t')}{\alpha_2}. \tag{2.19}$$

At the end of the evolution, the field ε and its derivatives must be equal to zero everywhere and necessarily (Gross and Haroche 1982)

$$\alpha_1 Q_{11'}(z, \infty) + \alpha_2 Q_{22'}(z, \infty) = 0. \tag{2.20}$$

This equation expresses that the emission stops either when all optical coherences are zero, which implies a complete de-excitation of the atoms, or when there exists a fully destructive interference between the fields emitted on the two transitions. This equation can also be written as

$$\alpha_1 \rho_{11} \sin(\theta_1(z, \infty)) + \alpha_2 \rho_{22} \sin(\theta_2(z, \infty)) = 0 \tag{2.21}$$

and the final state of the collective evolution can be unambiguously calculated from equations (2.19) and (2.21). If $|\alpha_1| \neq |\alpha_2|$, i.e. if the transition probabilities Γ_1 and Γ_2 are different, one cannot have θ_1 and θ_2 simultaneously equal to π and thus the atoms cannot be all de-excited†. The final state of the system is therefore subradiant, because of the fully destructive interatomic interference described by equation (2.20).

The proportion τ of non-de-excited atoms is plotted against the initial population ρ_{11} of the state $|1\rangle$ in figure 3 for the particular case $\Gamma_1/\Gamma_2 = 3$, which corresponds to the case of a $\frac{3}{2} \rightarrow \frac{1}{2}$ transition, when only the $m = \frac{3}{2}$ and the $m = \frac{1}{2}$ Zeeman sublevels of the upper level are initially populated. As a matter of fact, as the superradiant light propagates along the atomic sample axis, it cannot be π polarised when the quantisation axis is taken along this direction. The atomic level configuration of figure 4 is thus similar to the one of figure 1.

Numerical semiclassical calculations of the whole evolution of the system, taking into account both forward and backward propagation directions of the superradiant light, have been realised for various values of Γ_1/Γ_2 and of ρ_{11}/ρ_{22}. The results are in agreement with the above simplified treatment and subradiance rates of typically

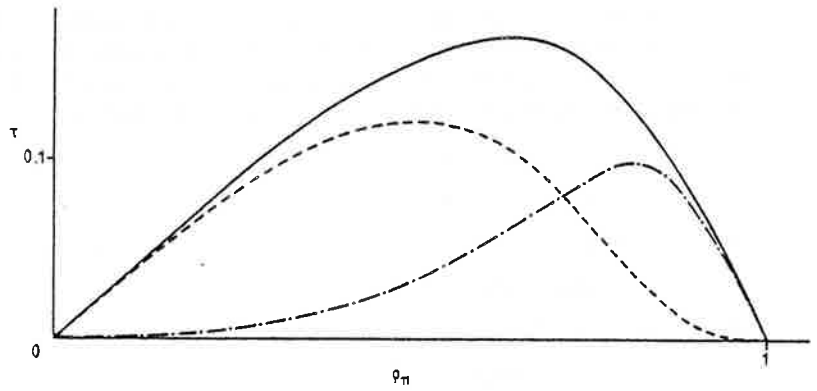

Figure 3. Subradiance rate τ plotted against initial population ρ_{11} of state $|1\rangle$ in the case $\Gamma_1/\Gamma_2 = 3$ (——); - - -, the proportion of atoms remaining the upper state $|1\rangle$; — · —, the proportion of atoms remaining in the upper state $|2\rangle$.

† The case $|\alpha_1| = |\alpha_2|$ is a very special one. R^+ and R^- are then infinitesimal operators of a SU(2) subgroup of the SU(4) 'cooperation group' (Crubellier 1977) which is associated to the N indistinguishable four-level atoms. The angular momentum associated with this SU(2) group is thus conserved. A collective state $|\psi\rangle$ with all atoms in the states $|1\rangle$ and $|2\rangle$ has an angular momentum $N/2$. As a subradiant state $|s(m)\rangle$ has an angular momentum $N/2$. As a subradiant state $|s(m)\rangle$ has an angular momentum equal to $|m|$, the de-excitation starting from the state $|\psi\rangle$ will necessarily lead to the fully de-excited state $|s(-N/2)\rangle$.

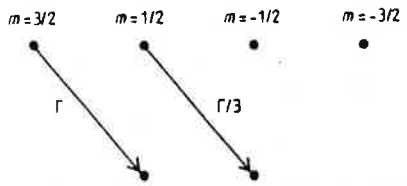

Figure 4. Diagram of the Zeeman sublevels of a $\frac{3}{2}\rightarrow\frac{1}{2}$ transition, showing the transitions on which superradiant emission occurs when only $m=\frac{3}{2}$ and $m=\frac{1}{2}$ sublevels are initially populated (the quantisation axis is assumed to be taken along the direction of propagation of the superradiant light).

10-20% are obtained. An example of semiclassical results, corresponding to the case of a $\frac{3}{2}\rightarrow\frac{1}{2}$ transition described just above (figure 4), is given in figure 5. The coupling between the two transitions which is induced by the interatomic interference on the different transitions and which is therefore due to the degeneracy of the transition, makes the emission on the $m=\frac{1}{2}\rightarrow m'=-\frac{1}{2}$ Zeeman transition occur almost simultaneously with (instead of three times later than) the emission on the $m=\frac{3}{2}\rightarrow m'=\frac{1}{2}$ transition (figure 5(b)). In addition one notices an important 'excitation exchange' between the two two-level systems, which makes the total populations of the Zeeman

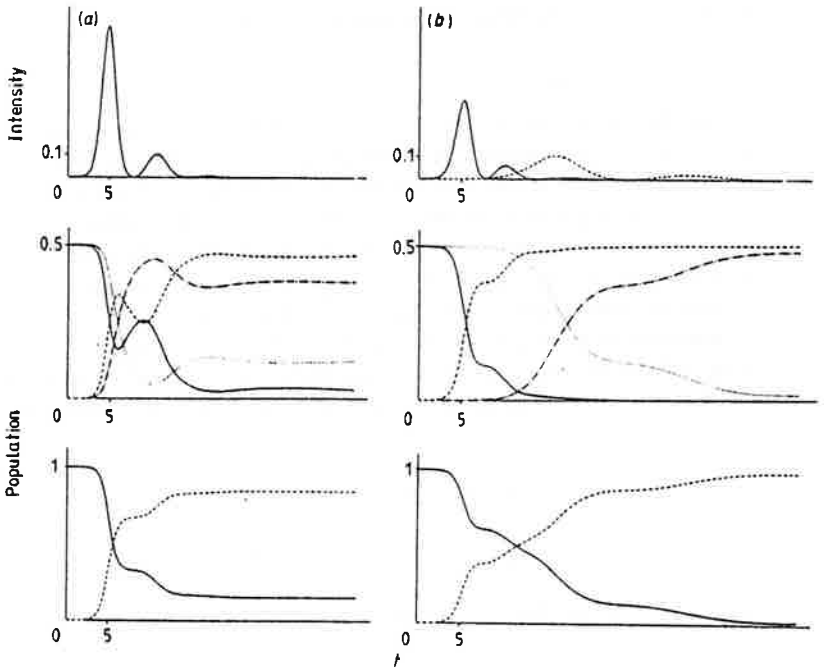

Figure 5. Semiclassical results corresponding to the configuration of figure 4, with equal initial populations of $m=\frac{3}{2}$ and $m=\frac{1}{2}$, upper Zeeman sublevels, in (a) the fully degenerate case and (b) when the level degeneracy is completely removed. The upper sets of curves represent the time evolution of the total radiated intensity in (a) and of the intensity radiated on each transition in (b) (——, $m=\frac{3}{2}\rightarrow m'=\frac{1}{2}$; - - -, $m=\frac{1}{2}\rightarrow m'=-\frac{1}{2}$). The intermediate sets of curves show the evolution of the total populations of the sublevels $m=\frac{3}{2}$ (——) and $m=\frac{1}{2}$ (· · · ·) of the upper level and of the sublevels $m=\frac{1}{2}$ (- - -) and $m=-\frac{1}{2}$ (— · —) of the lower level. The lower sets of curves represent the evolution of the total population of the upper $\frac{3}{2}$ level (——) and of the lower $\frac{1}{2}$ level (- - -).

sublevels oscillate (figure 5(*b*)), although the total populations of the upper and lower levels monotonously decrease and increase respectively, as expected in spontaneous emission. We have not numerically studied the fluctuations of the atom-field system in this case. We can, however, predict that the shape fluctuations of the emitted pulse will be rather similar to the fluctuations of superradiant emission of two-level systems: they are indeed caused by a single fluctuating field source (Polder 1979). Furthermore, the subradiance rate is always determined by equations (2.19) and (2.21) and does not thus fluctuate.

An indirect confirmation of these results can be found in previous experiments on atomic rubidium (Crubellier *et al* 1978, Pillet 1982). When excited from $5s\,\frac{1}{2}$ level to $6p\,\frac{3}{2}$ level with σ_+ polarised light, the atoms first radiate a single σ_- superradiant pulse, on the $6p\,\frac{3}{2} \rightarrow 6s\,\frac{1}{2}$ transition, followed by a σ_+ superradiant pulse emitted on the $6s\,\frac{1}{2} \rightarrow 5p\,\frac{3}{2}$ transition. The initial conditions for the first pulse are those of figure 5; in the subradiant state which is reached after the emission of this pulse, the $m = \frac{1}{2}$ sublevel of $6s\,\frac{1}{2}$ level is thus more populated than the $m = -\frac{1}{2}$ sublevel. Because of the competition and interference mechanisms which occur then during the emission of the second pulse, this small dissymmetry in the initial state explains quite well its practically pure σ_+ polarisation.

3. Three-level atoms with a degenerate transition

3.1. Two-atom case

In this section we study another case of two atomic transitions corresponding to the same degenerate transition; in addition, the two transitions share a common level. We thus consider the case of three-level atoms with two transitions sharing either the upper level ('Λ' configuration) or the lower level ('V' configuration) or the intermediate one (cascade configuration), when both transitions have the same frequency ω_0 and the same polarisation $\boldsymbol{e}$ (figure 6)†. The first two configurations might be experimentally realised by rather specific atomic-level systems, having in particular a small hyperfine (or fine) structure, the frequency splitting of which is small compared with the inverse of the characteristic time for superradiance but large compared with the frequency bandwidth of the excitation process. The 'degenerate'-cascade configuration, which

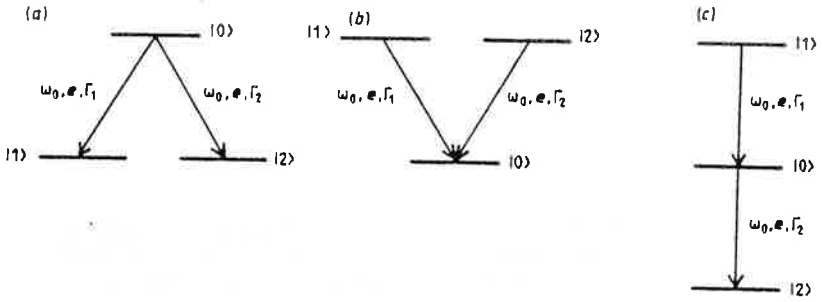

Figure 6. Three-level configurations with a degenerate transition: (*a*) 'Λ', (*b*) 'V' and (*c*) cascade configurations.

† The single-atom problem of the degenerate 'V' configuration and of the degenerate cascade has been discussed by Agarwal (1974).

contains two cascading transitions of the same frequency, would in fact be encountered in atomic-level systems only in the presence of an external field which suitably modifies the atomic frequencies†. In all cases, the matrix elements of the atomic dipole operator **d** are given by

$$\begin{aligned} d_{10} &= \langle 1|\mathbf{d}\cdot\boldsymbol{e}|0\rangle = \alpha_1 d \\ d_{20} &= \langle 2|\mathbf{d}\cdot\boldsymbol{e}|0\rangle = \alpha_2 d \end{aligned} \tag{3.1}$$

where, as in § 2, α_1 and α_2 are dimensionless real coefficients verifying $\alpha_1^2+\alpha_2^2=1$. The transition probabilities per unit time, Γ_1 and Γ_2, verify equation (2.2) with $\Gamma=\Gamma_1+\Gamma_2$.

We first consider two such three-level atoms and we assume that the interatomic distance is much smaller than the wavelength of the transition, so that interatomic interference may occur. Once a photon has been emitted, it is indeed impossible to know which atom has emitted it, or on which atomic transition. In this limit of fully cooperative emission, the evolution of the system is again ruled by equation (2.4), where

$$R^- = \sum_{i=1,2} \alpha_1 r_i^{-(1)} + \alpha_2 r_i^{-(2)} \tag{3.2}$$

$$r_i^{-(1)} = |1\rangle_{ii}\langle 0| \qquad r_i^{-(2)} = |2\rangle_{ii}\langle 0| \qquad \text{'}\Lambda\text{' configuration} \tag{3.3a}$$

$$r_i^{-(1)} = |0\rangle_{ii}\langle 1| \qquad r_i^{-(2)} = |0\rangle_{ii}\langle 2| \qquad \text{'V' configuration} \tag{3.3b}$$

$$r_i^{-(1)} = |0\rangle_{ii}\langle 1| \qquad r_i^{-(2)} = |2\rangle_{ii}\langle 0| \qquad \text{cascade configuration} \tag{3.3c}$$

and R^+ is the Hermitian conjugate of R^-. This evolution is most easily described by using a collective-state basis which diagonalises the R^+R^- operator. Rather different results will be found for the three particular level configurations under study.

3.1.1. 'Λ' *and* '*V*' *configurations.* For the 'Λ' and the 'V' level configuration (figures 6(*a*) and (*b*)), one first notices that states $|1\rangle$ and $|2\rangle$ are just a particular basis set for the states corresponding to the (upper or lower) degenerate level. In fact, because of the degeneracy of the transition, the choice of such a basis set is, as far as emission and absorption processes are concerned, fully arbitrary. We define two new basis states, $|a\rangle$ and $|b\rangle$, by

$$\begin{aligned} |a\rangle &= \alpha_1|1\rangle + \alpha_2|2\rangle \\ |b\rangle &= \alpha_2|1\rangle - \alpha_1|2\rangle. \end{aligned} \tag{3.4}$$

The two-atom states which diagonalise R^+R^- are then found to be the symmetrical and antisymmetrical states based on the monatomic states $|a\rangle$, $|b\rangle$ and $|0\rangle$, noted respectively:

$$\begin{aligned} |i,j\rangle_S &= (|i\rangle_1|j\rangle_2 + |j\rangle_1|i\rangle_2)/2^{1/2} \qquad (i\neq j) \\ |i,i\rangle_S &= |i\rangle_1|i\rangle_2 \\ |i,j\rangle_{AS} &= (|i\rangle_1|j\rangle_2 - |j\rangle_1|i\rangle_2)/2^{1/2} \qquad (i\neq j) \end{aligned} \tag{3.5}$$

† Such a configuration has actually been realised in the case of Na Rydberg atoms (Safinya *et al* 1981); resonant population transfers have been observed which can be explained by resonant Rydberg-atom–Rydberg-atom collisions (Gallagher *et al* 1982). These authors have found that the inhomogeneous broadening (due in particular to electric field inhomogeneity) in fact prevented the cooperative processes from occurring.

where i and j are a, b or 0. The evolution equations of the system cascading through cooperative emission among these collective states can be obtained from figure 7, which shows all collective transition probabilities.

For the 'Λ' level configuration (figure 7(a)), the evolution starting from the fully excited state $|0, 0\rangle_S$ is similar to the one of a pair of two-level atoms with a transition probability $\Gamma = \Gamma_1 + \Gamma_2$. For this '$\Lambda$' configuration, there exists only one subradiant state,

$$|0, a\rangle_{AS} = \alpha_1|0, 1\rangle_{AS} + \alpha_2|0, 2\rangle_{AS}. \tag{3.6}$$

A detailed analysis of the mean values of the different terms $r_i^{+(k)} r_j^{-(l)}$ of the R^+R^- operator shows that this state is characterised by a partially destructive interatomic interference on each transition separately and by a fully destructive interatomic interference between the two transitions, which explains its zero decay rate.

For the 'V' level configuration (figure 7(b)), there are four subradiant states, $|0, a\rangle_{AS}$, $|0, b\rangle_{AS}$, $|0, b\rangle_S$ and $|b, b\rangle_S$. The first two states would still be subradiant even if the transition was non-degenerate; their subradiance is due to their antisymmetrical nature

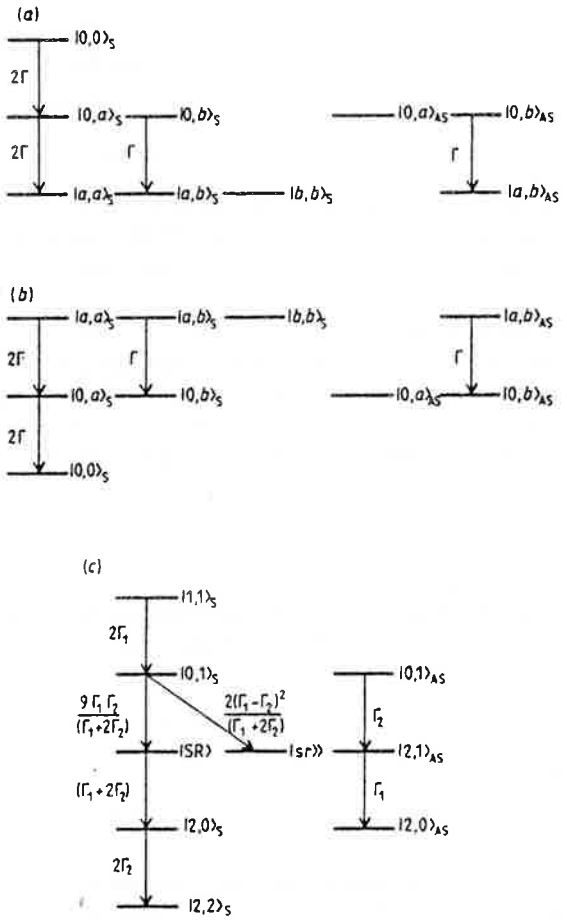

Figure 7. Collective energy level diagram for two three-level atoms with a degenerate transition, showing the collective transition probabilities. (a), (b) and (c) correspond to those in figure 6.

and thus, like in the non-degenerate 'V' configuration (I), it is due to a destructive interatomic interference on each transition separately. On the contrary, the other two subradiant states, which can be written as

$$\begin{aligned} |0, b\rangle_S &= \alpha_2|0, 1\rangle_S - \alpha_1|0, 2\rangle_S \\ |b, b\rangle_S &= \alpha_2^2|1, 1\rangle_S + \alpha_1^2|2, 2\rangle_S - 2^{1/2}\alpha_1\alpha_2|1, 2\rangle_S \end{aligned} \tag{3.7}$$

are specific to the degeneracy of the transition. The decay rate is zero because of a destructive interference between the two transitions, which appears to be an interatomic interference for the state $|0, b\rangle_S$, but which is intratomic as well as interatomic for the state $|b, b\rangle_S$; in this latter case, non-cooperative emission as well as cooperative emission are inhibited (because of the zero value of the matrix element of the atomic dipole operator between the states $|0\rangle$ and $|b\rangle$). It also appears in figure 7(b) that, for the 'V' configuration, the evolution starting from a fully excited two-atom state (with all the atoms either in state $|1\rangle$ or in state $|2\rangle$) will depend greatly on both the symmetry properties and the population ratio of the initial state. In particular, the subradiance rate may vary from 0 (no subradiance), for the initial state $|a, a\rangle_S$, to 1 (full subradiance, i.e. no cooperative de-excitation), for the initial state $|b, b\rangle_S$.

3.1.2. Cascade configuration. In the case of the cascade configuration (figure 6(c)) the choice of the basis set of monatomic states is of course not arbitrary. A two-atom state basis which diagonalises the R^+R^- operator has been found and is shown in figure 7(c) together with the collective transition probabilities; it contains in particular the following two states

$$\begin{aligned} |\mathrm{SR}\rangle &= (2^{1/2}\alpha_2|0, 0\rangle_S + \alpha_1|1, 2\rangle_S)/(\alpha_1^2 + 2\alpha_2^2)^{1/2} \\ |\mathrm{sr}\rangle &= (-\alpha_1|0, 0\rangle_S + 2^{1/2}\alpha|1, 2\rangle_S)/(\alpha_1^2 + 2\alpha_2^2)^{1/2}. \end{aligned} \tag{3.8}$$

These two states correspond respectively to fully constructive and fully destructive interatomic interferences between the two transitions. The first one has a decay rate equal to $\Gamma_1 + 2\Gamma_2$ and the second one is subradiant. Figure 7(c) indicates also the existence of another subradiant state, $|2, 0\rangle_{AS}$, which would still be subradiant without the transition degeneracy. The evolution of the two atoms will generally populate at least one of these two subradiant states (figure 7(c)). For example, if the two atoms are initially in the upper state $|1\rangle$ the probability for a photon to be trapped by the destructive interference is equal to $(\Gamma_1 - \Gamma_2)^2/[(2\Gamma_1 + \Gamma_2)(\Gamma_1 + 2\Gamma_2)]$; it is different from zero and subradiance is expected, except if $\Gamma_1 = \Gamma_2$. Incomplete de-excitation is also predicted when both atoms are initially in the intermediate state $|0\rangle$; the subradiant state $|\mathrm{sr}\rangle$ is then immediately populated and the proportion of trapped photons is $\Gamma_1/(\Gamma_1 + 2\Gamma_2)$.

This study of the elementary two-atom case already shows the intricate influence on the cooperative spontaneous emission of the two types of interatomic interference, either on each transition separately or between the two transitions. For the 'V' configuration one notices that, contrary to the case of the four-level configuration studied in the preceding section, the destructive interference between the two transitions can be intratomic as well as interatomic: the inhibition of spontaneous emission is then not a pure cooperative effect. As it will be shown hereafter, the existence of this intratomic interference reduces the problem of the spontaneous emission in the 'Λ' and the 'V' configurations to a two-level configuration problem, by simply making a suitable basis change. The effects of the interatomic and intratomic interferences

between the two transitions, however, appear, clearly in the old state basis; they will be briefly analysed, by comparing the results obtained in the degenerate and in the non-degenerate cases.

3.2. Degenerate 'Λ' and 'V' configurations: equivalence with two-level systems

For the 'Λ' and the 'V' configuration, the problem is greatly simplified if one uses the basis states $|a\rangle$ and $|b\rangle$ defined by equations (3.4). The matrix elements of the dipole operator correspond then to either a fully constructive or fully destructive intratomic interference between the 1↔0 and 2↔0 transitions; one has

$$\begin{aligned} d_{a0} &= \langle a|\mathbf{d}\cdot\boldsymbol{e}|0\rangle = (\alpha_1^2+\alpha_2^2)d = d \\ d_{b0} &= \langle b|\mathbf{d}\cdot\boldsymbol{e}|0\rangle = 0. \end{aligned} \tag{3.9}$$

The state $|b\rangle$ is therefore disconnected from the other two states (figure 8). It can neither be populated nor depopulated by emission or absorption processes. The

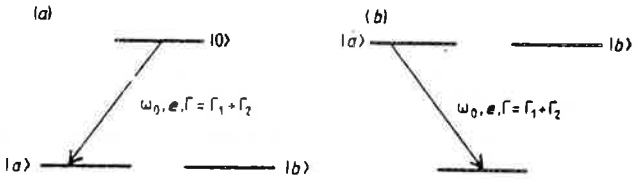

Figure 8. (*a*) Three-level 'Λ' and (*b*) three-level 'V' degenerate configurations in the basis where they reduce to two-level systems.

evolution of the collection of atoms through cooperative spontaneous emission can thus be calculated exactly as for two-level systems. The cooperation group of the system is a SU(2) group spanned by the R^+ and R^- operators (equations (3.3*a*) and (3.3*b*)) and the fully quantum-mechanical atom-field Hamiltonian commutes with the following operator

$$\begin{aligned} I_Q &= P_{bb} \\ &= \alpha_2^2 P_{11} + \alpha_1^2 P_{22} - \alpha_1\alpha_2(P_{12}+P_{21}) \end{aligned} \tag{3.10}$$

where, as in I, the P_{ij} operators are sums over locally undistinguishable atoms:

$$P_{ij} = \sum_{\alpha=1,N} |i\rangle_{\alpha\alpha}\langle j|. \tag{3.11}$$

The semiclassical equations keep invariant a quantity which is the semiclassical analogue of the operator (3.10):

$$I = \alpha_2^2 Q_{11} + \alpha_1^2 Q_{22} - \alpha_1\alpha_2(Q_{12}+Q_{21}) \tag{3.12}$$

where Q_{11} and Q_{22} are the populations of levels $|1\rangle$ and $|2\rangle$ and Q_{12} is the coherence between them (Pillet 1982). This conservation property which allows one to reduce the semiclassical equations to a set of two-level Bloch-Maxwell equations, implies a

specific restrictive condition on the final collective state reached by the atoms and thus, eventually, a specific kind of subradiance.

3.2.1. 'Λ' *configuration.* We consider here the case where all the atoms are initially in the upper state $|0\rangle$. They are then expected to simply fall to the lower state $|a\rangle$ by emitting a superradiant pulse which is the same as for two-level systems with a transition probability Γ and a complete initial-population inversion. Since the state $|b\rangle$ is never populated, the populations of the states $|1\rangle$ and $|2\rangle$ remain proportional to α_1^2 and α_2^2 during the whole evolution, exactly as they would in non-cooperative emission. It is interesting to compare the semiclassical results with those obtained for the same 'Λ' configuration but when the degeneracy of the transition is removed (figure 9). One

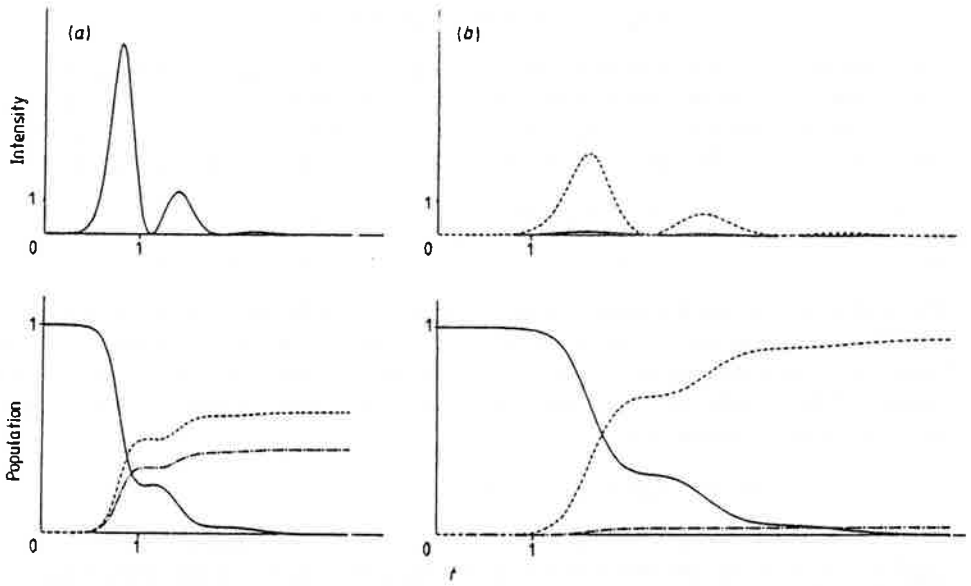

Figure 9. Comparison between semiclassical results corresponding to (a) degenerate and (b) non-degenerate three-level 'Λ' configurations. In both cases $\Gamma_2/\Gamma_1 = 1.44$ and all atoms are initially in the upper state. The upper sets of curves show the time evolution of the intensity radiated on both transitions in (a) and on the two transitions separately in (b) (——, $0 \rightarrow 1$; - - -, $0 \rightarrow 2$). The lower sets of curves show the time evolution of the total populations of the three states (— · —, 1; - - -, 2; ——, 0). The same arbitrary time and intensity units are used in all figures.

first remarks that the interference between the two transitions is essentially constructive since it accelerates and enhances the emission. In addition, whereas in the non-degenerate case emission on the less probable transition ($0 \rightarrow 2$) is almost completely inhibited, this quenching effect disappears in the degenerate case where, as expected, the populations of states $|1\rangle$ and $|2\rangle$ are always proportional to α_1^2 and α_2^2.

Semiclassical calculations have also been realised in the nearly degenerate case, when the energies of the two lower states differ by $\hbar\delta$, with $T = \pi/\delta$ larger than the characteristic time for superradiant emission. The transition can be considered as fully degenerate as long as T is twice as large as the delay time of the pulse. For larger δ

values, the pulse delay increases slightly and the quenching effect appears. In spite of this important effect (only 4% of the atoms decay in level 1), superradiant beats of period T with a contrast of about 50% are obtained when T becomes smaller than a fraction of the pulse delay time.

3.2.2. 'V' configuration. We consider that all atoms are initially excited either in state $|1\rangle$ or in state $|2\rangle$, the matrix elements of the monatomic density matrix being $\rho_{11}(0)$, $\rho_{22}(0)$, $\rho_{12}(0)$ and $\rho_{21}(0)$. In the state basis $\{|a\rangle, |b\rangle, |0\rangle\}$, the density matrix elements are given by:

$$\begin{aligned}
\rho_{aa}(0) &= \alpha_1^2\rho_{11}(0) + \alpha_2^2\rho_{22}(0) + \alpha_1\alpha_2(\rho_{12}(0) + \rho_{21}(0)) \\
\rho_{bb}(0) &= \alpha_2^2\rho_{11}(0) + \alpha_1^2\rho_{22}(0) - \alpha_1\alpha_2(\rho_{12}(0) + \rho_{21}(0)) \\
\rho_{ab}(0) &= \alpha_1\alpha_2(\rho_{11}(0) - \rho_{22}(0)) - \alpha_1^2\rho_{12}(0) + \alpha_2^2\rho_{21}(0).
\end{aligned} \tag{3.13}$$

The population ρ_{bb} will of course remain constant, whereas ρ_{aa} will decrease down to zero by emitting a superradiant pulse which is identical to that which one would obtain for two-level systems with a transition probability Γ and an initial population of the upper level $\rho_{aa}(0)$. The final state of the atoms, in the semiclassical model, is given by

$$\begin{aligned}
&\rho_{00}(\infty) = \rho_{aa}(0) \qquad \rho_{11}(\infty) = \alpha_2^2\rho_{bb}(0) \qquad \rho_{22}(\infty) = \alpha_1^2\rho_{bb}(0) \\
&\rho_{12}(\infty) = -\alpha_1\alpha_2\rho_{bb}(0) \qquad \rho_{01}(\infty) = \alpha_2\rho_{ab}(0) \qquad \rho_{02}(\infty) = -\alpha_1\rho_{ab}(0).
\end{aligned} \tag{3.14}$$

The final values of the populations and of the coherence between the upper states are exactly the same as they would be in non-cooperative spontaneous emission. In this latter case, however, the optical coherences would obviously remain equal to zero, because of the incoherent character of this type of emission. In the cooperative case, one immediately notices that

$$\alpha_1\rho_{01}(\infty) + \alpha_2\rho_{02}(\infty) = 0 \tag{3.15}$$

which is the exact analogue of equation (2.20) and shows the existence of a destructive interference between the two transitions. A detailed analysis of the mean values of the $r_i^{+(k)}r_j^{-(l)}$ operators (equations (3.3*b*)) in the final state given by equations (3.14) shows that in this case both the interatomic and the intratomic interference are negative: although the proportion of non-de-excited atoms is the same as in non-cooperative emission, this incomplete de-excitation can thus be called subradiance.

The subradiance rate, τ, which is obviously equal to $\rho_{bb}(0)$, is equal to 0 (no subradiance) if the atoms are prepared in the state $|a\rangle(\rho_{11}(0) = \alpha_1^2, \rho_{22}(0) = \alpha_2^2, \rho_{12}(0) = \alpha_1\alpha_2)$ and to 1 (no de-excitation) if they are prepared in the state $|b\rangle(\rho_{11}(0) = \alpha_2^2, \rho_{22}(0) = \alpha_1^2, \rho_{12}(0) = -\alpha_1\alpha_2)$. It is always larger than or equal to the smallest value, $\tau_0 = 1 - (2\sigma^2 - 1)^{1/2}$, which is compatible with the symmetry conservation (I). No additional subradiance occurs in the particular case of the 'most antisymmetrical' initial state ($\rho_{11}(0) = \rho_{22}(0) = 0.5, \rho_{12}(0) = 0$), which has a subradiance rate $\tau = \tau_0 = 50\%$, as in the case without the transition degeneracy. In contrast, an important effect due to this degeneracy is obtained when only one of the upper states, $|1\rangle$ or $|2\rangle$, is initially populated: the subradiance rate is then equal respectively to α_2^2 or to α_1^2, whereas $\tau_0 = 0$ (this result has already been obtained in the particular case $|\alpha_1| = |\alpha_2|$ (Gross and Haroche 1982)). In the latter case, a population trapping due to an intratomic destructive interference occurs in the non-cooperative regime (Agarwal 1974).

Semiclassical calculations including both the forward and backward emissions have been realised and confirm the predicted subradiance rates. Two examples are shown in figures 10 and 11. The first one concerns the above mentioned 'most antisymmetrical' initial state and figure 10 shows the time evolution of the system in this case, as compared with the non-degenerate case. The subradiance rate ($\tau = 50\%$) is the same in the two situations: in the degenerate case, the interference between the two transitions does not play any role in the final subradiant state, because $\rho_{01}(\infty) = \rho_{02}(\infty) = 0$. One remarks that the quenching of the emission on the less probable transition which accompanies the subradiance in the non-degenerate case completely disappears in the degenerate one; in this latter case, one has $\rho_{11}(\infty) = \alpha_2^2/2$ and $\rho_{22}(\infty) = \alpha_1^2/2$. Moreover, the interference between the two transitions is certainly constructive during the main part of the evolution since it clearly accelerates and enhances the emission (figure 10). In the nearly degenerate case, when the two upper levels are separated by $\hbar\delta$, with $T = \pi/\delta \gg T_{SR}$, the transition can be considered as fully degenerate provided that T is more than two or three times larger than the delay time of the pulse. For larger δ values, the delay time increases until it reaches the value of the non-degenerate case. As in this particular case subradiance is due to symmetry conservation, the subradiance rate does not change as δ increases, but the quenching effect modifies the final distribution of the atoms in the two upper states. In spite of the importance of this quenching effect (47% of the atoms remain in state $|1\rangle$ whereas 3% remain in state $|2\rangle$), superradiant beats with a contrast of about 50% are obtained for T smaller than a fraction of the pulse delay (Raimond 1979, Abraham and Bullough 1980).

In the second example, all the atoms are assumed to be initially in the same upper level (figure 11). Whereas the intensity radiated in the non-degenerate case is just a little smaller than in the non-degenerate one, the time evolution of the population is very different. In particular, the upper level which was initially empty is notably populated during the evolution (the same effect, with a similar origin, had already been obtained for a $\frac{1}{2} \rightarrow \frac{1}{2}$ transition in the small sample model (Crubellier and

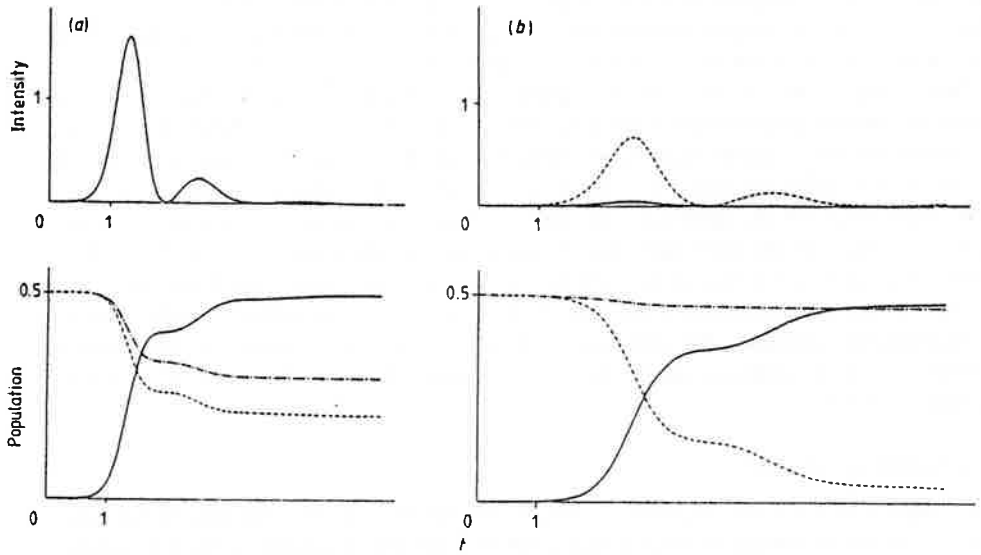

Figure 10. As figure 9 but for the three-level 'V' configuration, with $\Gamma_2/\Gamma_1 = 1.44$ and with an initial state defined by $\rho_{11}(0) = \rho_{22}(0) = 0.5$ and $\rho_{12}(0) = 0$.

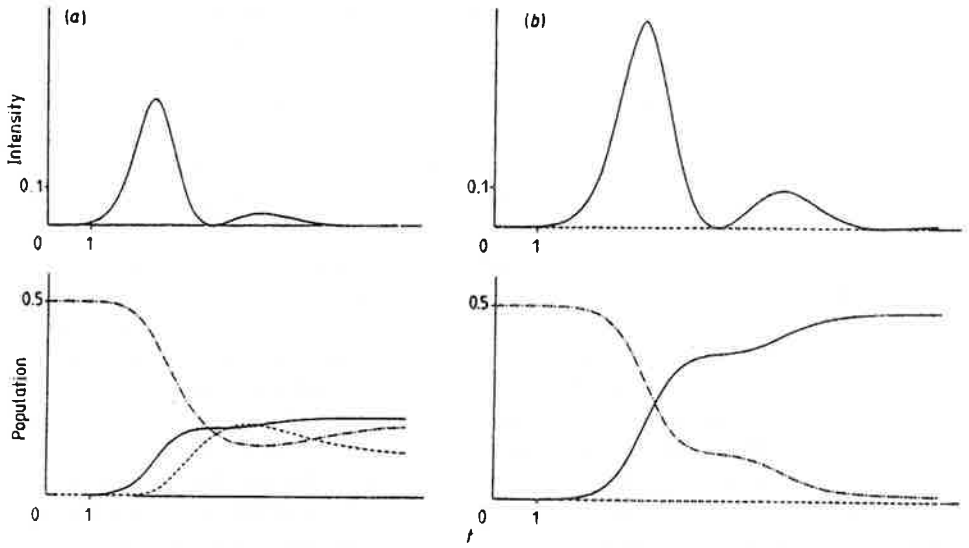

Figure 11. As figure 9 but for the three-level 'V' configuration, with $\Gamma_2/\Gamma_1 = 1.44$ and with all atoms initially in the upper state $|1\rangle$.

Schweighofer 1978)): any emitted photon can indeed be absorbed on any transition by any atom in the state $|0\rangle$. This process allows the collective dipole moments associated with both transitions to be different from zero and they finally reach the equilibrium defined by equation (3.15), in which a destructive interatomic and intratomic interference prevents the atoms from radiating. In both examples, shape fluctuations of the emitted light pulse are of course expected. As in § 2.4, they are caused by a single fluctuating-field source and they do not affect the subradiance rate. In this particular case, the transition can be considered as degenerate only if T is at least one order of magnitude larger than the delay time of the pulse. For larger δ values, subradiance disappears whereas state $|2\rangle$ always remains empty: superradiant beats cannot be obtained for these initial conditions.

These degenerate 'Λ' and 'V' configurations have allowed us to show how the interatomic and intratomic interference between the two transitions modifies the cooperative spontaneous emission and, in particular, for the 'V' configuration, the subradiance rate of an ensemble of such three-level atoms. However, studying the role of the interference between the different transitions in these cases is somehow artificial since a specific change of the state basis makes one transition disappear, so that this sort of interference no longer exists in the new basis. The situation was quite different in the case of the four-level configuration studied in § 2 and it is also different in the case of the 'degenerate' cascade. We will now study this latter case, where the interference between the two transitions causes new and peculiar effects in the cooperative spontaneous emission.

3.3. Degenerate cascade

3.3.1. Many-atom subradiant states. As in the four-level case, the existence of subradiance can be shown simply by exhibiting a general explicit expression for the subradiant states. We consider an ensemble of indistinguishable atoms in the degenerate-cascade level configuration (figure 6(*c*)); this can be, for example, the N indistinguishable

atoms of a slice with thickness smaller than $2\pi c/\omega_0$ of a pencil-shaped atomic sample. The master equation describing the evolution of the N atoms is given by equations (2.4), (3.2) and (3.3c), except that the sum in (3.2) runs now from 1 to N. We assume here that the atoms are initially (and thus remain) in a fully symmetrical state. As previously shown, this occurs in particular for initially uncorrelated atoms when their initial state is a pure state (I). The collective atomic state can then be represented in the occupation number representation and it is noted $|N_1 N_0 N_2\rangle$, where N_i is the population of the state $|i\rangle$. For p being any positive integer smaller than or equal to $N/2$, we consider the following non-normalised state:

$$|s(p)\rangle = \sum (-)^{N_0/2} \left(\frac{\alpha_1}{\alpha_2}\right)^{N_0/2} [2^{N_0/2}(N_0/2)!]^{-1} \left(\frac{N_0!\, N_2!}{N_1!}\right)^{-1/2} |N_1 N_0 N_2\rangle \tag{3.16}$$

where the sum runs over all even positive values of N_0 smaller than or equal to $2p$, the values of N_1 and N_2 verifying $N_0 + N_1 + N_2 = N$ and $N_1 - N_2 = -N + 2p$. The energy of these states is given by $E = (-N + 2p)\hbar\omega_0$ and they verify

$$R^-|s(p)\rangle = 0. \tag{3.17}$$

They are thus subradiant, unless $p = 0$, which corresponds to the fully de-excited state. Studying in more detail the expansion (3.16) in the particular case $p = N/2$ ($E = 0$), it appears that the only relevant components are those with $N_0 \sim N$ if $\alpha_1 > \alpha_2$ and those with $N_0 \sim 0$ if $\alpha_1 < \alpha_2$. More precisely one finds that the mean value of the population of the state $|0\rangle$ is

$$\langle N_0 \rangle = N - \frac{2}{\Gamma_1/\Gamma_2 - 1} \qquad \text{if } \Gamma_1 > \Gamma_2 \tag{3.18}$$

$$\langle N_0 \rangle = \frac{\Gamma_1/\Gamma_2}{1 - \Gamma_1/\Gamma_2} \qquad \text{if } \Gamma_1 < \Gamma_2 \tag{3.19}$$

both formulae being valid provided that Γ_1/Γ_2 is not too close to $1 (\Gamma_1/\Gamma_2 - 1 \ll 1/n$, with $n \ll N$ if $\Gamma_1 > \Gamma_2$ and $1 - \Gamma_1/\Gamma_2 \gg 1/N$ if $\Gamma_1 < \Gamma_2$). For large values of N, this particular subradiant state is therefore either a state with almost all the atoms in the state $|0\rangle$, as soon as Γ_1/Γ_2 is larger than one, or a coherent superposition with equal weights of states $|1\rangle$ and $|2\rangle$, the state $|0\rangle$ remaining almost empty, as soon as Γ_1/Γ_2 is smaller than one. This result can be generalised. Let us consider a subradiant state $|s(p)\rangle$ with $p = Na_1$, $N - p = Na_2$ ($a_1 \leqslant a_2$ and $a_1 + a_2 = 1$), i.e. with an energy $E = N(a_1 - a_2)\hbar\omega_0$. For $\Gamma_1 a_1 < \Gamma_2 a_2$ (zone I in figure 12), all subradiant states (3.16) are found to contain very few atoms in the state $|0\rangle$; one has then

$$\langle N_0 \rangle = \frac{\Gamma_1 a_1/\Gamma_2 a_2}{1 - \Gamma_1 a_1/\Gamma_2 a_2}. \tag{3.20}$$

This mean value is quite negligible provided that $\Gamma_1 a_1/\Gamma_2 a_2$ is not too close to one and the corresponding subradiant state is a coherent superposition of states $|1\rangle$ and $|2\rangle$ with respective weights a_1 and a_2. If, on the contrary, $\Gamma_1 a_1 > \Gamma_2 a_2$ (zone II in figure 12), the weight distribution of the components of the expansion (3.16) appears to be sharply peaked at a N_0 value such that

$$N_1 \alpha_1^2 = N_2 \alpha_2^2. \tag{3.21}$$

One remarks that for a state $|N_1 N_0 N_2\rangle$ which verifies equation (3.21), the induced emission rate from state $|1\rangle$ is equal to the absorption rate from state $|2\rangle$.

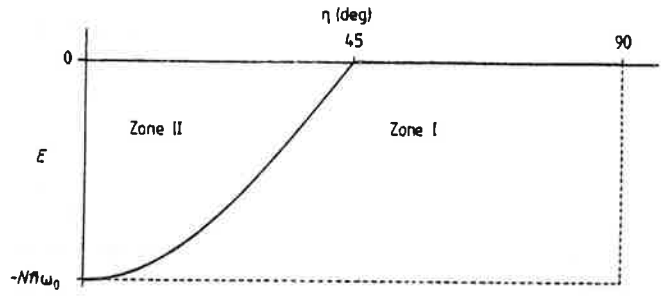

Figure 12. Diagram showing the two domains of energy (E) and η (with $\eta = \tan^{-1}(\Gamma_2/\Gamma_1)^{1/2}$) values of the subradiant states corresponding to the degenerate cascade configuration. In zone I each subradiant state is approximately a state $|N_1 0 N_2\rangle$; in zone II each subradiant state is very close to a state $|N_1 N_0 N_2\rangle$ with $N_1\alpha_1^2 = N_2\alpha_2^2$.

The mean values of the different populations are given by

$$\begin{aligned}
\langle N_0\rangle &= N\left(1-\frac{a_2-a_1}{\alpha_1^2-\alpha_2^2}\right)\\
\langle N_1\rangle &= \left(\frac{a_2-a_1}{\alpha_1^2-\alpha_2^2}\right)N\alpha_2^2 \qquad (3.22)\\
\langle N_1\rangle &= \left(\frac{a_2-a_1}{\alpha_1^2-\alpha_2^2}\right)N\alpha_1^2
\end{aligned}$$

and the corresponding subradiant state is a coherent superposition of the three states. These results strongly suggest that, conversely, either the states $|Na_1 0 Na_2\rangle$ or the states $|N_1 N_0 N_2\rangle$ verifying equations (3.21) might be practically subradiant if $\Gamma_1 a_1/\Gamma_2 a_1$ is respectively smaller or larger than one: this will be confirmed hereafter, by using the semiclassical model.

3.3.2. Semiclassical model. In the semiclassical model, using the plane-wave approximation, the evolution of a pencil-shaped sample of three-level atoms in the degenerate-cascade configuration (figure 6(c)) is given by the following Bloch–Maxwell equations:

$$\begin{aligned}
\frac{\partial}{\partial t'}Q_{11} &= \frac{\mathrm{i}d\alpha_1}{\hbar}(\varepsilon^* Q_{10}-\varepsilon Q_{10}^*)\\
\frac{\partial}{\partial t'}Q_{22} &= -\frac{\mathrm{i}d\alpha_2}{\hbar}(\varepsilon^* Q_{02}-\varepsilon Q_{02}^*)\\
\frac{\partial}{\partial t'}Q_{00} &= -\frac{\partial}{\partial t}(Q_{11}+Q_{22})\\
\frac{\partial}{\partial t'}Q_{12} &= \frac{\mathrm{i}d}{\hbar}\varepsilon(\alpha_2 Q_{10}-\alpha_1 Q_{02}) \qquad (3.23)\\
\frac{\partial}{\partial t'}Q_{10} &= \frac{\mathrm{i}d}{\hbar}[\alpha_1\varepsilon(Q_{11}-Q_{00})+\alpha_2\varepsilon^* Q_{12}]\\
\frac{\partial}{\partial t'}Q_{02} &= \frac{\mathrm{i}d}{\hbar}[\alpha_2\varepsilon(Q_{00}-Q_{22})-\alpha_1\varepsilon^* Q_{12}]\\
\frac{\partial}{\partial z}\varepsilon &= -\frac{\mathrm{i}kd}{2\varepsilon_0}(\alpha_1 Q_{10}+\alpha_2 Q_{02}).
\end{aligned}$$

In these equations, the Q_{ii} (with $i = 0, 1, 2$) are the populations of the different levels, whereas ε, Q_{10}, Q_{02} and Q_{12} are the slowly varying envelopes of the field and of the atomic coherences between the different levels. Time t' is the retarded time $t' = t - z/c$ and, for the sake of simplicity, equations (3.23) are written only for the wave packet travelling in the forward direction of the cylinder axis. We will study the evolution of the system either when all the atoms are initially in the upper state $|1\rangle$ or when they are initially in the intermediate state $|0\rangle$. In both cases, the early quantum period of cooperative spontaneous emission (Pillet 1977, Gross and Haroche 1982) will lock the phases of the optical coherences Q_{10} and Q_{02}, of the field ε and of the coherence Q_{12} to respective values ϕ, ϕ, $\phi - \pi/2$ and 2ϕ, ϕ being arbitrary. Putting then, in equations (3.23)

$$\begin{aligned} \varepsilon &= -i\varepsilon' \, e^{i\phi} \\ Q_{10} &= Q'_{10} \, e^{i\phi} \\ Q_{02} &= Q'_{02} \, e^{i\phi} \\ Q_{12} &= Q'_{12} \, e^{2i\phi}, \end{aligned} \tag{3.24}$$

it appears that the further semiclassical evolution of the system keeps ϕ constant. Equations (3.23) reduce then to a set of real equations

$$\begin{aligned} \frac{\partial}{\partial t'} Q_{11} &= -\frac{2d\alpha_1}{\hbar} \varepsilon' Q'_{10} \\ \frac{\partial}{\partial t'} Q_{22} &= \frac{2d\alpha_2}{\hbar} \varepsilon' Q'_{02} \\ \frac{\partial}{\partial t'} Q_{00} &= -\frac{\partial}{\partial t'}(Q_{11} + Q_{22}) \\ \frac{\partial}{\partial t'} Q'_{12} &= \frac{d}{\hbar} \varepsilon' (\alpha_2 Q'_{10} - \alpha_1 Q'_{02}) \\ \frac{\partial}{\partial t'} Q'_{10} &= \frac{d}{\hbar} \varepsilon' [\alpha_1 (Q_{11} - Q_{00}) - \alpha_2 Q'_{12}] \\ \frac{\partial}{\partial t'} Q'_{02} &= \frac{d}{\hbar} \varepsilon' [\alpha_2 (Q_{00} - Q_{22}) + \alpha_1 Q'_{12}] \\ \frac{\partial}{\partial z} \varepsilon' &= \frac{kd}{2\varepsilon_0} (\alpha_1 Q'_{10} + \alpha_2 Q'_{02}). \end{aligned} \tag{3.25}$$

These equations keep invariant the following quantity:

$$I = \alpha_2^2 Q_{11} + \alpha_1^2 Q_{22} + \alpha_1 \alpha_2 (Q'_{12} + Q'_{21}) \tag{3.26}$$

which is similar to the invariant (3.12) which has been found for the 'Λ' and 'V' degenerate configurations. For the degenerate cascade, however, one cannot find, at least in the rotating-wave approximation, an invariant operator similar to (3.10) and one notices that the R^+ and R^- operators do not belong to the Lie algebra of a SU(2) group. Contrary to the cases of the 'Λ' and 'V' degenerate transitions, the problem of

cooperative spontaneous emission in the degenerate-cascade configuration does not reduce to two-level superradiance. However, as in the first two cases, the conservation of the invariant (3.26) implies a specific restrictive condition on the final state which can be reached by the system and thus, eventually, a specific kind of subradiance. The evolution of the system will be studied for two cases of initial atomic states, with all atoms either in the upper state $|1\rangle$ or in the intermediate state $|0\rangle$.

3.3.3. Atoms initially in the upper state. We consider here that all atoms are initially in the upper state $|1\rangle$. If the cascade was non-degenerate, they would simply fall to the intermediate state $|0\rangle$ and then to the lower state $|2\rangle$ by emitting two successive superradiant pulses. Complications would arise if the probability of the lower transition was much larger than the probability of the upper transition, giving rise to 'truncated superradiance' (Molander and Stroud 1982). In any case, the final state of the evolution of the atoms would be fully de-excited. For a degenerate-cascade configuration, because of the existence of the invariant (3.26), the final state must verify

$$I(z,\infty) = I(z,0) \tag{3.27}$$

that is

$$\alpha_2^2 Q_{11}(z,\infty) + \alpha_1^2 Q_{22}(z,\infty) + 2\alpha_1\alpha_2 Q'_{12}(z,\infty) = \alpha_2^2. \tag{3.28}$$

This final state cannot thus be fully de-excited, unless $|\alpha_1| = |\alpha_2|$†. The destructive interatomic interference which is responsible for this incomplete de-excitation is described by the following equilibrium equation

$$\alpha_1 Q'_{10}(z,\infty) + \alpha_2 Q'_{02}(z,\infty) = 0 \tag{3.29}$$

which is similar to the equilibrium equations (2.20) and (3.15) found respectively in the cases of the four-level configuration and of the three-level 'V' configuration. The final populations of the three levels can be calculated, for any α_1 and α_2 values, by using equations (3.28) and (3.29) and the conservation of the trace of the squared monatomic density matrix. Figure 13 shows these values versus the angle η defined by $\eta = \tan^{-1}(\Gamma_2/\Gamma_1)^{1/2}$; the represented solution has been chosen among three possible ones as corresponding to the lowest final-energy value, i.e. to the most important de-excitation of the system. Numerical semiclassical calculations including both forward and backward emissions have been made for a number of α_1 and α_2 values and confirm the results of figure 13.

Since it is not fully de-excited, unless $|\alpha_1| = |\alpha_2|$, the final state of the evolution is subradiant; it should thus be identical to one of the subradiant states (3.16). Figure 13 can be divided into three parts, for η ranging from 0° to 30°, from 30° to 67.5° and from 67.5° to 90°. In the first part, the final monatomic state is given by

$$b_1|1\rangle + b_0|0\rangle + b_2|2\rangle \tag{3.30}$$

† As for the four-level configuration studied in § 2, the case $|\alpha_1| = |\alpha_2|$ is very particular. R^+ and R^- are infinitesimal operators of a SU(2) subgroup of the SU(3) cooperation group which is associated to N indistinguishable three-level atoms. The angular momentum associated with this SU(2) group is conserved. As the angular momentum of a state with all atoms in the upper state is equal to N whereas the angular momentum of a subradiant state $|s(p)\rangle$ is equal to $N-2p$, the evolution starting from the fully excited state will lead to the fully de-excited state $|s(0)\rangle$.

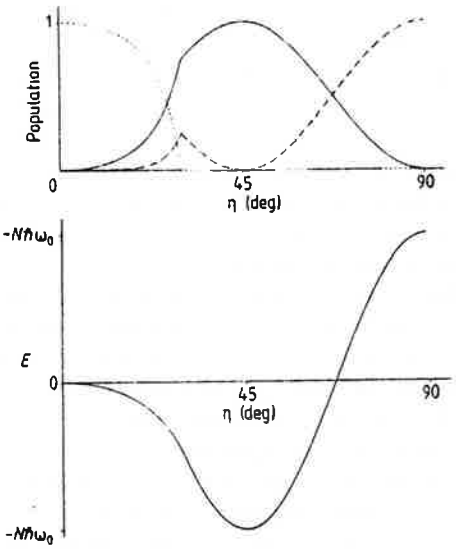

Figure 13. (*a*) Populations and (*b*) energy of the final state of the evolution plotted against $\eta = \tan^{-1}(\Gamma_2/\Gamma_1)^{1/2}$; the atoms are assumed to be initially in the upper state of the degenerate cascade. Populations of — · —, state $|1\rangle$, - - -, state $|0\rangle$ and ——, state $|2\rangle$.

with

$$b_1 = \alpha_2^2/(\alpha_1^2 - \alpha_2^2)$$
$$b_0 = [1 - \alpha_2^2/(\alpha_1^2 - \alpha_2^2)^2]^{1/2} \tag{3.31}$$
$$b_2 = -\alpha_1\alpha_2/(\alpha_1^2 - \alpha_2^2).$$

Equation (3.29) is thus verified: the evolution stops when the macroscopic dipole moment resulting from the interference between the two transitions is equal to zero. The collective final state can be written as a linear combination of fully symmetrical states $|N_1 N_0 N_2\rangle$, whose weight distribution is sharply peaked (with a width of the order of $N)^{1/2}$ to the state defined by

$$N_1 = Nb_1^2$$
$$N_0 = Nb_0^2 \tag{3.32}$$
$$N_2 = Nb_2^2.$$

This state obviously verifies equation (3.21) and the final state of the semiclassical evolution can be identified with a subradiant state of zone II in figure 12. In the second and third parts of figure 13, i.e. for $\eta > 30°$, the final state of the semiclassical evolution does not contain any atom in the intermediate state $|0\rangle$: in this case, the evolution stops when the macroscopic dipole moments corresponding to both the two transitions are separately equal to zero. The final monatomic state is a coherent superposition of states $|1\rangle$ and $|2\rangle$ given by

$$c_1|1\rangle + c_2|2\rangle \tag{3.33}$$

with

$$c_1 = \alpha_1^2 - \alpha_2^2$$
$$c_2 = 2\alpha_1\alpha_2. \tag{3.34}$$

The weight distribution of the collective final state on the fully symmetrical states $|N_1\ 0\ N_2\rangle$ exhibits a sharp maximum (of width of the order of $N)^{1/2}$; this collective state is thus approximately equal to a single state $|Nc_1^2\ 0\ Nc_2^2\rangle$ whose energy is equal to $N(c_1^2-c_2^2)\hbar\omega_0$, with $\alpha_1^2c_1^2<\alpha_1^2c_2^2$. For η varying from 30° to 67.5°, this energy is negative so that the final state of the semiclassical evolution can be identified with a subradiant state of zone I in figure 12. For η larger 67.5°, the energy of the final state is positive and there is no subradiant state which can be identified with such a state. However, the continuation for $\rho > N/2$ of the expression (3.16) provides us with states of very small decay rate, whose expansion reduces, like in zone I of figure 12, to a single state $|N_1\ 0\ N_2\rangle$. Such states might appear to be rigorously subradiant in the semiclassical model because, as is well known, this model deals with induced emission and absorption processes only and neglects in fact the spontaneous emission.

Two examples of numerical calculations, corresponding respectively to $\Gamma_2/\Gamma_1 = 0.307$ ($\eta = 29°$) and to $\Gamma_2/\Gamma_1 = 3.25$ ($\eta = 61°$), are represented in figures 14 and 15, for both the degenerate and non-degenerate configurations. For both η values, the two transitions of the degenerate cascade are strongly coupled by the interatomic interference, whereas the emission in the non-degenerate case consists essentially of two successive pulses (figures 14(b) and 15(b)). The first pulse emitted in the degenerate case contains photons emitted on both transitions (see the evolution of the populations in figures 14(a) and 15(a)). For $\Gamma_2 < \Gamma_1$ one observes, after the end of the emission, typical population oscillations from the intermediate state $|0\rangle$ to the extreme states $|1\rangle$ and $|2\rangle$ and vice versa, which are due to simultaneous emission and absorption processes. In both cases subradiance is observed in the degenerate case whereas the de-excitation is complete in the non-degenerate one; the obtained subradiance rates are in agreement with the results of figure 13. Fluctuations of the shape of the emitted light pulse (but not of the subradiance rate) are expected, as in the preceding cases of four-level atoms and of the 'Λ' and 'V' configurations.

Semiclassical calculations have also been realised in the nearly degenerate case, i.e. when the difference between the two atomic pulsations, δ, is much larger than T_{SR}^{-1}. The resonance condition is found to be quite severe: the transition can be considered

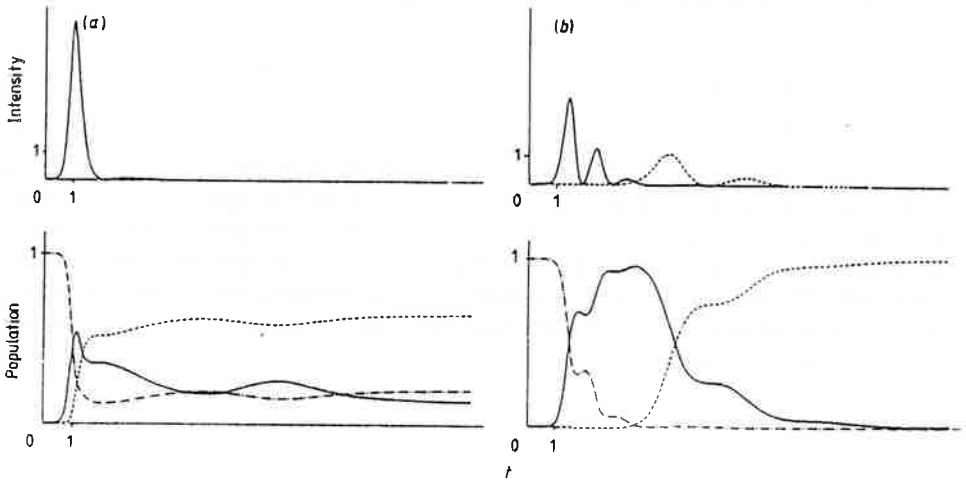

Figure 14. As figure 9 but for the cascade configuration, with $\Gamma_2/\Gamma_1 = 0.307$ ($\eta = 29°$) and with all atoms initially in the upper state.

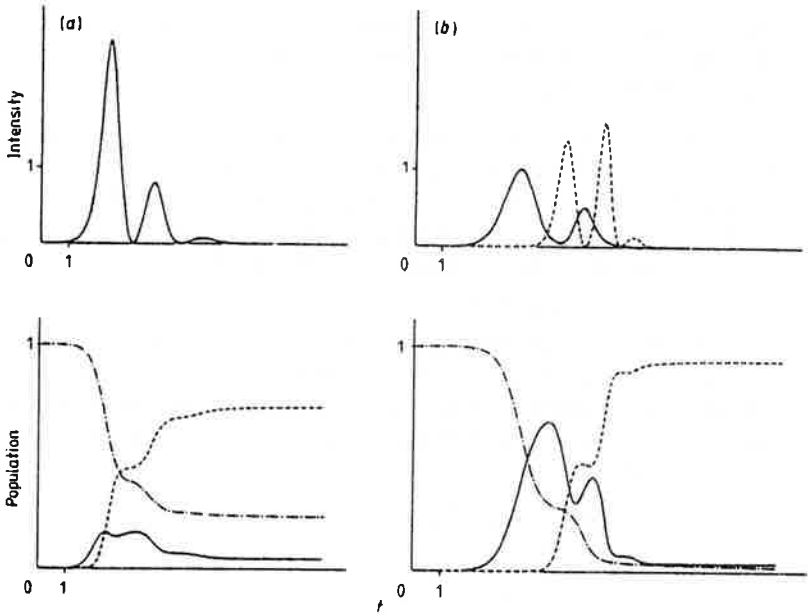

Figure 15. As figure 9 but for the cascade configuration, with $\Gamma_2/\Gamma_1 = 3.25$ ($\eta = 61°$) and with all atoms initially in the upper state.

as fully degenerate only if $T = \pi/\delta$ is larger than the pulse delay time by at least two orders of magnitude. For larger δ values, subradiance disappears whereas the emission occurs in two successive periods which correspond roughly to the two steps of the cascade (figures 16 and 17), as in the non-degenerate case. However, the second

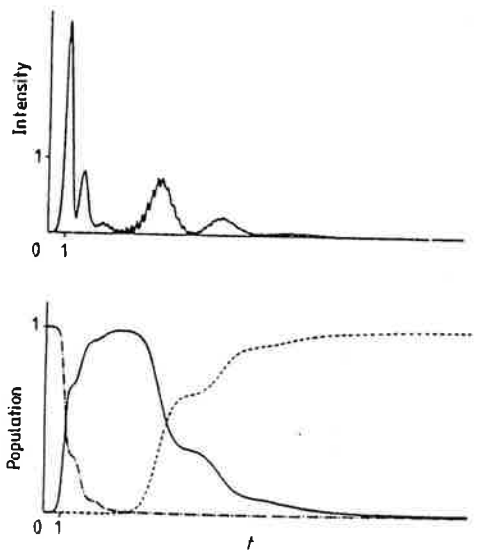

Figure 16. As figure 14 ($\eta = 29°$) but in the nearly degenerate case; the difference between the two atomic pulsations is δ, with $\pi/\delta = 0.31$.

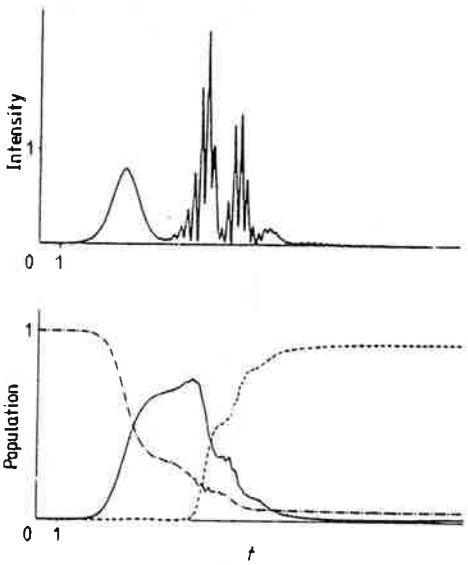

Figure 17. As figure 15 ($\eta = 61°$) but in the nearly degenerate case; the difference between the two atomic pulsations is δ, with $\pi/\delta = 0.31$.

emission period concerns in fact both transitions and superradiant beats are obtained (figures 16 and 17): in superradiant emission, even two cascading transitions can beat one against the other.

3.3.4. Atoms initially in the intermediate state. We now consider that all atoms are initially in the intermediate state $|0\rangle$. If the cascade was non-degenerate, the problem would obviously reduce to two-level superradiance and all atoms would in principle fall to the lower state $|2\rangle$ by emitting a superradiant pulse. For the degenerate cascade, the conservation of the invariant (3.26) implies that

$$\alpha_2^2 Q_{11}(z,\infty)+\alpha_1^2 Q_{22}(z,\infty)+2\alpha_1\alpha_2 Q'_{12}(z,\infty)=0.$$

The final state of the evolution cannot be fully de-excited unless $\alpha_1=0$. The final values of the populations can be calculated as in § 3.3.3 and the solution which gives the largest de-excitation is represented in figure 18. In fact the evolution of the system for the considered initial conditions is given by a peculiar solution of equation (3.25) which verifies

$$\begin{aligned}&\frac{Q_{11}}{\alpha_1^2}=\frac{Q_{22}}{\alpha_2^2}=-\frac{Q'_{12}}{\alpha_1\alpha_2}=Q_{nn}\\&\frac{Q'_{10}}{\alpha_1}=-\frac{Q'_{02}}{\alpha_2}=Q_{n0}.\end{aligned}\tag{3.35}$$

Equations (3.25) reduce then to

$$\begin{aligned}&\frac{\partial}{\partial t'}Q_{nn}=\mp\frac{2d}{\hbar}(\pm\varepsilon')Q_{n0}\\&\frac{\partial}{\partial t'}Q_{00}=\pm\frac{2d}{\hbar}(\pm\varepsilon')Q_{n0}\\&\frac{\partial}{\partial t'}Q_{n0}=\pm\frac{2d}{\hbar}(\pm\varepsilon')(Q_{nn}-Q_{00})\\&\frac{\partial}{\partial z}(\pm\varepsilon')=\frac{kd|\alpha_1^2-\alpha_2^2|}{2\varepsilon_0}Q_{n0}.\end{aligned}\tag{3.36}$$

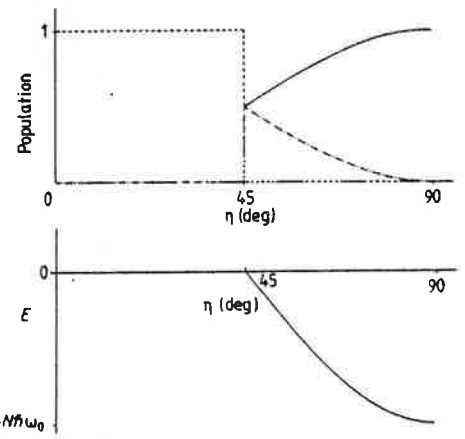

Figure 18. As figure 13 but when all atoms are initially in the intermediate state of the degenerate cascade.

These equations are the Bloch-Maxwell equations of an ensemble of two-level systems; one of the two levels is the intermediate state $|0\rangle$ and the other one is a fictitious state, $|n\rangle$, of population Q_{nn}. Q_{n0} is the optical coherence between the two states. For $\Gamma_1 < \Gamma_2$ one has to take the lower signs in equations (3.36) and the upper state of the transition appears to be the state $|0\rangle$, whereas for $\Gamma_1 > \Gamma_2$ one has to take the upper signs and the state $|0\rangle$ is the lower one.

If $\Gamma_1 < \Gamma_2$, all atoms will leave the state $|0\rangle$; the largest part ($N\alpha_2^2$) will fall in the lower state $|2\rangle$ whereas the others ($N\alpha_1^2$) will absorb one photon and reach the upper state $|1\rangle$, these transfers being accompanied by the emission of a superradiant pulse containing $N(\alpha_2^2 - \alpha_1^2)$ photons only. The monatomic final state is given by

$$\alpha_1|1\rangle - \alpha_2|2\rangle. \tag{3.37}$$

The weight distribution of the collective state on the fully symmetrical states $|N_1\ 0\ N_2\rangle$ is sharply peaked (with a width of the order of $N^{1/2}$) at $N_1 = N\alpha_1^2$ and $N_2 = N\alpha_2^2$: the final state of the semiclassical evolution can thus be identified with a subradiant state of zone I in figure 12.

If, on the contrary, $\Gamma_1 > \Gamma_2$, the system cannot evolve at all. The probability of the atoms absorbing one photon is larger than the probability of emitting one. No cooperative emission is then possible and the initial state $|0\ N\ 0\rangle$ can be identified with a subradiant state of zone II in figure 12. The atoms will thus simply decay by ordinary spontaneous emission in the non-cooperative field modes (I) toward the state $|2\rangle$: the presence of the upper level prevents any superradiant emission.

An example of semiclassical numerical results (including both forward and backward emissions) in the case where $\Gamma_2/\Gamma_1 = 1.42$ ($\eta = 50°$) is given in figure 19, for both the degenerate and non-degenerate cascades.

As expected, the evolution in the degenerate case consists essentially of a population transfer from the intermediate state $|0\rangle$ toward the extreme states $|1\rangle$ and $|2\rangle$. A small superradiant pulse is still emitted, not much later than in the non-degenerate case, but its area is only about 17% that of the pulse which is emitted in this latter case; this is

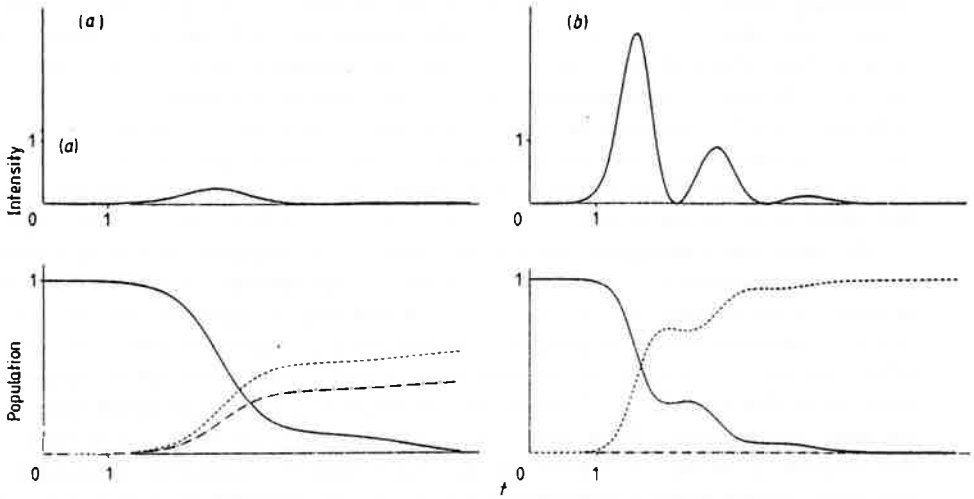

Figure 19. As figure 9 but for the cascade configuration, with $\Gamma_2/\Gamma_1 = 1.42$ ($\eta = 50°$) and with all atoms initially in the intermediate state.

of course closely related to the important (42%) population of the upper state in the final subradiant collective state. As in all other considered cases, fluctuations of the shape of the emitted field but not of the final state are predicted. In this case, the transition can be considered as fully degenerate as long as $T = \pi/\delta$ (where δ is the difference between the two atomic pulsations) is more than 20 times larger than the pulse delay time. For larger δ values, subradiance progressively disappears. Superradiant beats are never observed in this particular case.

4. Conclusion

In atomic systems with degenerate transitions, superradiance and subradiance are due to two sorts of interatomic interference, either on each transition separately, as in the absence of degenerate transition, or between the different transitions of the same frequency and polarisation. These two sorts of interference have many identical properties. Firstly, they are both basically due to the indistinguishability of the atoms with respect to the emission or absorption processes and they therefore occur in the same conditions and with the same limitations, concerning the 'dephasing' processes, for example. Secondly, these two sorts of interatomic interference have a similar influence on the evolution of the system: either an acceleration and enhancement of the emission when constructive or an inhibition of the emission (in the cooperative field modes) when destructive. Important differences also exist between the two sorts of interatomic interference and therefore between the two corresponding subradiance phenomena. For instance, while the constructive or destructive nature of the interatomic interference on the same transition depends simply on the symmetry properties of the collective atomic state (I), so that the subradiance rate is easily calculated from the initial symmetry properties, the subradiance rate for an atomic system with a degenerate transition depends in a complicated manner on the initial populations and on the different transition probabilities. In addition, the subradiance phenomenon which is due to the presence of a degenerate transition completely disappears when the degeneracy is removed, i.e. when the interatomic interference is replaced by interatomic beats. Four typical examples of level configurations with a degenerate transition have been studied. The first one concerns a four-level configuration with two transitions of the same frequency and polarisation. As an interatomic interference between the different Zeeman transitions of same polarisation is expected in cooperative emission on a $j \rightarrow j'$ transition between degenerate levels, these results open the way to the study of this latter case. An example, corresponding to the experimental situation of the first observation of subradiance (Pavolini *et al* 1985), will be discussed in paper IV.

The other three examples concern the three-level configurations with a degenerate transition ('degenerate' 'Λ', 'V' and cascade configurations). The first two have a mainly formal interest. In the case of the degenerate or nearly degenerate cascade, new and surprising effects are predicted. It is in particular shown that the two transitions of the cascade can beat one against the other, giving rise to a new sort of superradiant beat. It is also found that if the atoms are excited in the intermediate state of the cascade, superradiance from this level is expected to be resonantly quenched when the frequencies of the two transitions become equal; the quenching is complete and the atoms remain in the intermediate state if the most probable transition is the upper one; in the reverse case, the atoms are transferred to the extreme states of the cascade by the emission of a small superradiant pulse whose area is proportional to the difference

between the two transition probabilities. The observation of these effects remains, however, to be seen; an experimental realisation of degenerate cascades has indeed already been studied, for Na Rydberg atoms (Safinya *et al* 1981), but the cooperative effects were quenched by the inhomogeneous broadening, so that only resonant collision effects were observed (Gallagher *et al* 1982).

Before giving, in paper IV, the detailed theoretical description of the subradiance phenomenon which has been observed in the case of a $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ atomic cascade (Pavolini *et al* 1985), we shall briefly study, in paper III, the 'limited superradiance' expected in the small samples (Friedberg *et al* 1972, Friedberg and Hartmann 1974a, b, Gross and Haroche 1982), which is another kind of subradiance. As in paper I, the symmetry properties of the collective state are essential for the description of the evolution of the system: subradiance is in fact due to a sudden symmetry breaking caused by the dipole–dipole interaction.

Acknowledgment

This work has greatly benefited by the continuing cooperation of S Liberman and P Pillet.

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5.2: Configurations impliquant deux niveaux dégénérés

Dans le paragraphe (§ 3) de l'article ci-après ("Superradiance and subradiance: IV. Atomic cascades between degenerate levels"), on considère une configuration simple pour laquelle les deux sortes d'interférence, sur chaque transition et sur les différentes transitions de même fréquence et même polarisation, sont simultanément présentes dans l'évolution globale du système. Il s'agit du cas d'une transition $3/2 \rightarrow 1/2$, lorsque tous les sous-niveaux Zeeman du niveau supérieur sont initialement peuplés, sauf le sous-niveau de $m = 3/2$, et lorsqu'on choisit l'axe de quantification dans la direction de propagation de la lumière (figure 6). La configuration étudiée est en fait quelque peu fictive, dans

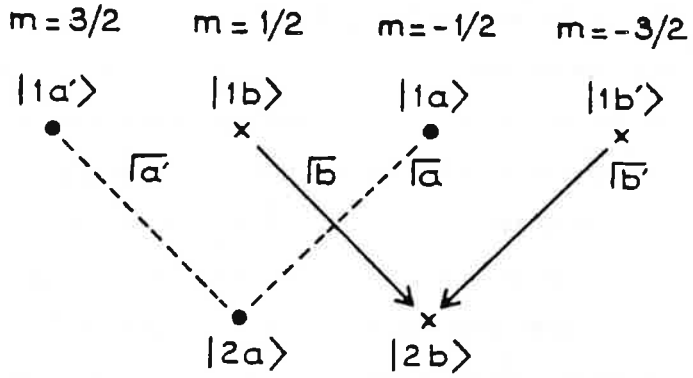

figure 6: configuration simple impliquant à la fois la subradiance "par antisymétrie" et la subradiance "Zeeman"

la mesure où on fait varier les deux probabilités de transitions aussi bien que les populations initiales, afin de prévoir qualitativement à travers cet exemple simplifié l'évolution d'un système plus général impliquant une transition entre niveaux dégénérés j et j' quelconques. Afin de simplifier encore davantage, on considère le cas où les deux sous-niveaux $m = 1/2$ (état $|1b>$) et $m = -3/2$ (état $|1b'>$) sont également peuplés initialement et où il n'y a pas de cohérence entre eux. On fait varier

la population initiale du sous–niveau $m = -1/2$ (état $|1a>$) par rapport aux deux autres, afin d'étudier l'influence des populations sur le taux de subradiance totale. On choisit en premier lieu comme probabilités de transition les probabilités de transition de la transition $3/2 \rightarrow 1/2$, qui sont dans le rapport $\Gamma_{b'} = 3\Gamma_b = 3\Gamma_a$. On montre les taux de subradiance τ, τ_Z et τ_V qui sont respectivement obtenus lorsque les trois états supérieurs sont initialement peuplés, lorsque l'état $|1b>$ n'est pas peuplé $(n_{1a}(0) + n_{1b'}(0) = 1)$, et lorsque l'état $|1a>$ n'est pas peuplé $(n_{1b}(0) + n_{1b'}(0) = 2n_{1b}(0) = 1)$. Les taux τ_V et τ_Z correspondent donc respectivement à la subradiance "par antisymètrie" (transitions $|1b> \rightarrow |2b>$ et $|1b'> \rightarrow |2b>$) et à la subradiance "Zeeman" (transitions $|1a> \rightarrow |2a>$ et $|1b'> \rightarrow |2b>$). Comme on peut le prédire à partir des propriétés de symétrie de l'état collectif d'un système qui est initialement dans un mélange statistique d'états, 50% de la population initialement dans les deux états $|1b>$ et $|1b'>$ reste dans le niveau supérieur. D'autre part, on peut calculer τ_Z à l'aide des équations (2.19 et 2.21) de l'article présenté dans le paragraphe précédent. Quand les états $|1b>$ et $|1b'>$ ne sont pas peuplés initialement, le système se réduit à un système à deux niveaux, avec une inversion totale de population: il se désexcite par émission d'une impulsion superradiante qui vide complètement le niveau supérieur. Quand l'état $|1a>$ n'est pas peuplé initialement, on retrouve une configuration en "V" avec un mélange statistique initiale des deux états supérieurs, et donc un taux de subradiance de 50% de la population initiale. Ailleurs, τ est aussi simplement la somme des deux taux de subradiance associés à la subradiance "par antisymètrie" et à la subradiance "Zeeman". On verra (§ 3.3) que ceci est souvent le cas, mais que, pour des probabilités de transition différentes, la competition entre les deux effets peut diminuer ainsi le taux de subradiance totale.

SUPERRADIANCE AND SUBRADIANCE: IV. ATOMIC CASCADES BETWEEN DEGENERATE LEVELS

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ABSTRACT

This paper is the fourth of a sequence devoted to the theoretical study of subradiance, i. e. of the cooperative inhibition of spontaneous emission. It first describes how the main results of the first paper have been developed in the general case of $j \rightarrow j'$ transitions between degenerate levels. In particular, the link between statistical mixing and symmetry properties is now derived in the general case of $n-$level atoms. We also explain how these results have allowed us to conceive the experiment which has recently provided the first evidence for the subradiance phenomenon, in the case of the $3/2 \rightarrow 1/2 \rightarrow 1/2$ cascade of atomic gallium. The specific influence of the degenerate transitions which are associated with the Zeeman transitions of same polarisation is then discussed on a simplified example. Numerical simulations corresponding to the case of the experiment and to similar cases are finally presented and analysed.

1. INTRODUCTION

In the preceding papers of the sequence (Crubellier et al 1985a, Crubellier and Pavolini 1986, Crubellier 1986, hereafter referred to as, respectively, I, II, III), we studied theoretically three different and typical situations for which cooperative inhibition of spontaneous emission, i. e. subradiance, is predicted. In the first paper we showed that a destructive interatomic interference can occur in many−atom collective states which are not fully symmetrical with respect to the atomic permutations. Because of the conservative law which governs the symmetry of the collective state, subradiance was in particular predicted in the typical case of the three−level "V" configuration for any initial collective state which is not fully symmetrical. We showed in addition that, for uncorrelated atoms, the "amount" of statistical mixing fully characterises the symmetry of the collective state. This close link between two different characteristics of a collective state, statistical mixing and permutation symmetry, provided us with the key for the observation of the subradiance phenomenon: it connects in fact the usually non−cooperative excitation process, which prepares uncorrelated atoms, and the cooperative deexcitation, which is determined by the symmetry of the collective state. Aiming at the experimental evidence for subradiance, we have then looked for a realistic generalisation of the "V" configuration, and the case of a transition between degenerate levels, precisely a $j \rightarrow j-1$ transition, has finally been chosen.

Studying cooperative emission between degenerate levels has led us to new theoretical developments, the interest of which is of course not restricted to the elaboration of a subradiance experiment. We have for instance studied both theoretically and experimentally the remarkable polarisation properties of the light emitted on such transitions (Crubellier et al 1978, 1981, 1984, 1986, Pillet 1982). We have also noticed that a $j \rightarrow j'$ transition generally contains several Zeeman transitions of same polarisation, i. e. several transitions corresponding to the same degenerate transition. We have then shown that, quite generally, the presence of a degenerate transition

involves an additional interatomic interference (II). Since this interference concerns different transitions, its either constructive or destructive nature does no longer depend on the permutation symmetry properties of the collective state; simple typical examples (four–level and three–level configurations) are studied in II.

For an atomic configuration including only the two relevant j and $j-1$ levels, the evidence for a subradiance effect could not be given without an absolute measure of the total number of the emitted photons, or of the number of either deexcited or non–deexcited atoms. The situation is quite different in the case of atomic cascades between degenerate levels, which has allowed us to elaborate and then to realise the experiment which has given the first experimental evidence for the existence of the subradiance phenomenon (Crubellier et al 1985b, Pavolini et al 1985). In such a case, a subradiant state in which the emission is cooperatively inhibited on a given transition can indeed be destroyed by cascading cooperative emission on a lower transition. Superradiant emission on the first transition can then resume, giving rise to what we call a "subradiance echo", which is a clear manifestation of the former incomplete deexcitation.

The present paper is devoted to the theoretical study of cooperative spontaneous emission on such atomic cascades and, especially, to the interpretation of our experiment. The two kinds of subradiance phenomena which occur in such a configuration are taken into account. The first one, which is due to the symmetry conservation (I), is called "V" subradiance, whereas the second one, which we call "Zeeman" subradiance, is due to the presence of degenerate transitions (II).

In the rest of the paper we first study, in the general case of a $j \rightarrow j'$ transition, the "V" subradiance which can quite generally be predicted from symmetry considerations (§ 2). We now demonstrate in the general case of n–level atoms the close link which connects statistical mixing and symmetry of collective states of uncorrelated atoms. As a consequence, we show that a collective state spontaneously evolve, through cooperative emission, to a subradiant state provided that j' is equal to $j-1$ and that

the "amount" of statistical mixing is large enough. The respective influence of the "V" subradiance and of the "Zeeman" subradiance is then analysed in simplified examples (§ 3). We finally discuss two experimental situations (§ 4). The first one concerns a $3/2 \rightarrow 1/2 \rightarrow 1/2$ cascade, i. e. the case of the atomic cascade of gallium on which the observation of subradiance has recently been made (Crubellier et al 1985b, Pavolini et al 1985). In this case, due to the ground $4p$ configuration of gallium, the excitation of the upper $3/2$ level of the cascade can be achieved, with either linearly or circularly polarised light, from either a $1/2$ or a $3/2$ level. All the corresponding initial states are considered here, assuming in addition that the light intensity is either large enough for the transition to be saturated or small enough for the absorption regime to be linear. The second example concerns a $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ cascade and an initial state which is a complete statistical mixing with equal weights of the upper Zeeman sublevels. This is the case of the HF superradiance experiment (Skribanowitz et al 1972, Skribanowitz et al 1973, Herman et al 1974), when the excitation uses the molecular P branch: symmetry considerations then predict a subradiance effect.

2. Symmetry properties and subradiance in a $j \rightarrow j'$ transition

2.1 Statistical mixing and subradiance

A $j \rightarrow j'$ dipole transition between degenerate levels of angular momentum j and j' (with j' equal to either $j+1$, j or $j-1$) is considered. As it was the case for the three level "V" configuration (I), a subradiance effect can be predicted within simple considerations about the conservation of the trace σ^2 of the squared monatomic density matrix. Suppose that all the atoms are initially in the upper j level (initial full population inversion); then

$$\frac{1}{2j+1} \leq \sigma^2 \leq 1 \tag{1}$$

If the system completely deexcites to the lower level one has necessarily:

$$\frac{1}{2j'+1} \leq \sigma^2 \leq 1 \tag{2}$$

If $j' \geq j$, this condition is less restrictive than condition (1) and a complete deexcitation may occur. If, on the other hand, $j' = j-1$ and if, in addition,

$$\frac{1}{2j+1} \leq \sigma^2 \leq \frac{1}{2j-1} \tag{3}$$

condition (2) cannot be fulfilled, and incomplete deexcitation, i. e. subradiance, has to be expected. As for the three–level system, this result is physically understood as being due to a destructive interference between the atoms.

2.2 Statistical mixing and symmetry

We showed in I that the symmetry properties of the considered initial collective state, which was in fact a two–level atom state, were fixed by the initial statistical mixing. A similar link between statistical mixing and symmetry properties can be found in the general case of n–level atoms, provided that the collective density matrix

is factorisable and homogeneous. Consider a (spatially) homogeneous factorisable density matrix describing the collective state of a large number N of identical $n-$level atoms,

(4) $$\rho = \prod_{\alpha=1..N} \rho^{\alpha}$$

The diagonal form of the monatomic density matrix is written as

(5) $$\rho^{\alpha} = \sum_{i=1,n} \rho_{ii} |i><i|_{\alpha\alpha}$$

$|i>$ being a monatomic state, and the eigenvalues ρ_{ii} are assumed to decrease as i increases. The trace of the squared monatomic density matrix is then

(6) $$\sigma^2 = \sum_{i=1,n} \rho_{ii}^2$$

Being symmetrical under the atomic permutations, ρ is diagonal with respect to the symmetry type and can be written as a sum of density operators with well-defined symmetry types,

(7) $$\rho = \sum_{\chi} a(\chi)\rho(\chi)$$

where the sum runs over all the symmetry types χ, and where the trace of each $\rho(\chi)$ operator is assumed to be equal to one. It is shown hereafter that this sum essentially reduces to a single term, the symmetry type of which is given by

(8) $$\{N\rho_{ii}\} \equiv \{N\rho_{11}, N\rho_{22}, ..., N\rho_{nn}\}$$

The mean value corresponding to the state ρ of the Casimir operator of the $S_N \otimes SU(n)$ cooperation group (see I, eqs (5.4) and (5.5)), W, is given by

(9) $$< W >= Nn + N(N-1)\sigma^2 \sim N^2\sigma^2$$

It is recalled that the W operator is diagonal with respect to the symmetry type. Moreover, it provides a "symmetry scale" (by analogy with a temperature scale)

since its eigenvalues are decreasing when the symmetry "decreases", i. e. when one or several cells of the corresponding Young diagram are removed from one row to a lower one (I). The mean value $< W >$ can be written as a linear combination of eigenvalues of W

$$< W >= \sum_{\chi} a(\chi) W(\chi) \tag{10}$$

It is also approximately equal to the eigenvalue of W associated with the particular symmetry type given by eq. (8), which is

$$W(\{N\rho_{ii}\}) = \sum_{i=1,n} N\rho_{ii}(N\rho_{ii} + n - 2i + 1) \sim N^2\sigma^2 \tag{11}$$

Consequently the sum (7) can contain symmetry types higher than $\{N\rho_{ii}\}$ only if it also contains lower symmetry types. One notices then that $\{N\rho_{ii}\}$ is the lowest symmetry type compatible with the values $N\rho_{ii}$ of the different populations of the levels. The presence of terms with a symmetry lower than $\{N\rho_{ii}\}$ in the development (7) would therefore necessarily imply non−zero fluctuations of the different populations $< N_i >$. As these fluctuations are given by

$$\sqrt{\frac{< N_i^2 > - < N_i >^2}{< N_i >^2}} = \sqrt{\frac{1 - \rho_{ii}}{N\rho_{ii}}} \tag{12}$$

they can be neglected if N is large and the development (7) of ρ does not contain any term of symmetry notably lower than (and thus higher than) $\{N\rho_{ii}\}$: the symmetry type of the considered collective state is therefore simply determined by the diagonal form of the monatomic density matrix. For large N, it is in fact well−defined, although the atoms are assumed to be uncorrelated. This somehow paradoxal situation is already explained in I. Like for the "V" configuration, it provides in the general case a valuable link between the usually non−cooperative excitation process and the cooperative deexcitation.

2.3 Subradiance rate on a $j \rightarrow j'$ transition

Like in I, we consider now a pencil–shaped sample in the plane wave approximation. The whole active medium is thought of as formed by successive slices S_k of thickness smaller than the transition wavelength. As already shown (I), the atom–field Hamiltonian is invariant under the atomic permutations inside any given slice S_k, that is under the permutation group S_N where N is the number of atoms contained in the slice. Cooperative spontaneous emission conserves therefore the symmetry types, i. e. the irreducible representations of the groups S_N. When, as it is usually the case, uncorrelated atoms are prepared, the initial atomic state is represented by a factorisable density matrix. We assume, in addition, that it is at least locally homogeneous, i. e. that it is homogeneous in each slice. If, in the initial state, all the atoms are in the upper j level, the corresponding wavefunctions are represented by Young tableaux containing at most $2j + 1$ rows. On the other hand, final fully deexcited states are represented by Young tableaux containing at most $2j' + 1$ rows. For $j' \geq j$, the symmetry type conservation always allows the complete deexcitation of the system. On the contrary, a complete deexcitation in the case $j' = j - 1$ is only possible if the initial symmetry type is represented by a Young diagram containing at most $2j - 1$ rows. Subradiance is otherwise predicted. Consider, for instance, the case of a density matrix with maximum statistical mixing, i. e. the complete statistical mixing with equal weights of all the Zeeman sublevels of the upper j level. In this case, $\sigma^2 = \frac{1}{2j+1}$. As shown in section 2.2, this state has the most antisymmetrical symmetry type $\{\frac{N}{2j+1}, \frac{N}{2j+1}, \ldots\ldots, \frac{N}{2j+1}\}$. The corresponding Young diagram contains $(2j + 1)$ rows and $\frac{N}{2j+1}$ columns. The most deexcited collective states having this symmetry type are Young tableaux containing $N/(2j + 1)$ atoms in each of the $(2j - 1)$ states of the lower level. The $2N/(2j + 1)$ remaining cells of the tableaux necessarily still contain excited atoms: the final state is thus subradiant. In this case, the destructive interference takes place inside ensembles of $(2j + 1)$ atoms which include two excited and $(2j - 1)$ deexcited atoms.

These results can be generalised to any case of initially fully excited atoms. The amount of atoms which are prevented from getting deexcited because of the destructive interference, i. e. the subradiance rate, can be calculated for any symmetry type $\{l_1, l_2, ..., l_h\}$. It is of course equal to zero if $h \leq (2j-1)$, and one has

$$\begin{aligned} \tau_V &= \frac{l_h}{N} \qquad \text{if} \quad h = 2j \\ \tau_V &= \frac{l_{h-1} + l_h}{N} \qquad \text{if} \quad h = 2j+1 \end{aligned} \tag{13}$$

Using the results of § 2.2, the subradiance rate τ_V can thus simply be derived from the eigenvalues of the initial monatomic density matrix; one has

$$\tau_V = \rho_{2j+1,2j+1}(0) + \rho_{2j,2j}(0) \tag{14}$$

where the eigenvalues $\rho_{ii}(0)$ of the initial density matrix are assumed to monotonically decrease with increasing i (between 1 and $2j+1$).

2.4 Subradiance and atomic cascades between degenerate levels

If the atomic configuration only contained the relevant $j \rightarrow j-1$ transition, the experimental observation of subradiance would imply a simultaneous measurement of the number of emitted photons (or, equivalently, the number of either deexcited or non−deexcited atoms) and of the number of initially excited atoms. The presence of additional atomic levels completely changes the situation. We show hereafter that in the case of a $j \rightarrow j-1 \rightarrow j'$ cascade configuration, for instance, the occurrence of the subradiance effect drastically upsets the qualitative behaviour of the cooperative emission.

We consider a $j \rightarrow j-1 \rightarrow j'$ cascade between degenerate levels. For the sake of simplicity, we assume that $j' \geq j-1$, and we consider that a complete statistical mixing with equal weights of all the Zeeman sublevels has initially been prepared in the upper j level. The symmetry type of the initial state is then $\{\frac{N}{2j+1}, \frac{N}{2j+1}, ..., \frac{N}{2j+1}\}$. As previously shown, subradiance is then expected on the $j \rightarrow j-1$ transition. Therefore, after the emission of a superradiant pulse on the $j \rightarrow j-1$ transition,

the Young tableaux describing the collective state still contain $2N/(2j+1)$ excited monatomic states: a subradiant state is thus reached. The presence of the j' level still allows the emission to resume via a superradiant pulse on the lower $j-1 \rightarrow j'$ transition. As we assume that $j' \geq j-1$, no subradiance is predicted on this transition and this pulse is expected to completely empty the intermediate level of the cascade. The atoms are then in a collective state which contains $2N/(2j+1)$ atoms in the upper j level and $(2j-1)N/(2j+1)$ atoms in the j' level. Since the j and j' levels are not connected by any allowed transition, this collective state, although partially antisymmetrical, is not subradiant. Emission on the first transition can then start again, and it can completely empty the upper j level via a superradiant pulse. This pulse, which we call the "subradiance echo", testify to the existence of an intermediate state in which the emission was cooperatively inhibited on the $j \rightarrow j-1$ transition. Such an echo is shown to occur if and only if the initial symmetry type is such that subradiance is expected on the first transition. The number of photons emitted in the subradiance echo is of course proportional to the predicted subradiance rate (14).

The qualitative evolution of a configuration which implies degenerate levels is most simply described if the quantisation axis is taken along the axis of the pencil-shaped sample, as it will always be the case in the following. Superradiant emission, which occurs in the same direction, cannot be $\pi-$polarised: it will then concern σ_+ and σ_- polarisations only. Two distinct level–subsystems, a and b (figures 1,4,9), can thus separately be considered. The atoms remain in a same subsystem during the whole evolution. One can thus define, at least statistically, two classes of atoms with constant number of atoms N_a and N_b (Pavolini et al 1985). As shown in II, the interclass permutation symmetry properties influence only the density matrix elements connecting the two subsystems and they play no role in the interatomic interference which occurs among different Zeeman transitions of same polarisation. Therefore the groups S_{N_a} and S_{N_b} of local permutations among atoms of a same class will provide all the relevant symmetry properties of the collective state.

The two subsystems are coupled one to the other by the interference between the Zeeman transitions of same polarisation: this interference is in particular responsible for the simultaneous occurrence of the emission by the two subsystems a and b. Moreover, a second subradiance effect, due to the possibility of this interference being destructive, can also manifest itself. We will see in the next section how the two subradiance phenomena compete in a simple configuration involving a degenerate transition.

3."V" subradiance and "Zeeman" subradiance

According to the choice of the quantisation axis along the cylinder axis, a $j \rightarrow j'$ transition includes $j + j'$ different Zeeman transitions exhibiting each of the two polarisations, σ_+ and σ_-, of the emission. As shown in II, the presence of the two corresponding degenerate transitions implies an additional interatomic interference. In this section, we study, on a simplified and somehow fictitious example, the contributions to subradiance of the two kinds of interatomic interference, on each transition separately and between different Zeeman transitions of same polarisation.

3.1 Description of the atomic configuration

In order to qualitatively estimate the role of the two kinds of subradiance in the evolution of an atomic configuration involving degenerate levels, we consider here a simple situation in which the two kinds of destructive interference are simultaneously concerned. It is the case of a $3/2 \rightarrow 1/2$ transition, when all Zeeman sublevels of the upper $3/2$ level are initially populated, $m = 3/2$ sublevel excepted (figure 1). In fact, we actually deal with a somewhat fictitious system, since we unrestrainedly choose the different probabilities of the Zeeman transitions. In addition, we let the initial populations vary independently on the realistic possibilities. In order to further simplify the problem, we assume that the transition probabilities Γ_a and Γ_b (resp. $\Gamma_{a'}$ and $\Gamma_{b'}$) are equal, that identical populations in sublevels $|1b>$ and $|1b'>$ are initially prepared, and that there is no initial coherence between these two sublevels. The initial state is thus characterised by the initial populations of the different upper states, $n_{1a'}(0) = 0$, $n_{1a}(0)$ and $n_{1b}(0) = n_{1b'}(0)$; the initial population of the upper level, $n_1(0)$, is equal to one:

$$n_1(0) = n_{1a}(0) + 2n_{1b}(0) = 1 \tag{15}$$

Both kinds of subradiance are generally expected. A "V" subradiance arises from the presence of the b subsystem (sublevels $|1b>$, $|1b'>$ and $|2b>$), whereas a "Zeeman" subradiance is due to the interference between the $|1a> \rightarrow |2a>$ and $|1b'> \rightarrow |2b>$

transitions. Whatever the transition probabilities and the initial populations be, the contribution of the "V" subradiance can be separately considered by merely cancelling the initial population of state $|1a>$: $n_{1a}(0) = 0$. The contribution of the "Zeeman" subradiance is similarly obtained by cancelling the initial population of state $|1b>$: $n_{1b}(0) = 0$ (but $n_{1b'}(0) \neq 0$). It is recalled that in the latter case the subradiance rate can beobtained directly, without solving the Bloch–Maxwell equations (II).

3.2 Influence of population rates

Semi–classical numerical calculations have been performed for various values of $n_{1a}(0)$ in the case $\Gamma_{a'} = 3\Gamma_a$, which corresponds to the real case of a $3/2 \rightarrow 1/2$ transition. We report in figure 2 the obtained subradiance rate, τ, which is equal to $n_1(\infty)$, as plotted against $n_{1b}(0)$. The other two reported quantities, τ_V and τ_Z, represent the specific influence of the "V" and "Zeeman" subradiances when isolated one from the other. τ_V (resp. τ_Z) is equal to the value of $n_1(\infty)$ one would obtain by cancelling $n_{1a}(0)$ (resp. $n_{1b}(0)$). τ_V is predicted from symmetry properties conservation to be 50 % of the initial populations of sublevels $|1b'>$ and $|1b>$ whereas τ_Z is calculated from eqq. (2.19) and (2.20) of II. When $n_{1b}(0) = n_{1b'}(0) = 0$, all the atoms are initially in sublevel $|1a>$, and the system reduces to a two–level configuration which spontaneously completely deexcites via a superradiant pulse. When $n_{1b}(0) = n_{1b'}(0) = .5$, sublevel $|1a>$ is initially empty: one obviously finds the behaviour of a "V" configuration system, and therefore a subradiance rate of 50 % of the total population. Elsewhere, the "V" and the "Zeeman" subradiance are also found to simply add their contributions.

3.3 Influence of transition probabilities

In figure 3 we report how the calculated subradiance rate depends on the transition probabilities. We consider initial populations such that $n_{1a}(0) = n_{1b}(0) + n_{1b'}(0)$, i. e. $n_{1a}(0) = .5$ and $n_{1b}(0) = n_{1b'}(0) = .25$, and we study the subradiance rate while varying the ratio $\Gamma_{a'}/\Gamma_a$. For $\Gamma_{a'} > \Gamma_a/4$, the final subradiance rate is found to be

exactly the sum of the two contributions one obtains in considering the two kinds of subradiance separately. This is not the case if $\Gamma_{a'} < \Gamma_a/4$. The two subradiance effects then interfere one with the other and, instead of increasing up to .5 while $\Gamma_{a'}$ decreases to zero, the total subradiance rate decreases monotonically to its limit value, $\tau = .25$, which is obviously due to the zero value of $\Gamma_{a'}$.

4. Atomic cascades between degenerate levels

4.1. $3/2 \rightarrow 1/2 \rightarrow 1/2$ cascade

In this section we consider the atomic cascade which has recently allowed us to give the first experimental evidence for the subradiance phenomenon. It is the case of the $4d_{3/2} \rightarrow 5p_{1/2} \rightarrow 5s_{1/2}$ cascade of gallium (figure 4). Besides the presence of a $j \rightarrow j-1$ transition, the $4d_{3/2} \rightarrow 5p_{1/2}$ transition, noted (1) $\rightarrow$ (2), on which subradiance can be expected, the gallium level configuration possesses two main remarkable features: i) the ground configuration is a p one, ii) a lower $5s_{1/2}$ level, noted (3), is connected with the $5p_{1/2}$ level.

i) The $4p$ ground configuration is made of a ground level of angular momentum $1/2$ and of a metastable level of angular momentum $3/2$, which can suitably be thermally populated. This allows the excitation of the upper level ($4d_{3/2}$) to be achieved, with either linearly or circularly polarised light, starting from either a $1/2$ level ($4p_{1/2}$) or a $3/2$ level ($4p_{3/2}$). Moreover, two different cases can be considered, in assuming that the excitation intensity is either large enough for the transition to be saturated (saturated regime), or small enough for the absorption regime to be linear (linear regime). These different possibilities of exciting the atoms in the upper $3/2$ level of the cascade provide us with the different initial states which are analysed hereafter: we will first consider the cases corresponding to a circularly polarised exciting light, when the state before excitation is a statistical mixing with equal weights of all Zeeman sublevels of either a $1/2$ or a $3/2$ level, and in the either saturated or linear regime. We then study the same initial situations but in the case of a linearly polarised exciting light.

ii) The presence of the lower level, as already indicated (§ 2.4), allows the subradiance phenomenon to be put in evidence by the occurrence of a second pulse on the upper (1) $\rightarrow$ (2) transition, the "subradiance echo". One can thus distinguish three periods in the cooperative spontaneous emission on the (1) $\rightarrow$ (2) $\rightarrow$ (3) atomic cascade. A

first superradiant pulse on the (1) → (2) transition ("pulse 1") generally leaves a certain amount of atoms in the upper level; a superradiant pulse on the (2) → (3) transition ("pulse 2") will then empty the intermediate level, thus destroying the subradiant state previously created; finally the subradiance echo will occur on the first transition. For the various initial conditions considered hereafter, we have also studied the polarisation properties of the superradiant light emitted during these three periods. For the (1) → (2) transition, the polarisation of pulse 1 is fully determined by the excitation, except in the case of (§ 4.2 c)) (Pillet 1982, Crubellier et al 1986). In the latter case, the polarisation is essentially random, and the initial polarisation has been chosen to be a linear polarisation of arbitrary polarisation direction. The same has been made in all cases for the (2) → (3) transition. Is is emphasised that the polarisation of the subradiance echo has always been obtained from the semi–classical calculations without any further assumption.

4.1.1. Excitation with circularly polarised light

We first study the case of an excitation of the atoms onto the upper 3/2 level achieved with circularly polarised light; the initial density matrix depends on the pumping level as well as from the exciting regime.

a) We consider first the cases corresponding to an excitation from a 1/2 level with, for instance, σ_- polarised light. Only the $|1a>$ $(m=-1/2)$ and $|1b'>$ $(m=-3/2)$ sublevels of the upper level are then initially populated. In the saturated (resp. linear) regime, one has $n_{1a}(0) = 1/2$ (resp. 1/4) and $n_{1b'}(0) = 1/2$ (resp. 3/4). In both cases, no subradiance is predicted from the symmetry properties of the collective state. The initial collective state is indeed found to be the product of two fully symmetrical states, corresponding to classes a and b respectively. If "Zeeman" subradiance is omitted, each of the two classes is expected to completely empty the upper level and the intermediate level one after the other, by emitting two successive superradiant pulses on the two transitions. On the other hand, the "Zeeman" subradiance rate due to the interference between the $|1a> \rightarrow |2a>$ and the $|1b'> \rightarrow |2b>$ transitions

has been calculated in II for any values of $n_{1a}(0)$ and $n_{1b'}(0)$. Semiclassical numerical calculations made for the two different excitation regimes are in perfect accord with the "Zeeman" subradiance rate r_Z given in figure 2. The three previously described periods are observed in the cooperative spontaneous emission on the (1) $\rightarrow$ (2) $\rightarrow$ (3) cascade; pulse 1 and 2 are respectively σ_+ and linearly polarised, whereas the maximum intensity of the subradiance echo, which is σ_+ polarised like the pulse 1, represents in both cases only a few per cent of the one of pulse 1.

b) We consider now the case of an excitation achieved from a 3/2 level with σ_- polarised light. Then the $|1a'>$ level is not initially populated, while one has $n_{1a}(0) = 1/3$ (resp. 4/10) and $n_{1b}(0) = n_{1b'}(0) = 1/3$ (resp. 3/10) in the saturated (resp. linear) regime. No "V" subradiance is expected for atoms of class a: this subsystem is in fact found to be represented by a symmetry type which is fully symmetrical. On the other hand, a "most antisymmetrical" symmetry type is found for the atoms of class b, namely $\{\frac{N}{3}, \frac{N}{3}\}$ (resp. $\{\frac{3N}{10}, \frac{3N}{10}\}$). A "V" subradiance rate of 33 % (resp. 30 %) is then to be expected. It has been shown in § 3.2 that, for the two particular cases considered here, the two kinds of subradiance simply add their influence, giving rise to a total subradiance rate r of about 44 % (resp. 41 %). Semiclassical calculations confirm these previsions and exhibit, once again, the three emission periods. In particular, a subradiance echo is predicted in both cases. In figure 5 we give the exemple corresponding to an σ_- polarised excitation in the saturated regime. A strongly σ_+ polarised superradiant pulse on the (1) $\rightarrow$ (2) transition is first observed, whereas the emission on the $|1b> \rightarrow |2b>$ transition is almost completely inhibited. An almost purely linearly polarised superradiant pulse on the (2) $\rightarrow$ (3) transition destroys the subradiant state previously created. A σ_- polarised subradiance echo with a maximum of about 20 % of that of pulse 1 terminates the emission. It is worthwhile to remark that the evolution does not end up with the total deexcitation of the atoms: owing to symmetry properties conservation, the final collective state, whose global symmetry type, with respect to $S_{N_a+N_b}$, is $\{\frac{N}{3}, \frac{N}{3}, \frac{N}{3}\}$

necessarily contains $\frac{N}{3}$ atoms either in the upper or in the intermediate level. It is found that the atoms of class b are prevented from emitting a second pulse on the (2) $\rightarrow$ (3) transition and 33 % of the atoms are finally left in the (2) level.

4.1.2. Excitation with linearly polarised light

Three remarkable situations are considered hereafter, which correspond to an initial state prepared with linearly polarised light. For this type of excitation, the evolution of each $|j,m>$ state is identical to the one of the $|j,-m>$ state, and the evolution of the two "Y" subsystems a and b are thus exactly the same. In particular, the two subsystems have in any case the same symmetry type and any prevision of group theory will be valid for the two atom classes.

a) In the first case we are considering here, we suppose that the excitation is achieved from a 1/2 level. With our choice of the quantisation axis, a full initial coherence between the Zeeman sublevels of $|\Delta m| = 2$ of the upper 3/2 level is obtained, and one has $n_{1a'}(0) = n_{1b'}(0) = 3/8, n_{1a}(0) = n_{1b}(0) = 1/8$. In this case, the collective state of each subsystem is fully symmetrical with regard to local permutations. After the emission on the two successive transitions, a complete deexcitation is thus predicted from group theory considerations. We have found in numerical calculations that "Zeeman" subradiance does not either occur, and the evolution simply consists in two periods of emission: as shown in figure 6, the pulse 1, which has the same polarisation as the exciting light, completely empties the upper level, and the pulse 2, which is linearly polarised too, but with an arbitrary polarisation direction, brings all the atoms in the lower level of the cascade.

b) The second considered situation corresponds to the case of an excitation which starts from a 3/2 level in the linear regime. We then find $n_{1a'}(0) = n_{1b'}(0) = 3/20, n_{1a}(0) = n_{1b}(0) = 7/20$, and a partial coherence, of modulus equal to $\frac{\sqrt{3}}{10}$, between the Zeeman sublevels of $|\Delta m| = 2$. The symmetry type corresponding to the collective state of each subsystem is $\{\frac{9N}{20}, \frac{N}{20}\}$: "V" subradiance is then predicted to inhibit 10 % of the atoms to emit. We have found in numerical calculations (figure 7)

that the "Zeeman" subradiance simply adds its contribution to the total subradiance rate, and about 30 % of the total population is then predicted to remain in the upper 3/2 level after the pulse 1, which has the same polarisation as the exciting light. After the emission of a subradiance echo, which has an orthogonal polarisation and which is found to be a few percent as compared to the pulse 1, about 10 % of the total population is still in the upper (1) level, due to Zeeman subradiance.

c) We finally consider the certainly most interesting case of an initial state prepared with a linearly polarised excitation which saturates the transition connecting a 3/2 pumping level and the upper 3/2 level of the cascade. The statistical mixing with equal weights of the lower 3/2 level is simply transferred in the upper 3/2 level of the cascade. Whatever the quantisation axis is, the corresponding initial density matrix is thus proportional to unity. The initial collective state is represented by a complete statistical mixing, with equal weights, of all the Zeeman sublevels of the upper level, and no coherence initially exists between the sublevels of $|\Delta m| = 2$. A subradiance rate τ_V of 50 % of the total population is predicted from symmetry properties conservation: for each subsystem the collective state has the "most antisymmetrical" symmetry type $\{\frac{N}{4}, \frac{N}{4}\}$, and at least $\frac{N}{2}$ atoms are therefore expected to be inhibited to emit. Figure 8 reports the time evolution of the system for these initial conditions, and assuming an initial linear polarisation for the light emitted on (1) $\rightarrow$ (2). After the emission of the pulse 1, which is therefore linearly polarised, half the atoms are left in the upper (1) level. The linearly polarised pulse 2 destroys the subradiant state spontaneously created with respect to the 3/2 $\rightarrow$ 1/2 transition. Emission on the latter transition starts then again, giving rise to a subradiance echo the polarisation of which is linear and orthogonal to that of pulse 1. The maximum intensity of this echo is about 50% of the one of the pulse 1. This spectacular effect has been recently put in evidence (Pavolini et al 1985), and the experimental results qualitatively agree with the previous numerical calculations. Due to "Zeeman" subradiance, the subradiance echo does not completely empty the upper 3/2 level: as it is shown in figure 8,

about 13 % of the atoms are left in the upper (1) level at the end of the cooperative spontaneous emission. Moreover, as expected from the antisymmetrical character of the collective state, a subradiance effect also occurs on the (2) $\rightarrow$ (3) transition, and the atoms which have emitted the subradiance echo (about 37 %) are thus prevented to further emit, and are left in the intermediate level.

4.2 $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ **cascade**

The second cascade configuration we consider here is the $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ cascade (figure 9). This was in fact the case of the first observation of superradiance (Skribanowitz et al 1973),when the excitation used the molecular P−branch. It is possible, from symmetry properties considerations, to predict a subradiance phenomenon to occur on the upper $3 \rightarrow 2$ transition, and, due to the cascade configuration, to manifest itself by a subradiance echo. Nevertheless, although the system was actually prepared in an initial state which is not fully symmetrical, the experimental observation never reported on such an echo. We give hereafter a brief description of the cooperative evolution of a system in this typical cascade configuration.

The initial state is supposed to be a full statistical mixing with equal weights of all the Zeeman sublevels of the upper $j = 3$ level. The symmetry type associated with class a (resp. b) is therefore $\{\frac{N}{7}, \frac{N}{7}, \frac{N}{7}, \frac{N}{7}\}$ (resp $\{\frac{N}{7}, \frac{N}{7}, \frac{N}{7}\}$). This means that the corresponding wavefunctions can be thought of as an ensemble of quartets (resp. trios) of atoms in a fully antisymmetrical function and that subradiance in class a (resp. b) is due to a 4−atom (resp. 3−atom) destructive interatomic interference.

We first follow the evolution of a quartet of class a atoms, as it can be predicted from symmetry properties. We note $(ijkl)$ the collective energy levels of the quartet; in this notation each index is equal to the j value of a monatomic level. The four atoms are initially in the upper $j = 3$ level, and the corresponding level of the quartet is thus (3333). One notices that the 4−atom state, being fully antisymmetrical, cannot contain more than 3 (resp. 2 and 1) atoms in the $j = 2$ (resp. $j = 1$ and $j = 0$) level. Figure 10 shows the different energy levels of the quartet, together with the cascading

transitions between these levels. The evolution, through cooperative emission, of such a quartet is of course not independent from the other ones. From a semiclassical point of view, for instance, the cooperative emission of the quartet is essentially stimulated emission due to the field radiated by all the atoms. Therefore, each pulse emitted on a $i \rightarrow j$ transition will stimulate the emission by the quartet of the maximum number of $i \rightarrow j$ photons. It will thus first emit 3 photons on $3 \rightarrow 2$ and then 2 photons on $2 \rightarrow 1$. One cannot predict the order of the last three photons which can be emitted before the quartet reaches the collective level (2110) which correspond to a subradiant final state. In any case, the emission on the $3 \rightarrow 2$ transition is first inhibited when N/7 atoms at least are still in the upper $j = 3$ level and this emission cannot further resume before a superradiant pulse on $2 \rightarrow 1$ is emitted. A subradiance echo containing N/7 photons is thus expected to be emitted by the class a atoms.

The evolution of a trio of class b is even easier to qualitatively understand. Starting from the collective level (333), it will first emit two photons on $3 \rightarrow 2$, then one photon on $2 \rightarrow 1$, and finally one photon on $3 \rightarrow 2$ again, reaching then the subradiant state associated with the (221) level. The subradiance echo is thus also expected to contain N/7 photons at least.

The interference between the different transitions of same polarisation and frequency implies that atoms of class a and atoms of class b are expected to simultaneously emit each pulse of given frequency and polarisation. The first pulse on $3 \rightarrow 2$ will thus contain 5N/7 photons; the subsequent pulse on $2 \rightarrow 1$ will then contain 3N/7 photons. We cannot know the order of the further pulses which occur on the three different transitions. We cannot in particular precisely know the temporal position of the subradiance echo, which consists of the emission of 2N/7 photons on the $3 \rightarrow 2$ transition and which ends the deexcitation of atoms towards the $j = 2$ level. The cooperative spontaneous emission will in any way terminate when the system is in a collective state which contains 3N/7 atoms in the $j = 2$ level, 3N/7 in the $j = 1$ level, and N/7 in the lowest $j = 0$ level, which is a subradiant state.

Figure 11 reports the time evolution of populations and intensities as they were calculated with the aid of Bloch–Maxwell semiclassical equations. Due to the "Zeeman" subradiance which occurs on the $3 \rightarrow 2$ and on the $2 \rightarrow 1$ transitions, the amount of atoms which are inhibited to emit on these two transitions is greater than it can be predicted by "V" subradiance only. The extreme smallness of the subradiance echo as compared to the first superradiance pulse on $3 \rightarrow 2$ can thus be partially explained as due again to "Zeeman" subradiance. In addition, we have observed that the size of the echo depends also on the polarisation of the different pulses previously emitted. The polarisation of these superradiant pulses is indeed expected to be random. The semiclassical calculations are performed (Crubellier et al 1986) in arbitrarily choosing the σ_+ and σ_- components of the vacuum field which simulate the beginning of the cooperative spontaneous emission (in the calculations of figure 11, for instance, the two modulus were chosen to be equal, whereas the relative phase was arbitrary). If, for instance, an initial σ_+ polarisation is chosen for the three fields corresponding to the three transitions (figure 12 a)), the a (resp. b) subsystem will evolve to a state where N/7 atoms are left in the Zeeman sublevel of $m = j$ (resp. $m = j - 1$) of each j level. No subradiance echo can thus take place. If, on the contrary, an initial σ_+ polarisation has been chosen for the fields correponding to $3 \rightarrow 2$ and $1 \rightarrow 0$ transitions, whereas an initial σ_- polarisation characterises the beginning of the emission on $2 \rightarrow 1$ (figure 12 b)), a subradiance echo has to be expected. In this case, the a (resp. b) subsystem will evolve to a state in which only the $m = j$ (resp. $m = j - 1$) Zeeman sublevels of the $j = 3$ and $j = 1$ levels and the $m = -j$ (resp. $m = -j + 1$) sublevel of the $j = 2$ level are populated. A σ_- polarised subradiance echo can then take place on the $3 \rightarrow 2$ transition and completely empty the upper $j = 3$ level. A more detailed analysis of the polarisation properties of the superradiant light in this case will be given elsewhere.

5. Conclusion

This paper, the fourth of our theoretical study on subradiance, is devoted to the analysis of realistic cases in which the phenomenon can be expected to occur. Dipole transitions between degenerate levels and pencil–shaped samples have only been considered. Two kinds of results are presented.

Qualitative results are first obtained from rather simple and intuitive considerations about the symmetry of the collective state with regard to the atomic permutations. These symmetry properties determine, like in a pair of two–level atoms (Dicke 1954), the either constructive or destructive nature of the interatomic interference. We have shown, in particular, how a subradiant state may be predicted to spontaneously be reached after the emission of a superradiant pulse. For this aim, we have generalised in the case of n–level atoms the link between statistical mixing and permutation symmetry which was established, in a different manner and for three–level atoms only, in the first paper of the sequence. We recall here that the present derivation quite generally applies to a large number of identical and uncorrelated multilevel atoms. A qualitative destription of the evolution, again based on symmetry properties, allows us to show that the mere observation of the sequence of pulses emitted on a $j \rightarrow j-1 \rightarrow j'$ cascade can reveal unambiguously the phenomenon of subradiance. This is the method we actually used to give the first experimental evidence for subradiance (Pavolini et al 1985), by observing superradiant emission on the $4d_{3/2} \rightarrow 5p_{1/2} \rightarrow 5s_{1/2}$ cascade of atomic gallium. This method does in particular avoid the delicate measure of both the numbers of initially excited and of finally deexcited atoms which would directly prove the inhibition of the emission.

Quantitative results have otherwise been obtained, within the semiclassical formalism (McGillivray and Feld 1976), by numerically solving the Maxwell–Bloch equations, the general form of which has already been given in the case of a $j \rightarrow j'$ transition (Pillet 1982, Crubellier et al 1986). As shown in the second paper of the sequence, the presence of degenerate transitions involves a second kind of interatomic

interference, between the different Zeeman transitions of same polarisation, which is completely ignored in the qualitative description based on symmetry properties. Studying first a suitably simplified configuration, we have found that the two kinds of interference most often simply add their influence on the evolution of the system. We finally analyse two particular experimental situations. The first one is the case of a $3/2 \rightarrow 1/2 \rightarrow 1/2$ cascade, on which we performed the first observation of subradiance. A number of initial conditions are studied, for which the respective role of "V" subradiance and of "Zeeman" subradiance is again analysed. Emphasis is of course given to the interpretation of the subradiance experiment. The second case is a $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ cascade, with an initial complete statistical mixing of all Zeeman sublevels of the $j = 3$ upper level. Such an experimental situation was actually encountered in the HF experiment (Skribanowitz 1972, Skribanowitz et al 1973, Herman et al 1974), which provided the first observation of superradiance, when the exciting light was tuned at the wavelength of the molecular P−branch. In this case, the symmetry conservation predicts a subradiance echo on the first transition, the total energy of which should be 2/5 of the one of the first pulse. However, it is found that the size of this subradiance echo is lessered by the "Zeeman" subradiance and moreover that it greatly depends on the polarisation of the two previously emitted pulses. This could explain why no subradiance echo has ever been mentioned in the HF experiment.

The numerical simulations show a polarisation of the subradiance echo which is always orthogonal to the one of the first pulse. This result is in fact quite general, and closely related to the antisymmetry of the collective state. A more detailed analysis will be given elsewhere and the corresponding experimental study is presently in progress.

As pencil−shaped samples are here considered, the cooperative inhibition of spontaneous emission only holds in the so−called "end−fire" cooperative modes. It is shown in the third paper of the sequence that in "small samples", i. e. atomic sam-

ples with dimensions smaller than the emitted wavelength, the inhibition concerns all the field modes. Provided that the dephasing effects remain small enough, the complete deexcitation of the atoms should then actually spend a time larger than the spontaneous lifetime: this phenomenon still remains to be checked; no suitable experimental situation has yet been proposed.

FIGURE CAPTION

Fig. 1 Zeeman sublevels of a $3/2 \rightarrow 1/2$ transition.

Fig. 2 Final population $n_1(\infty)$ of the upper level plotted against the initial population $n_{1b}(0)$ of the $|1b>$ sublevel (in the case $n_{1a'}(0) = 0$, $n_{1b}(0) = n_{1b'}(0)$ and $\Gamma_a = \Gamma_b = \Gamma_{a'}/3 = \Gamma_{b'}/3$).

Fig. 3 Final population $n_1(\infty)$ of the upper level plotted against $\eta = tan^{-1}(\sqrt{\Gamma_{a'}/\Gamma_a})$ (in the case $n_{1a}(0) = .5$, $n_{1b}(0) = n_{1b'}(0) = .25$).

Fig. 4 a and b level–subsystems of a $3/2 \rightarrow 1/2 \rightarrow 1/2$ cascade.

Fig. 5 Semiclassical results for a $3/2 \rightarrow 1/2 \rightarrow 1/2$ cascade, in the case of an excitation achieved from a $3/2$ level with σ_- circularly polarised light in the "saturated" regime. The upper set of curves represents the time evolution of the intensity of the light emitted on both transitions with σ_+ and σ_- polarisations (——, $3/2 \rightarrow 1/2$, σ_+; – – –, $3/2 \rightarrow 1/2$, σ_-; — —, $1/2 \rightarrow 1/2$, σ_+; —•, $1/2 \rightarrow 1/2$, σ_-); the lower set of curves represents the time evolution of the average populations of the three levels (——, upper level; – – –, intermediate level; —•, lower level).

Fig. 6 Semiclassical results for a $3/2 \rightarrow 1/2 \rightarrow 1/2$ cascade, in the case of an excitation achieved from a $1/2$ level with linearly polarised light. The upper set of curves represents the time evolution of the intensity of the light emitted on both transitions, which is always linearly polarised (——, $3/2 \rightarrow 1/2$; – – –, $1/2 \rightarrow 1/2$); the lower set of curves represents the time evolution of the average populations of the three levels (——, upper level; – – –, intermediate level; —•, lower level).

Fig. 7 As figure 6 but for an excitation achieved from a $3/2$ level with linearly polarised light in the linear regime.

Fig. 8 As figure 6 but for an excitation achieved from a $3/2$ level with linearly polarised light in the saturated regime.

Fig. 9 Zeeman sublevels of a and b subsystems of a $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ cascade (a subsystem: ● and $\dashrightarrow$, b subsystem: × and $\longrightarrow$).

Fig. 10 Energy levels $(ijkl)$ (resp. (ijk)) of an antisymmetrical "quartet" (resp. "trio") of class a (resp. b) atoms; the different arrows correspond to the emission on the different transitions ($\longrightarrow$: $j = 3 \rightarrow j = 2$, $\Longrightarrow$: $j = 2 \rightarrow j = 1$, $\blacktriangleright$: $j = 1 \rightarrow j = 0$).

Fig. 11 Semiclassical results for a $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ cascade, in the case of a complete initial statistical mixing with equal weights of all upper Zeeman sublevels and in the case of respective linear initial polarisations u, u′, u (with u orthogonal to u′) for the $3 \rightarrow 2$, $2 \rightarrow 1$ and $1 \rightarrow 0$ transitions. The upper set of curves represents the time evolution of the intensity of the light radiated on the three transitions, which is always linearly polarised (——, $3 \rightarrow 2$; – – –, $2 \rightarrow 1$; — · —, $1 \rightarrow 0$). The lower set of curves represents the time evolution of the average populations of the four levels (——, $j = 3$; – – –, $j = 2$; — · —, $j = 1$; — —, $j = 0$).

Fig. 12 Schematic representation of superradiant emission on a $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ cascade in two particular cases. In case (a) (resp. (b)) the emission on the three successive transitions is assumed to be initially σ_+, σ_+ and σ_+ (resp. σ_+, σ_- and σ_+) polarised. The sublevels which remain populated after the emission of the corresponding three pulses are circled: in case (a) the corresponding state is subradiant whereas in case (b) a subradiance echo can be expected.

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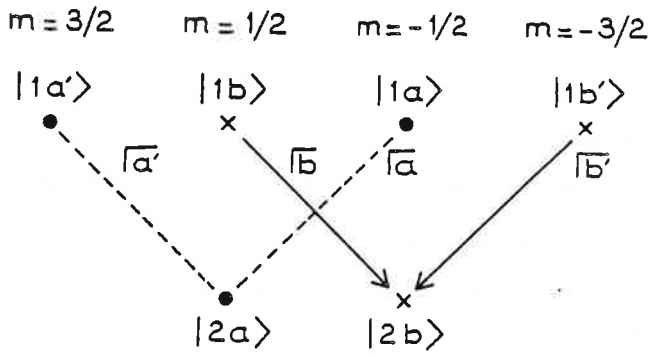
figure 1



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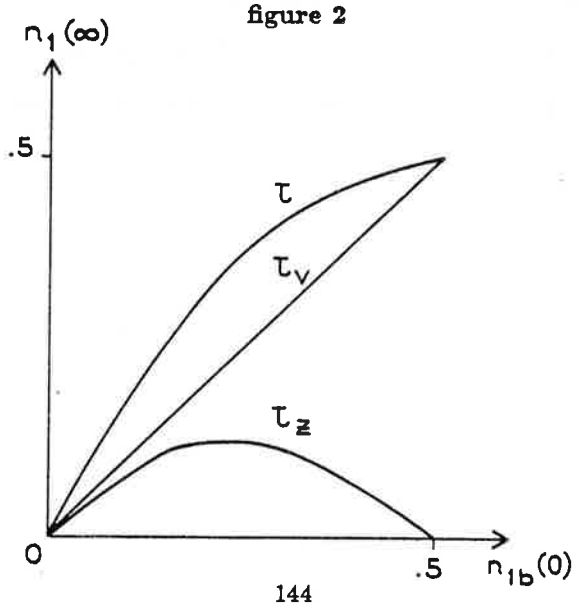

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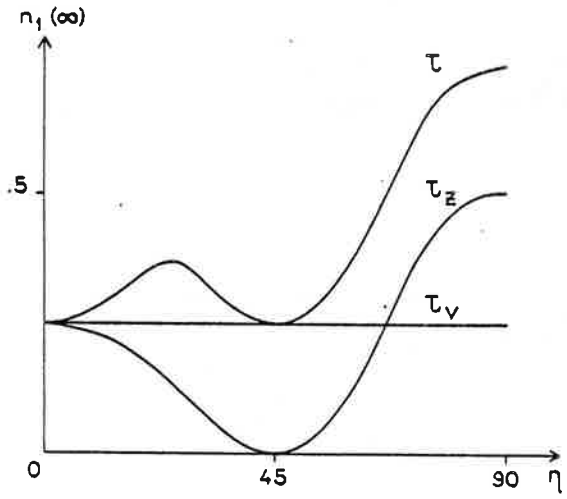

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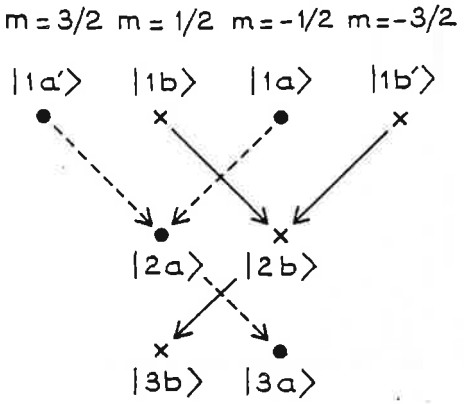

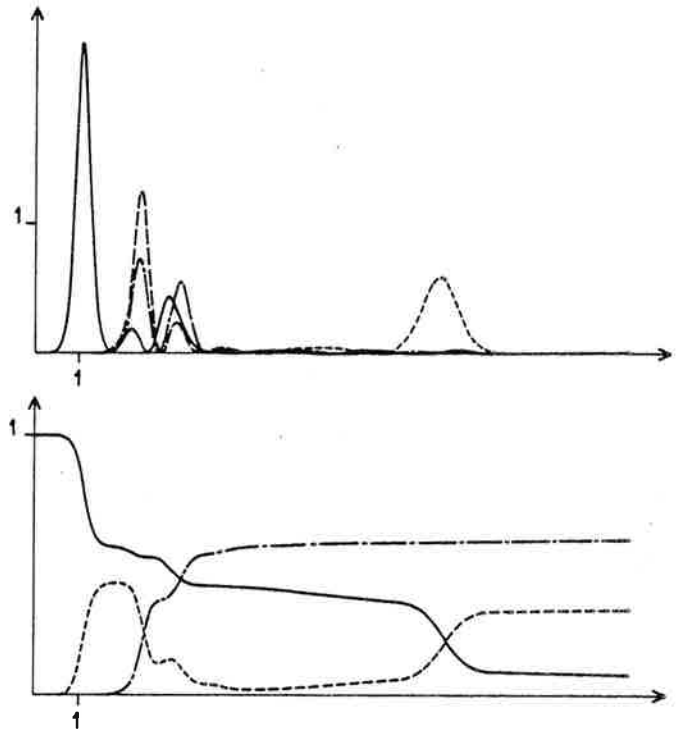

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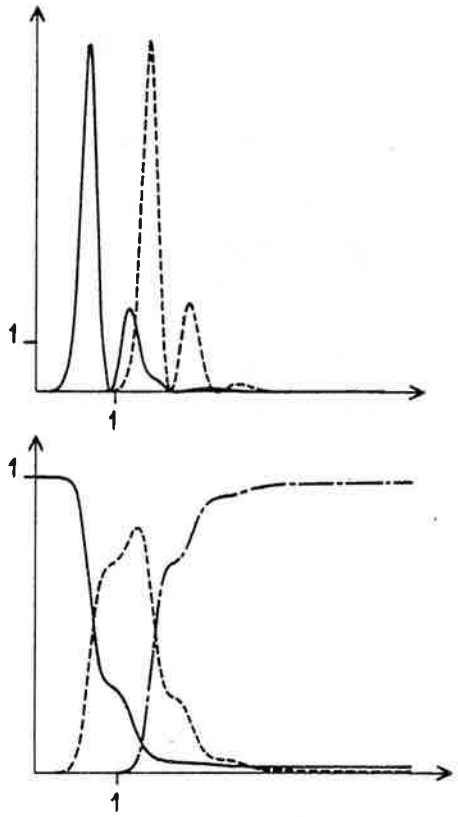

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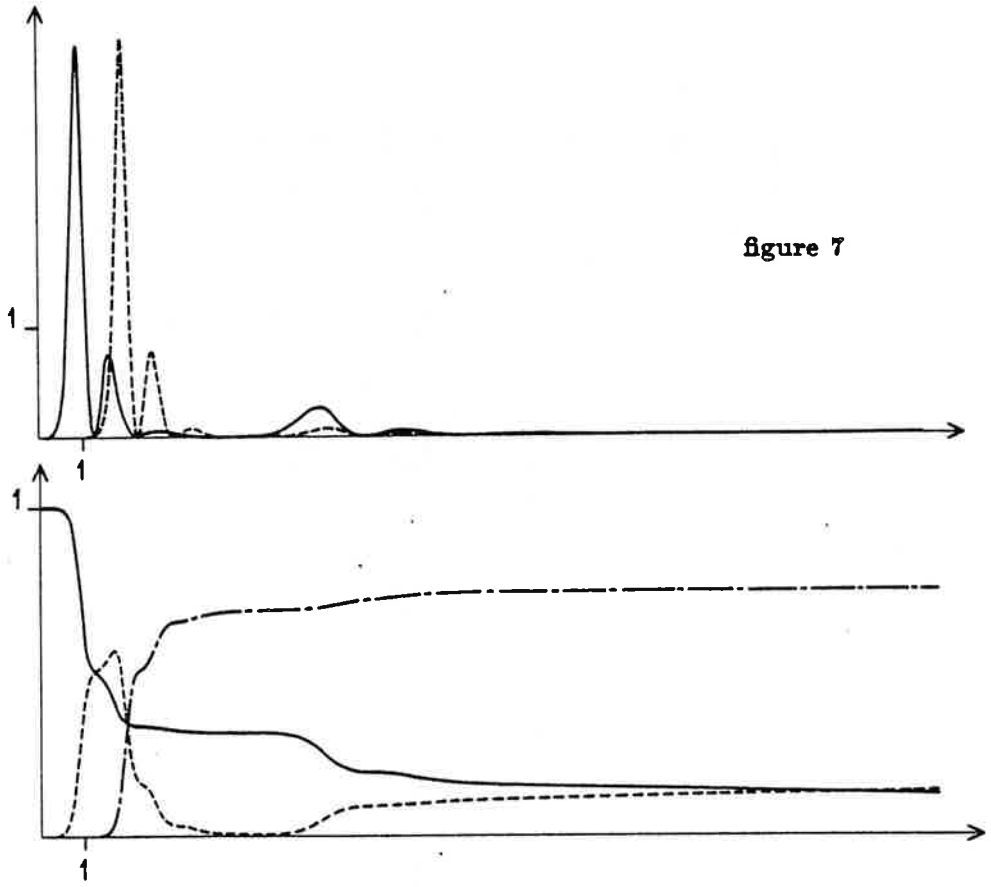

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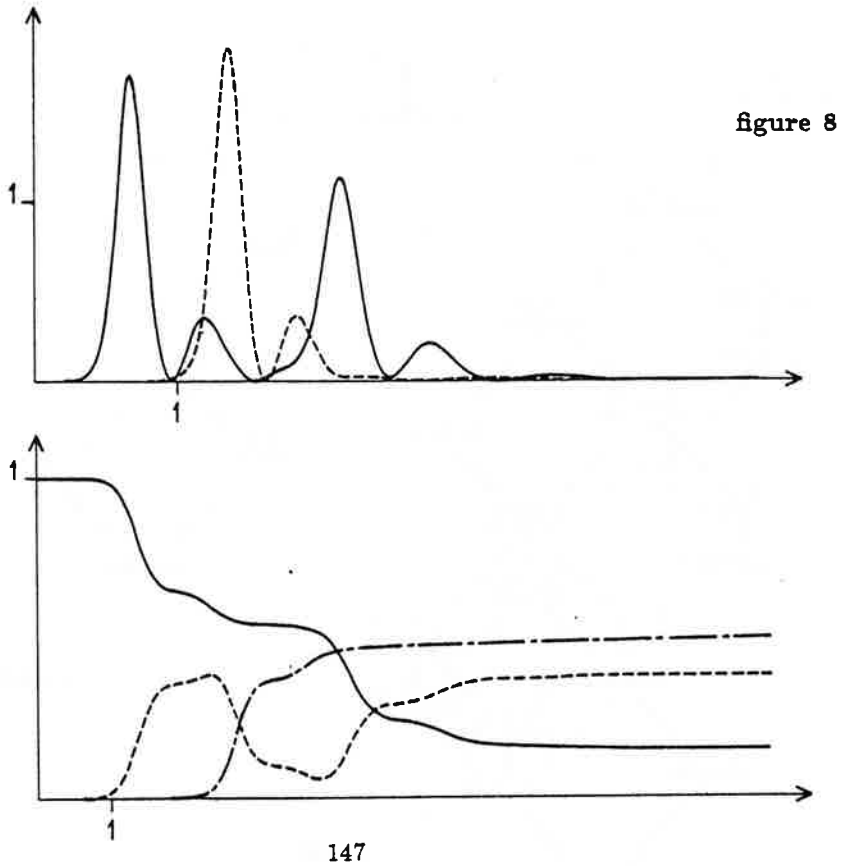

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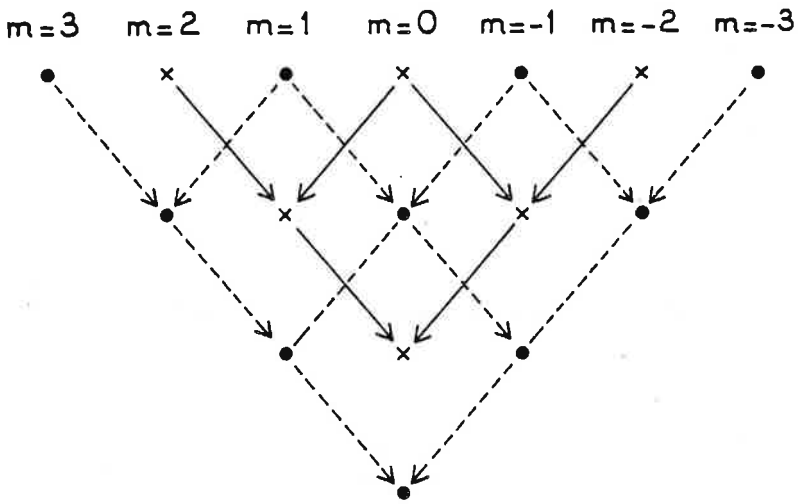

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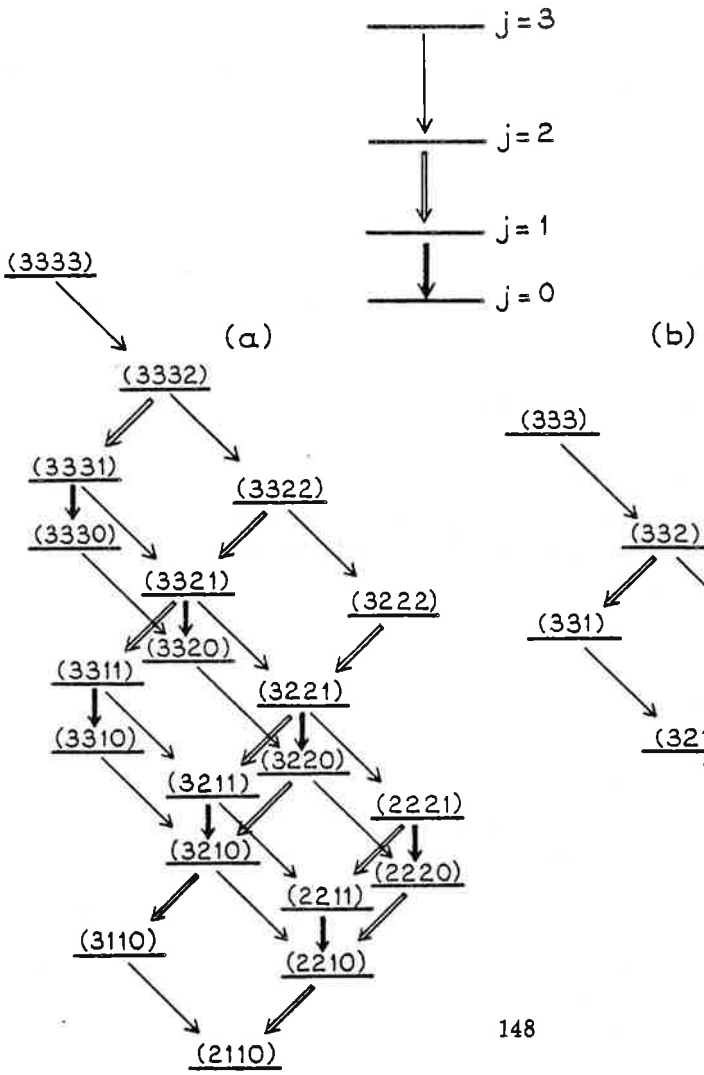

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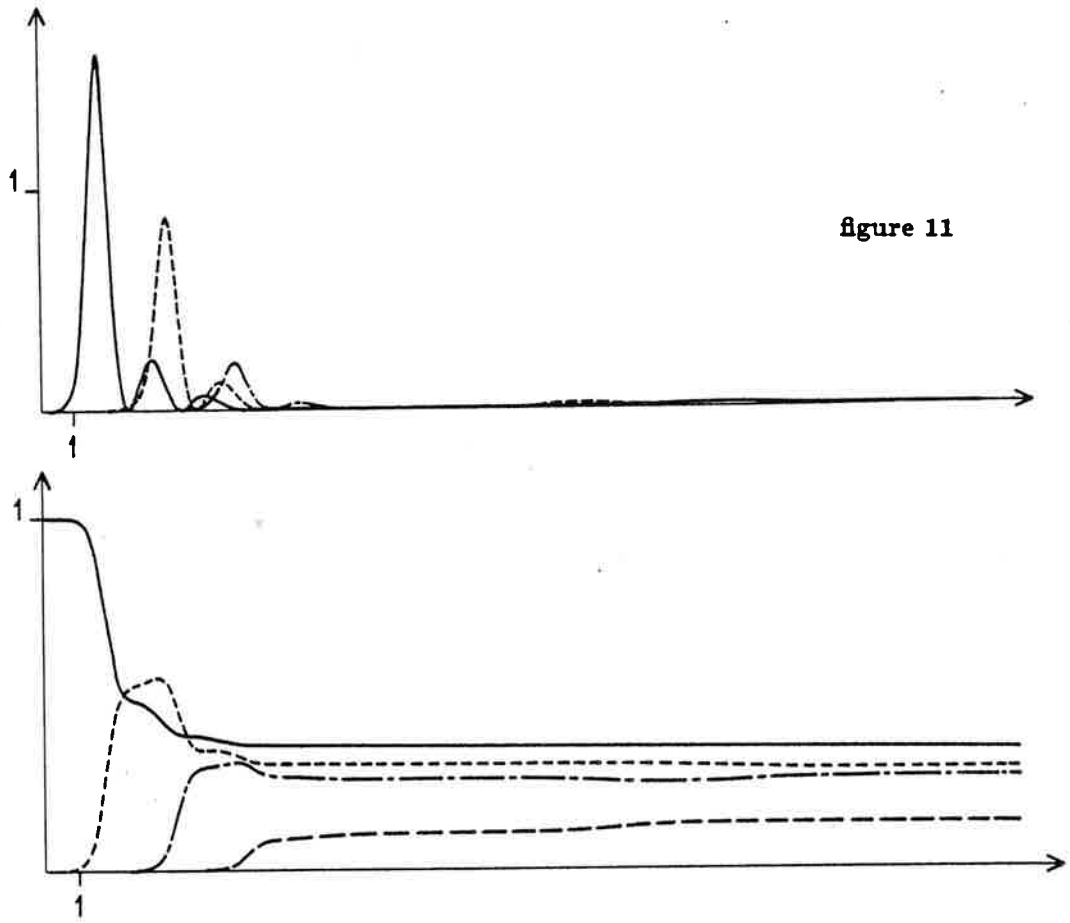

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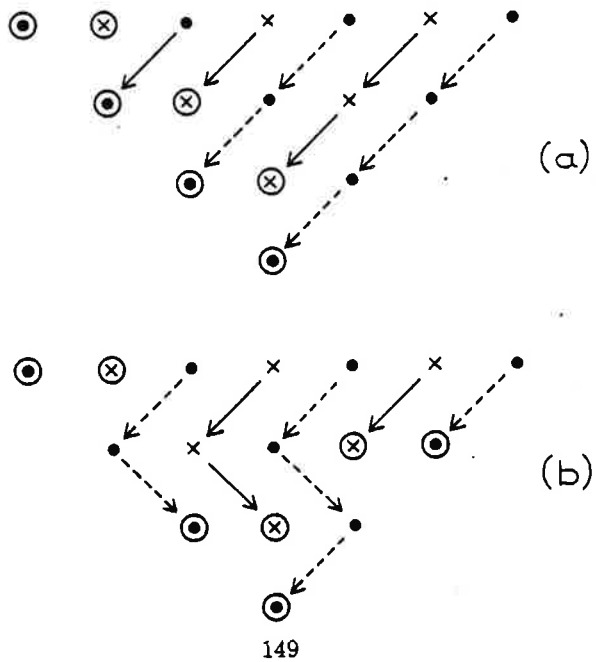


figure 12

6. MISE EN EVIDENCE EXPERIMENTALE DE LA SUBRADIANCE

6.1. Le système de niveaux du gallium

Pour mettre en évidence le phénomène de subradiance, le système choisi a été celui du gallium (figure 7). Ce système possède en effet plusieurs avantages. Le niveau fondamental est le niveau $4p_{1/2}$; le niveau $4p_{3/2}$, métastable, se trouve à environ $800cm^{-1}$ du fondamental $4p_{1/2}$, et est convenablement peuplé thermiquement: pour des températures de l'ordre de 1300 K, typiquement, 30 % des atomes se trouvent dans ce niveau. On parvient alors en particulier à atteindre, avec un pompage laser linéairement polarisé, deux situations initiales remarquables.

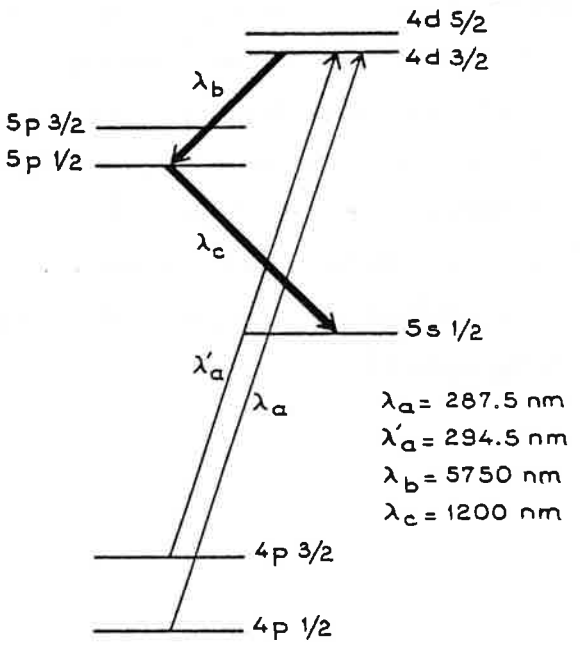


figure 7: niveaux profonds du gallium

Quand le peuplement est fait à partir du fondamental $4p_{1/2}$ et que l'on choisit comme axe de quantification la direction de la polarisation du laser, seuls les états de $m = \pm 1/2$ sont ainsi peuplés. Si ensuite on place l'axe de quantification dans la direction de propagation de la lumière superradiante, c'est à dire suivant l'axe de symétrie du cylindre, on s'aperçoit facilement que ceci entraîne une cohérence complète entre les sous–niveaux Zeeman de $|\Delta m| = 2$ du niveau $4d_{3/2}$. En revanche, dans le cas où le pompage a lieu à partir du métastable $4p_{3/2}$, le mélange statistique existant initialement dans ce dernier niveau est simplement transféré dans le niveau supérieur: la matrice densité monoatomique est donc initialement représentée par un mélange statistique complet à poids égaux de tous les sous–niveaux Zeeman du niveau supérieur. Par ailleurs, lorsque les conditions de coopérativité sont remplies, l'émission sur la transition $4d_{3/2} \rightarrow 5p_{3/2}$, transition qui est 5 fois moins probable que sa rivale $4d_{3/2} \rightarrow 5p_{1/2}$, devient négligeable par rapport à cette dernière: les atomes "choisissent" coopérativement l'émission sur l'autre transition. On est donc dans le cas d'une transition $j \rightarrow j - 1$, sur laquelle, avec des conditions initiales opportunes, on prévoit un phénomène de subradiance. Finalement, la présence du niveau inférieur $5s_{1/2}$ de la cascade permet, la mise en évidence de l'éventuel état subradiant précédemment créé, suivant le processus décrit dans le paragraphe 4.2.2.

6.2 Montage expérimental

Le gallium a une tension de vapeur très basse jusqu'à des températures très élévées (10^{-4} Torr pour 1300 K). Il est donc évaporé dans un four en graphite (figure 8) qui permet de monter suffisamment haut en température (~ 2000 K) en faisant circuler dans la masse du four un courant très important (~ 350A). Le four est fermé par un couvercle muni d'une fente de 30 mm de longueur et 0,5 mm de largeur, ce

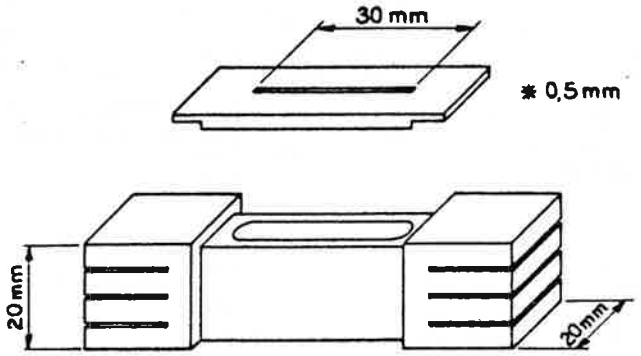


figure 8: schéma du four

qui permet d'évaporer le gallium en une espèce de "multijet" atomique, non collimaté. La lumière excitatrice (figure 9) est fournie par un laser à colorant pompé par un laser à azote et sa fréquence est doublée dans un cristal A.D.A. Le colorant est la rhodamine 590 pour le pompage à partir du métastable (294,5 *nm*), et un mélange de rhodamines 590 et 610 pour l'excitation à partir du fondamental (287,5 *nm*). Avec des lentilles de distances focales convenablement choisies, on arrive à obtenir un faiseau laser bien collimaté qui est focalisé dans la zone où il traverse le multijet, parallélement à la fente du four et à une distance d'environ 2 mm. Ceci nous permet d'être dans les conditions où la transition excitatrice est saturée et d'appliquer l'approximation

des ondes planes: notre échantillon a bien la forme d'un cylindre allongé, avec $L = 30mm$, $a^2 = 0,008mm^2$ environ, et les nombres de Fresnel relatifs aux deux transitions étudiées sont de l'ordre le l'unité: $\mathcal{F}_{4d_{3/2}\to 5p_{1/2}} \simeq 0,5$ et $\mathcal{F}_{5p_{1/2}\to 5s_{1/2}} \simeq 2$. Les signaux correspondant aux deux

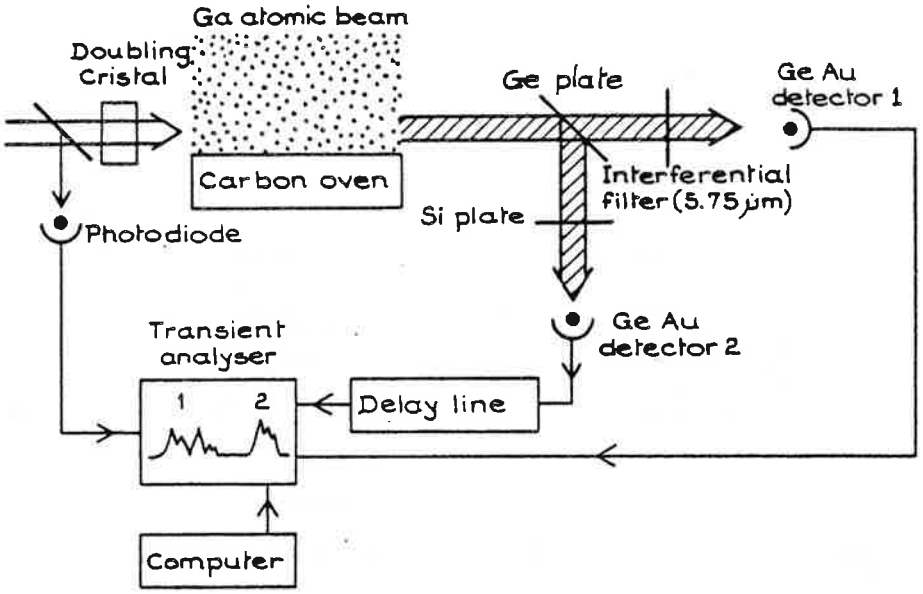


figure 9: montage expérimental

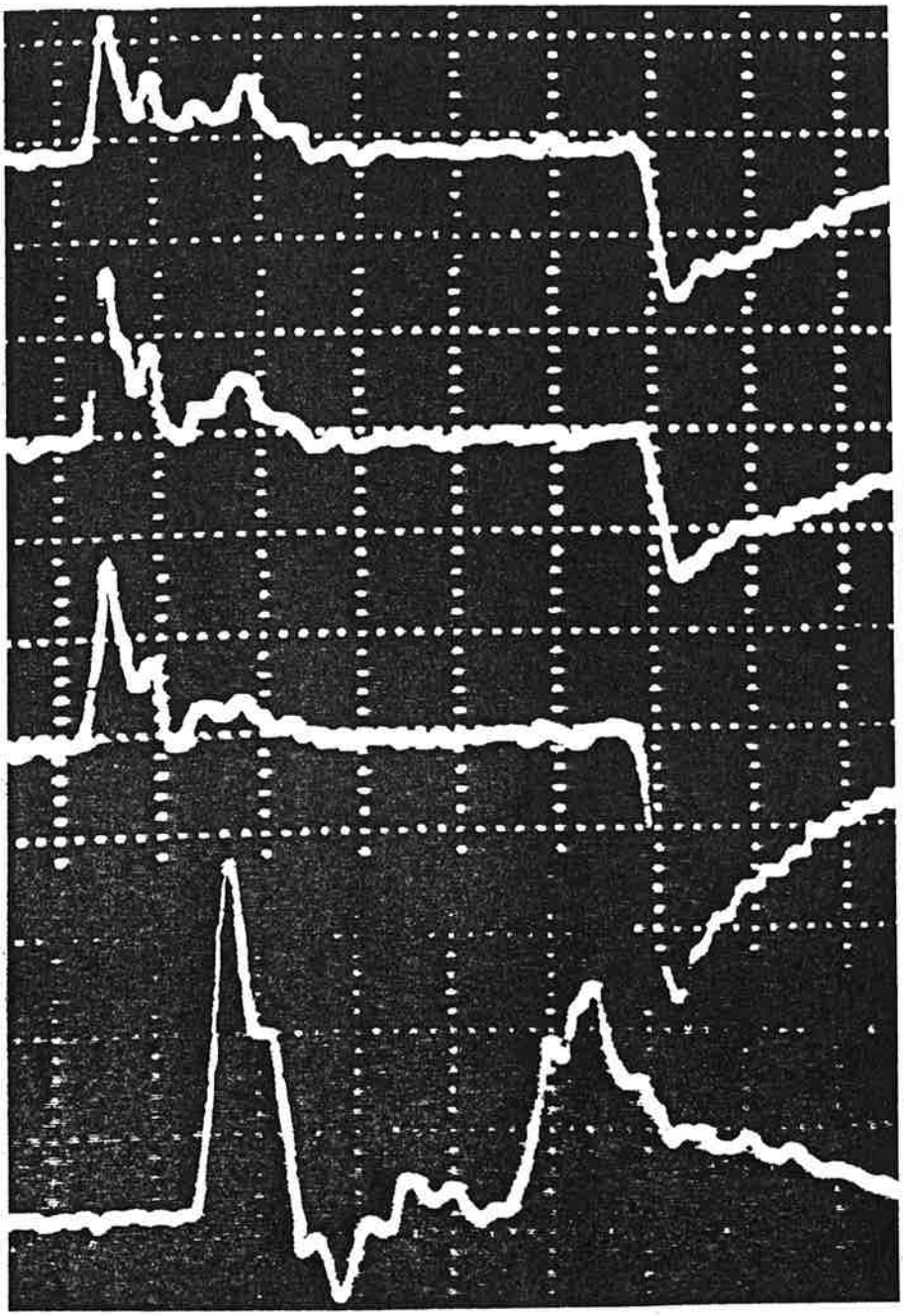
transitions $4d_{3/2} \to 5p_{1/2}$ et $5p_{1/2} \to 5s_{1/2}$ sont recueillis séparément par deux détecteurs germanium–or identiques, de temps de réponse 2ns, dans la direction de propagation du laser. Ceci est rendu possible à l'aide d'une lame de germanium qui laisse passer l'impulsion superradiante sur la transition $4d_{3/2} \to 5p_{1/2}$ $(5,75\mu)$. La lumière refléchie passe à travers une lame de silicium qui empêche le signal ultraviolet du laser d'arriver au détecteur. Les signaux sont visualisés et enregistrés par un

analyseur de transitoires, et traités par un micro-ordinateur.

6.3 Mise en évidence expérimentale de la subradiance

On donne dans la figure 10 quelques signaux enregistrés typiques. Dans le cas où on a préparé un mélange statistique complet à poids égaux des sous–niveaux Zeeman du niveau supérieur, c'est à dire lorsque l'excitation démarre du niveau $4p_{3/2}$, on observe deux impulsions successives sur la transition $4d_{3/2} \rightarrow 5p_{1/2}$. Ceci n'est jamais le cas lorsque le niveau supérieur est initialement peuplé à partir du fondamental $4p_{1/2}$, ce qui entraîne une cohérence initiale complète entre les sous–niveaux Zeeman de $|\Delta m| = 2$ du niveau $4d_{3/2}$. Les oscillations des impulsions sur la transition $4d_{3/2} \rightarrow 5p_{1/2}$ sont très probablement dues à la structure hyperfine du niveau $5p_{1/2}$. La valeur calculée de cette structure est, pour l'isotope ^{69}Ga, de 278 MHz, ce qui correspond à 3,6 nm environ (Jönsson et al 1985); malheureusement, aucune étude expérimentale n'a jusqu'à présent confirmé cette valeur. Dans l'article présenté ci–après, "Experimental evidence for subradiance" (voir aussi Crubellier et al 1985), on donne le compte rendu de la première mise en évidence expérimentale de la subradiance, avec les prévisions théoriques et les calculs semi–classiques accomplis dans les cas correspondants aux situations expérimentales étudiées, qui confirment qualitativement les résultats obtenus.

figure 10: signaux typiques. Les trois traces supérieures (resp. la trace inférieure) correspondent à une excitation partant du niveau $4p_{3/2}$ (resp. $4p_{1/2}$). Les signaux reçus par les deux détecteurs, qui correspondent à la lumière émise sur les deux transitions $4d_{3/2} \rightarrow 5p_{1/2}$ et $5p_{1/2} \rightarrow 5s_{1/2}$ sont sommés après qu'on ait retardé le second signal de 28ns (resp. 15ns). L'échelle est telle que 1 carreau= 5ns.



Experimental Evidence for Subradiance

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(Received 11 February 1985)

The first observation of the subradiance phenomenon is reported. As previously predicted, destructive interatomic interference may prevent cooperative deexcitation of a collection of atoms from being complete. In the case of a $j=\frac{1}{2}\rightarrow j'=\frac{1}{2}$ transition, for example, half the atoms remain excited when the initial atomic state is a full statistical mixture with equal weights of all Zeeman sublevels of the upper level. The experiment has been realized with gallium atoms. The results agree with semiclassical calculations.

PACS numbers: 32.50.+d, 32.80.−t

The origin of superradiant emission lies in constructive interatomic interference. The possibility of destructive interference was already indicated in the original paper of Dicke,[1] in the example of two neutrons in a uniform magnetic field. If the conditions for cooperative emission are fulfilled, a pair of two-level systems cannot radiate when it is in the antisymmetrical state, the so-called subradiant state, which still contains one excited system. In the same way, spontaneous emission of a collection of atoms can be inhibited or quenched by destructive interatomic interference, while the atoms are not all deexcited. This phenomenon is called subradiance. In the case of two-level atoms, although most of the theoretical models of cooperative spontaneous emission could account for this effect, subradiance is explicitly mentioned by Freedhof and van Kranendonk[2] and Stroud *et al.*[2] only. Moreover, the many-atom subradiant states, which can simply be considered as ensembles of pairs of atoms in antisymmetrical states, are neither easy to create nor to observe: This is probably why subradiance has not been experimentally studied so far. Recent studies of problems involving level degeneracy[3,4] and polarization[5] in superradiance have led us to consider the cases of many-level atoms, which open new possibilities for the observation of subradiance. In the case of three-level atoms in the V configuration (two transitions sharing a common lower level),[6] partially antisymmetrical collective states evolve spontaneously, through cooperative emission, to subradiant states. Furthermore it has been shown that any statistical mixing of the two upper levels provides an example of such states. As a generalization of these theoretical results, we have pointed out that in the case of a $j\rightarrow j-1$ transition between degenerate levels, incomplete deexcitation due to subradiance is predicted for a collection of initially excited atoms when the "amount" of statistical mixing is large enough.[7,8] For a $\frac{1}{2}\rightarrow\frac{1}{2}$ transition, for instance, half the atoms are expected to remain excited when the initial atomic state is a full statistical mixture (diagonal density matrix) with equal weights of all Zeeman sublevels of the upper level. This paper reports the first experimental evidence for subradiance, on such a $\frac{1}{2}\rightarrow\frac{1}{2}$ transition.

Since subradiant states are characterized by an inhibition of the emission, they cannot be simply observed, unless destroyed for instance, by superradiant emission from the lower level toward another level. To make manifest the existence of subradiant states, we have chosen to study the cooperative spontaneous emission on the $4d_{3/2}\rightarrow 5p_{1/2}\rightarrow 5s_{1/2}$ cascade of gallium atoms (Fig. 1). When the atoms are initially excited in the $4d_{3/2}$ level starting from the thermally populated metastable $4p_{3/2}$ level, a first pulse is expected to deexcite the system incompletely, because of subradiance on the $4d_{3/2}\rightarrow 5p_{1/2}$ transition. Moreover, cascading superradiant emission is predicted to occur on the $5p_{1/2}\rightarrow 5s_{1/2}$ transition and then to break the subradiant state. A second pulse, the "subradiance echo," is thus expected on the first transition. Exciting the same $4d_{3/2}$ level from the $4p_{1/2}$ ground level provides a situation in which no subradiance is expected at all.

In our experimental arrangement, a nitrogen-pumped dye laser is amplified and frequency doubled in a thermally stabilized ammonium dihydrogen arsenate crsytal. The peak power of the exciting light is about 100 W, and is large enough to saturate the atomic transition; its duration is 5 ns and its beam waist is $r=0.2$ mm, in the interaction region. The exciting light is tuned to either 287.5 nm ($4p_{1/2}\rightarrow 4d_{3/2}$) or

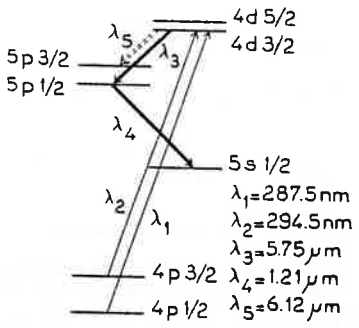

FIG. 1. Relevant level diagram of gallium.

294.5 nm ($4p_{3/2} \rightarrow 4d_{3/2}$). Gallium atoms are evaporated through the slot (20 mm length, 0.5 mm width) of a carbon oven. The Fresnel numbers of the pencil-shaped active volume corresponding to the two relevant wavelengths (λ_3 and λ_4) are of the order of a few units. The laser beam propagates 2 mm above the oven slot. The oven temperature is typically 1700 K. A rough estimate of the density of gallium atoms in the interaction region is about 10^{12} atoms/cm^3. The corresponding characteristic time for cooperative emission on the transition at λ_3 (λ_4) is 0.04 ns (0.02 ns).[9] The relative population of the $4p_{3/2}$ metastable level is 30% at 1700 K. The light emitted in the direction of propagation of the exciting light hits a germanium plate, which is fully reflective for wavelengths shorter than 2 μm and transparent for longer wavelengths. The pulses emitted on $4d_{3/2} \rightarrow 5p_{1/2}$ (5.75 μm) and on $5p_{1/2} \rightarrow 5s_{1/2}$ (1.21 μm) are detected, through suitable filters, by two GeAu photoconducting cells whose response time is about 2 ns. It has been verified, with an interferential filter, that cooperative emission never occurs on $4d_{3/2} \rightarrow 5p_{3/2}$ (6.12 μm); because of its 5 times weaker transition probability, this emission is quenched[4] by the cooperative emission on the competing $4d_{3/2} \rightarrow 5p_{1/2}$ transition. Signals are dealt by a transient digitalizer and a computer.

Typical superradiant signals are displayed in Fig. 2. In Fig. 2(a), trace *a* exhibits two separate pulses on $4d_{3/2} \rightarrow 5p_{1/2}$, emitted before and after the pulse on $5p_{1/2} \rightarrow 5s_{1/2}$ (trace *b*). The second pulse on the first transition indicates that the $4d_{3/2}$ remained populated after the emission of the first pulse; such a feature gives experimental evidence for the existence of an intermediate subradiant state. In contrast, no second pulse on the first transition has ever been observed in the case of Fig. 2(b). Characteristic features of superradiant pulses (in particular, delay times and fluctuations) have been observed in all cases.

The results of semiclassical calculations for a $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ cascade, using the plane-wave approximation[10] and including level degeneracy, are shown in Fig. 3 and confirm the experimental results. Case (a) corresponds to an initial full statistical mixture with equal weights of all Zeeman sublevels of the $\frac{3}{2}$ upper level, that is, to an excitation with saturating linearly polarized light from a $\frac{3}{2}$ level; case (b) corresponds to an excitation with linearly polarized light from a $\frac{1}{2}$ level. By choice of the quantization axis along the direction of propagation of the exciting light, it is very easy to see that, contrary to case (a), full initial coherence between the pairs of Zeeman sublevels with $|\Delta m| = 2$ of the $\frac{3}{2}$ upper level is obtained in case (b). The time evolution of the populations in Fig. 3(a) clearly shows that the $\frac{3}{2}$ level remains populated as long as the emission on the $\frac{1}{2} \rightarrow \frac{1}{2}$ transition does not occur.[11] The emission on the $\frac{3}{2} \rightarrow \frac{1}{2}$ transition exhibits two distinct periods, with ringing oscillations,[10] well separated by a period of nonemission. Coherence between the Zeeman sublevels with $|\Delta m| = 2$ of the upper level, initially equal to zero, tends to be total. Figure 3(b) shows that the upper $\frac{3}{2}$ level is emptied after a single emission period on the $\frac{3}{2} \rightarrow \frac{1}{2}$ transition. $|\Delta m| = 2$ Zeeman coherences remain total during the whole evolution.

Comparison between cases (a) and (b) shows that the temporary trapping of photons which is at the origin of the "subradiance echo" depends upon the "amount" of statistical mixing in the initial state. This can be understood, in the semiclassical model, from the conservation property of the trace of the

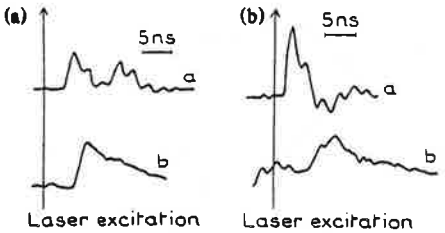

FIG. 2. Typical superradiant signals. (a) corresponds to the excitation with saturated linearly polarized light from the $4p_{3/2}$ level; (b), the $4p_{1/2}$ level. In both cases, traces *a* and *b* correspond respectively to $4d \rightarrow 5p$ and to $5p \rightarrow 5s$ transitions. The visible oscillations of the signals are most likely due to the hyperfine structure of the $5p_{1/2}$ level.

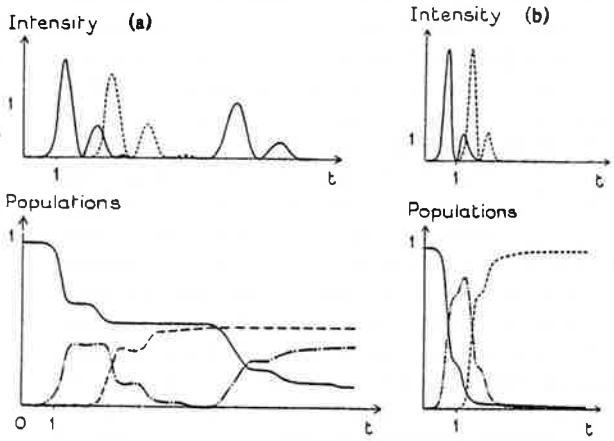

FIG. 3. Results of semiclassical calculations of cooperative spontaneous emission on a $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ cascade. In both cases (a) and (b) the upper curves show the total intensity radiated on $\frac{3}{2} \rightarrow \frac{1}{2}$ (solid line) and on $\frac{1}{2} \rightarrow \frac{1}{2}$ (dashed line); the lower curves show the time evolution of the populations of the $\frac{3}{2}$ upper level (solid line), of the $\frac{1}{2}$ intermediate level (dot-dashed line), and of the $\frac{1}{2}$ lower level (dashed line). The (arbitrary) time and intensity units are the same in both cases.

square monatomic density matrix.[6] However, these considerations give no physical understanding of the inhibition of cooperative emission. The physical origin of this phenomenon is in fact quite naturally found in destructive interatomic interference. As in the two two-level atom case, the constructive or destructive nature of the interatomic interference in cooperative spontaneous emission of a collection of many-level atoms is fixed by the (conserved) symmetry properties of the collective state with respect to the permutations of the undistinguishable atoms.[6,8] We can then interpret all features of the evolution of the atomic system by studying the symmetry properties of the initial collective state.

As shown in Fig. 4, the atomic level configuration consists in two four-level systems in the "Y" configuration. If the quantization axis is taken along the axis of the atomic sample, superradiant emission occurs in σ_+ and σ_- polarizations only and, therefore, inside two distinct sublevel systems, a and b, which are only coupled one to the other by the interference between the Zeeman transitions of the same polarization.[7,8,12] The atoms therefore form two classes, according to whether they are initially in states a or in states b, and no atom exchange between the two classes occurs during the cooperative spontaneous evolution of the system.[13] The number of locally undistinguishable atoms in the two classes, N_a and N_b, remains thus constant. The relevant conserved symmetry properties of the collective state are those with respect to the groups $\mathcal{S}_{N_a}$ and $\mathcal{S}_{N_b}$ of local permutations among atoms of a same class.[14] For initially uncorrelated atoms, these properties can be obtained in exactly the same way as in the case of three-level atoms[6] and they depend simply upon the amount of statistical mixing.[8] The initial collective state in case (a) is found to be a statistical mixture of collective states which all have the same symmetry properties. The actual spontaneous evolution of the system is the same if the initial state was any one of these collective states, i.e., any of the "most antisymmetrical" collective states. Such a state contains $N/4$ pairs of atoms in an antisymmetrical state formed with $|1a\rangle$ and $|1a'\rangle$ monatomic states, denoted as $|1a, 1a'\rangle_{AS}$, and $N/4$ analogous pairs corresponding to the b-level system. In case (b), the initial collective state includes two fully symmetrical $\frac{1}{2}N$-atom states, the one being formed with $|1a\rangle$ and $|1a'\rangle$ monatomic states, the other with $|1b\rangle$ and $|1b'\rangle$ states. Cooperative emission of one photon on the $\frac{3}{2} \rightarrow \frac{1}{2}$ transition is accompanied by the replacement, in the collective wave function, of one monatomic state $|1a\rangle$ or $|1a'\rangle$ by a state $|2a\rangle$ (the b-level system is henceforth omitted, since it evolves exactly as the a-level system). Because of the symmetry conservation, each pair of atoms initially in a state $|1a, 1a'\rangle_{AS}$ cannot emit more than one photon and reaches then either a state $|2a, 1a'\rangle_{AS}$ or a state $|1a, 2a\rangle_{AS}$, which is subradiant with respect to the $\frac{3}{2} \rightarrow \frac{1}{2}$ transition. The same pair can still emit on the $\frac{1}{2} \rightarrow \frac{1}{2}$ transition and reach either a state $|3a, 1a'\rangle_{AS}$ or a state $|1a, 3a\rangle_{AS}$. These states are not subradiant, since they involve monatomic states which are not connected by an atomic transition, and the cooperative emission on the $\frac{3}{2} \rightarrow \frac{1}{2}$ transition can start again. The three periods of emission which are observed and computed in case (a) are thus pretty well understood. The first emission of $N/2$ photons on $\frac{3}{2} \rightarrow \frac{1}{2}$ leads to a subradiant state, where half the atoms are left in the upper level. $N/2$ photons are then emitted on $\frac{1}{2} \rightarrow \frac{1}{2}$. Finally, the $N/2$ last photons emitted on $\frac{3}{2} \rightarrow \frac{1}{2}$ constitute the "subradiance echo." In case (b), as the initial collective state is fully symmetrical with respect to a and b permutations, a complete deexcitation is predicted for the emission on each transition, in agreement with both observed and computed results.

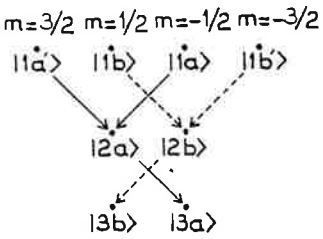

FIG. 4. Zeeman sublevels of the considered $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ atomic cascade.

In our experiment, spontaneous emission is inhibited only in the end-fire field modes which are involved in the cooperative process (i.e., for which the atoms are locally undistinguishable). For these modes, the photon trapping which is observed is the exact generalization of the one predicted for two two-level atoms; the antisymmetry of the subradiant states indicates precisely that many-atom subradiance is actually due to two-atom interaction. This Letter aims to contribute to answering the fundamental question of knowing how the elementary process of atomic absorption[15] or spontaneous emission[16] can be modified. An experimental study on noteworthy polarization properties of the "subradiance echo" is presently in progress.

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[13]As there is no initial coherence between a and b states, the case of a coherent superposition of the two types of states can be ignored. Furthermore, the two classes are only statistically defined.

[14]The atom-field Hamiltonian is indeed invariant under all local permutations and all atoms are locally undistinguishable. Moreover, interatomic interference occurs also between the two classes, because of the presence of degenerate transitions, but we have shown in Ref. 8 that its influence does not depend on the symmetry properties of the collective state.

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7. INTERPRETATION THEORIQUE DE L'EXPERIENCE

7.1 Interprétation qualitative de l'expérience

On donne ici une description qualitative de l'évolution spontanée coopérative dans le cas de la cascade $4d_{3/2} \rightarrow 5p_{1/2} \rightarrow 5s_{1/2}$ du gallium et pour les conditions initiales qui correspondent à une excitation en lumière polarisée linéairement à partir soit du niveau $4p_{1/2}$ soit du niveau $4p_{3/2}$. Puisque l'excitation est linéairement polarisée, l'évolution de chaque état $|j,m>$ est identique à celle de l'état $|j,-m>$ et les évolutions des deux sous–systèmes a et b en configuration "Y", qu'on peut (§ 4.2.1), considérer séparément si l'on ignore l'interférence existant entre les transitions Zeeman de même polarisation, sont exactement les mêmes. En particulier, pour des atomes initialement non corrélés, les propriétés de symétrie de l'état collectif peuvent alors être obtenues exactement de la même façon que dans le cas d'atomes à trois niveaux, et elles dépendent simplement du mélange statistique.

L'état collectif initial dans le cas où l'excitation part du niveau métastable $4p_{3/2}$ en régime saturant est un mélange statistique d'états collectifs ayant tous les mêmes propriétés de symétrie. L'évolution spontanée d'un tel système est alors la même que si l'état initial était n'importe lequel de ces états collectifs, c'est à dire n'importe lequel des états collectifs "les plus antisymétriques". Un tel état contient $N/4$ paires d'atomes dans un état antisymétrique formé avec des états monoatomiques $|1a>$ et $|1a'>$, qu'on écrit $|1a,1a'>_{AS}$, et $N/4$ paires analogues correspondant au système de niveaux b. L'émission coopérative d'un photon sur la transition $3/2 \rightarrow 1/2$ est accompagnée par le remplacement, dans la fonction d'onde collective, d'un état monoatomique $|1a>$ ou $|1a'>$ par un état $|2a>$, et pareillement pour le système b, qui est dorénavant omis, puisqu'il évolue exactement de la même façon que

le système a. A cause de la conservation de la symétrie, chaque paire d'atomes initialement dans un état $|1a, 1a' >_{AS}$ ne peut pas émettre plus d'un photon, et atteint donc soit l'état $|2a, 1a' >_{AS}$, soit l'état $|1a, 2a >_{AS}$, et les deux états sont subradiants par rapport à la transition $3/2 \rightarrow 1/2$. Cette paire peut néanmoins émettre sur la transition $1/2 \rightarrow 1/2$, et atteint ainsi soit un état $|3a, 1a' >_{AS}$, soit un état $|1a, 3a >_{AS}$. Ces états ne sont pas subradiants, puisqu'ils impliquent des états monoatomiques qui ne sont pas connectés par une transition atomique, et l'émission coopérative sur la transition $3/2 \rightarrow 1/2$ peut donc recommencer. On comprend ainsi aisément les trois périodes d'émission. La première émission de $N/2$ photons sur la transition $3/2 \rightarrow 1/2$ conduit à un état subradiant, dans lequel la moitié des atomes est restée dans le niveau supérieur. $N/2$ photons sont ensuite émis sur la transition $1/2 \rightarrow 1/2$. Les $N/2$ photons émis finalement sur la transition $3/2 \rightarrow 1/2$ constituent l' "écho de subradiance".

Dans le cas où l'excitation a lieu à partir du fondamental $4p_{1/2}$, l'état collectif initial inclut deux états complètement symétriques à $N/2$ atomes, l'un formé avec des états monoatomiques $|1a >$ et $|1a' >$, l'autre avec des états $|1b >$ et $|1b' >$. Une désexcitation complète est donc prévue après l'émission de deux impulsions successives qui dépeuplent complètement le niveau supérieur et le niveau intermédiaire.

7.2 Cascades entre niveaux dégénérés: subradiance "par antisymètrie" et subradiance "Zeeman"

Dans l'article intitulé "Superradiance and subradiance: IV. Atomic cascades between degenerate levels", qui est présenté dans le paragraphe 5.2, on étudie en détail le phénomène de subradiance tel qu'il peut apparaître dans les systèmes macroscopiques dans le cas général d'une cascade entre niveaux dégénérés. On analyse ainsi en particulier, dans le paragraphe § 4, les situations expérimentales qui ont donné lieu à la première mise en évidence de la subradiance (Pavolini et al 1985), et à la première mise en évidence de la superradiance (Skribanowitz et al 1973). Dans le premier cas, il s'agit d'une cascade $3/2 \rightarrow 1/2 \rightarrow 1/2$, tandis que dans le deuxième cas il s'agit d'une cascade $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$. On examine l'influence sur l'évolution du système des deux types d'interférence que peuvent apparaître, et qui peuvent donner lieu à un phénomène de subradiance. La cascade $3/2 \rightarrow 1/2 \rightarrow 1/2$ est étudiée pour tous les différents états initiaux qu'on peut atteindre avec une excitation qui peut démarrer d'un niveau de moment angulaire $1/2$ ou bien $3/2$; la cascade $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$, en revanche, est étudiée seulement dans le cas d'un état initial qui est un mélange statistique à poids égaux des sous–niveaux Zeeman du niveau $j = 3$ supérieur, c'est à dire le cas le plus favorable à la manifestation d'un phénomène de subradiance.

Des simulations numériques dans le modèle semi–classique ont été effectuées. Pour cela, nous avons écrit un programme qui construit et résoud les équations de Bloch–Maxwell pour un ensemble de niveaux dégénérés quelconques, en utilisant d'ailleurs les simplifications apportées par la séparation des équations de Bloch en deux sous–systèmes indépendants liés aux deux classes a et b d'atomes. Les calculs réalisés reproduisent correctement les résultats prévus. En particulier, lors-

qu'on prévoit qu'un état subradiant peut être atteint, à cause de la subradiance "par antisymétrie" ou/et de la subradiance "Zëeman" sur la transition supérieure, un écho de subradiance est émis après l'émission d'une impulsion sur la transition inférieure de la cascade, comme prévu en présence du niveau inférieur de la cascade (§ 4.3). L'intensité maximum de l'écho dépend en général à la fois de la proportion d'atomes qui avaient précédemment été empêchés d'émettre, et de la possibilité qu'un phénomène de subradiance "Zeeman" se produise à nouveau sur la même transition. Les résultats des calculs numériques sont en général en bon accord avec les prévisions théoriques qui tiennent compte séparément des deux types d'interférence. Cependant, il faut souligner que les équations de Bloch–Maxwell, à cause de leur origine semi–classique, sont incapables de reproduire le démarrage de l'évolution du système, qui a une origine purement quantique: on simule le démarrage de l'évolution en choisissant un champ initial qui représente les fluctuations du vide de photons. L'intensité maximum de l'écho de subradiance se trouve dépendre aussi du choix fait pour la polarisation de ce champ. Dans le cas de la cascade $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$, on a trouvé que ce choix peut, à la limite, faire disparaître totalement l'écho. L'étude des propriétés de polarisation de l'écho de subradiance est le sujet du chapitre suivant.

8. PROPRIETES DE POLARISATION

8.1 Cas d'une cascade $3/2 \rightarrow 1/2 \rightarrow 1/2$.

On considère ici le cas, étudié dans le paragraphe 4.1, d'un état collectif initial qui est un mélange statistique complet à poids égaux de tous les sous-niveaux Zeeman du niveau supérieur $3/2$ d'une cascade $3/2 \rightarrow 1/2 \rightarrow 1/2$. On a prédit et observé (Pavolini et al 1985) que, à cause d'un effet de subradiance, l'émission sur la transition $3/2 \rightarrow 1/2$ se fait en deux impulsions bien séparées temporellement par l'émission d'une impulsion superradiante sur la transition $1/2 \rightarrow 1/2$. On a de plus trouvé par un calcul semi-classique que si la polarisation de la première impulsion est linéaire et de direction arbitraire, celle de l'écho de subradiance est aussi linéaire et orthogonale à la précédente. Ce résultat n'est pas surprenant si l'on prend comme axe de quantification cette direction de polarisation. La figure 11 indique les niveaux qui restent

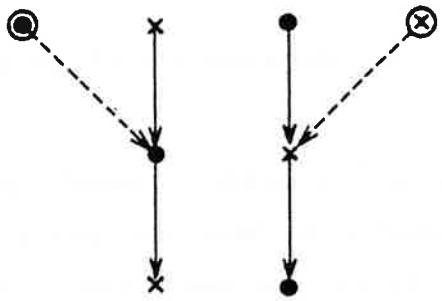


figure 11: cascade $3/2 \rightarrow 1/2 \rightarrow 1/2$ On a entouré les sous-niveaux Zeeman des sous-systèmes a ($\bullet$) et b ($\times$) qui restent peuplés après émission des deux impulsions polarisées linéairement ($\longrightarrow$). L'écho de subradiance ($- - \rightarrow$) a donc une polarisation orthogonale à celle de la première impulsion

alors peuplés après l'émission d'une impulsion sur chacune des deux transitions. On comprend parfaitement que la polarisation de l'écho de subradiance soit linéaire et orthogonale à celle de la première impulsion émise sur $3/2 \rightarrow 1/2$. Le même raisonnement s'applique aussi au cas où la polarisation de la première impulsion est $\sigma_{\pm}$. On choisit cette fois comme axe de quantification la direction de propagation de la lumière

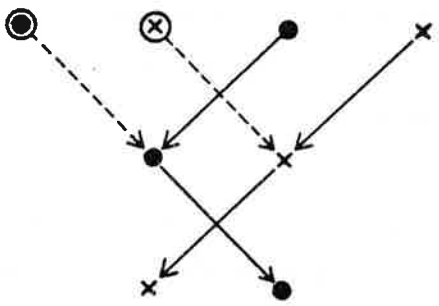


figure 12: même légende que pour la figure 11 mais dans le cas où la première impulsion (⟶) est polarisée σ_+. L'écho de subradiance (- - ⇢) est alors polarisé σ_-

superradiante (figure 12): la polarisation de l'écho de subradiance est alors évidemment $\sigma_{\mp}$.

En fait la polarisation de la première impulsion superradiante est aléatoire; il est donc opportun de déterminer préalablement la distribution de probabilité: c'est ce que nous faisons à l'aide de calculs semi-classiques dans le paragraphe 8.1.1. Dans le cas le plus général d'une polarisation elliptique, le raisonnement précédent est impossible à généraliser. Les calculs montrent toutefois (paragraphe 8.1.2) que la polarisation de l'écho de subradiance est encore pratiquement orthogonale à celle de la première impulsion quelles que soient les conditions initiales choisies, c'est à dire quelle que soit la polarisation du premier pulse.

Ce résultat peut en fait être démontré de façon tout à fait générale en utilisant les propriétés de symétrie (ou plutôt d'antisymétrie) de l'état atomique collectif (paragraphe 8.1.3).

8.1.1 Polarisation de la première impulsion

La distribution de probabilité de la polarisation de la première impulsion superradiante émise sur la transition $3/2 \rightarrow 1/2$ est parfaitement déterminée par les deux histogrammes des figures 13 et 14. Ces deux histogrammes décrivent en effet ce que l'on obtiendrait en analysant la lumière superradiante suivant deux polarisations orthogonales, σ_+ et σ_- (resp. u_1 et u_2, où u_1 et u_2 sont deux polarisations linéaires quelconques orthogonales entre elles), et en cherchant l'histogramme qui représente la distribution de probabilité de l'angle ϕ (resp. θ) défini comme $tg^{-1}\sqrt{\frac{I_1}{I_2}}$, où I_1 et I_2 sont les intensités maximales correspondant aux deux polarisations d'analyse. La méthode utilisée pour calculer ces deux histogrammes repose sur un calcul quantique, dans l'approximation linéaire, du démarrage de l'émission (Pillet 1982, Crubellier et al 1986).

On remarque sur les figures 13 et 14 que, alors que pour une transition $1/2 \rightarrow 1/2$ la polarisation prévue et observée (Crubellier et al 1978, 1981,1984, 1986) était pratiquement linéaire, la probabilité d'une polarisation fortement elliptique, voire pratiquement circulaire, est ici très importante. Ceci peut être attribué à l'interférence entre les transitions Zeeman de même polarisation. Si l'on choisit l'axe de quantification suivant la direction de propagation de la lumière superradiante, l'émission a lieu dans deux configurations en "V". Si l'on ignore l'interférence entre transitions différentes, ces deux configurations sont indépendantes: on s'attendrait alors à une inhibition pratiquement totale de l'émission sur les transitions $|1a> \rightarrow |2a>$ et $|1b> \rightarrow |2b>$,

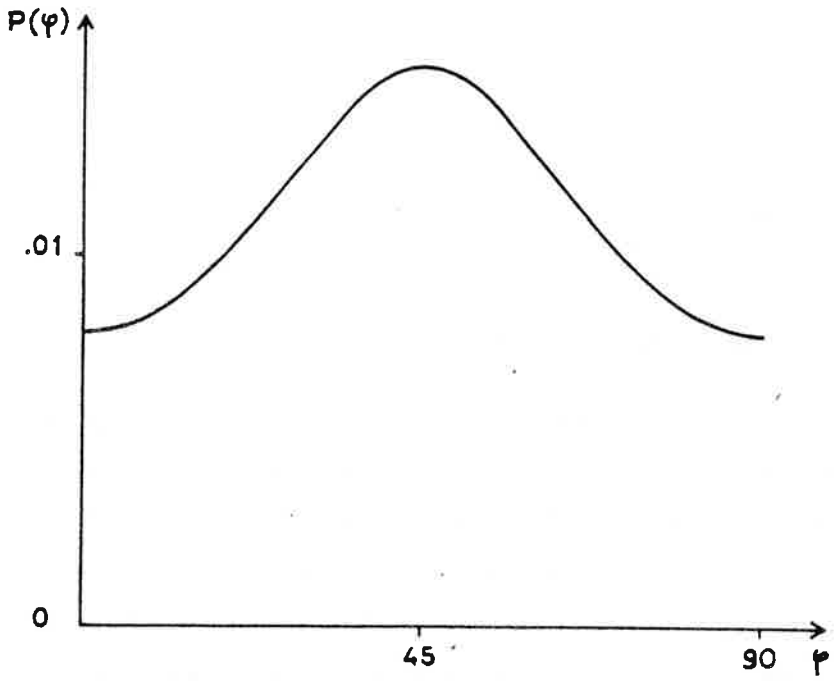
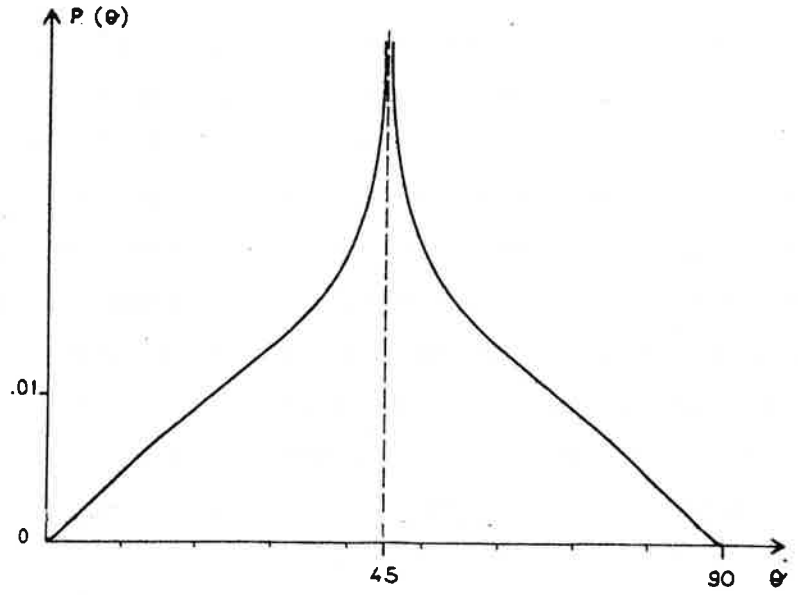


figure 13: distribution de la probabilité de l'angle $\phi = Arctg\sqrt{I_+/I_-}$ où I_+ et I_- sont les intensités maximales des composantes σ_+ et σ_- de la lumière superradiante

figure 14: même légende qhe pour la figure 13 mais dans le cas d'une analyse suivant deux polarisations linéaires orthogonales



dont les probabilités de transitions sont trois fois plus faibles que celles des transitions $|1a' > \rightarrow |2a >$ et $|1b' > \rightarrow |2b >$. La situation serait alors exactement la même que pour une transition $1/2 \rightarrow 1/2$: les différences observées entre les deux cas sont donc bien dues à la présence de transitions dégénérées.

8.1.2 Polarisation de l'écho de subradiance

L'écho de subradiance peut être calculé de façon simplifiée en ignorant la transition $1/2 \rightarrow 1/2$. Pour cela, on part de l'état subradiant atteint après émission de la première impulsion et on y annule purement et simplement les populations et cohérences du niveau intermédiaire $1/2$. On trouve ainsi que, à cause de la cohérence presque totale qui existe dans l'état subradiant entre les paires de sous–niveaux Zeeman telles que $|\Delta m| = 2$, la polarisation de l'écho de subradiance n'est pas véritablement aléatoire mais est imposée par la polarisation de la première impulsion: on trouve par exemple que l'angle ϕ' qui est défini de façon analogue à ϕ et qui caractérise l'ellipticité de la polarisation de l'écho est déterminé à mieux qu'un degré près, tandis que la position des axes de l'ellipse est parfaitement fixée. Les grands axes des ellipses associées aux polarisations de la première impulsion et de l'écho sont toujours orthogonaux. Par ailleurs la corrélation qui existe entre les angles ϕ et ϕ' est indiquée dans la figure 15. On voit qu'on a, en première approximation, $\phi' = \pi/2 - \phi$. Ceci signifie que le rapport entre la longueur du petit axe de l'ellipse et celle du grand axe est le même pour les deux impulsions, tandis que les sens de rotation sont opposés. Ceci, joint au résultat concernant les positions des axes des ellipses, prouve que, en première approximation, les deux polarisations, celle de la première impulsion et celle de l'écho, sont orthogonales. En fait les écarts à cette loi sont, comme on le verra plus loin, dus encore

une fois à l'interférence entre transitions Zeeman de même polarisation.

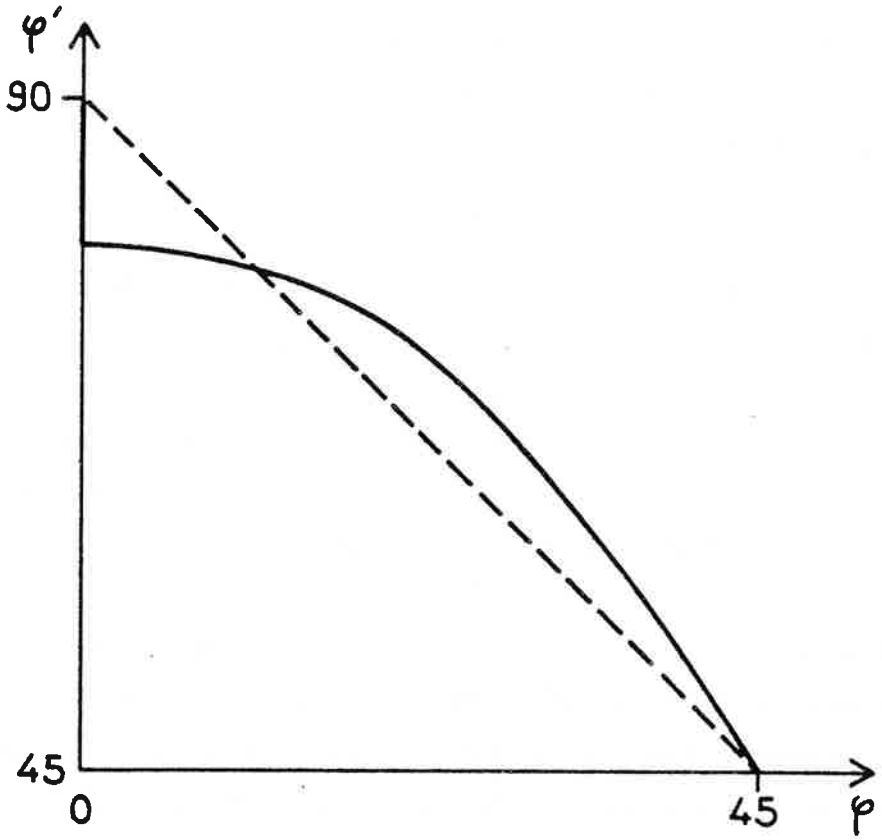


figure 15: corrélation entre les angles ϕ et ϕ' correspondant respectivement à une analyse en polarisations circulaires de la première impulsion et de l'écho

8.1.3 Antisymétrie et polarisation

L'état collectif initial considéré ici peut être, comme on l'a déjà vu, visualisé comme un ensemble de paires d'atomes de classe a et de classe b dans un état antisymétrique. Considérons par exemple une paire d'atomes de classe a; leur état initial est

(20) $$|\psi_o >= |1a, 1a' >_{AS}$$

En émission coopérative, chaque atome ou paire d'atomes rayonne essentiellement par émission stimulée. La polarisation de la lumière rayonnée sur la transition $3/2 \rightarrow 1/2$ par la paire d'atomes est donc identique à la polarisation $\mathbf{u}$ de la première impulsion superradiante. Cette polarisation est intrinsèquement aléatoire (Crubellier et al 1986), et on écrit

$$\mathbf{u} = \alpha_+ \mathbf{e}_{+1} + \alpha_- \mathbf{e}_{-1} \tag{21}$$

où

$$\mathbf{e}_{\pm 1} = \mp(\mathbf{e}_x \pm i\mathbf{e}_y)/\sqrt{2} \tag{22}$$

sont les vecteurs de base associés aux polarisations σ_+ et σ_- et où α_+ et α_- sont des coefficients aléatoires complexes vérifiant $|\alpha_+|^2 + |\alpha_-|^2 = 1$. A cause de la conservation de la symétrie l'émission sur $3/2 \rightarrow 1/2$ ne peut pas désexciter les atomes plus qu'à moitié (§ 7.1) et l'état qui est alors atteint par la paire peut s'écrire

$$|\psi_1 > \propto (\mathbf{R} \cdot \mathbf{u}^*)|\psi_o > \tag{23}$$

où $\mathbf{R}$ est l'opérateur "échelle" associé à la transition $3/2 \rightarrow 1/2$, c'est à dire

$$\mathbf{R} = \sum_{\alpha=1..N} \sqrt{3}|2a >_{\alpha\alpha} < 1a'|\mathbf{e}_{-1} + |2a >_{\alpha\alpha} < 1a|\mathbf{e}_{+1} \tag{24}$$

On trouve ainsi

$$|\psi_1 > = [\alpha_+^*|2a, 1a' >_{AS} + \sqrt{3}\alpha_-^*|1a, 2a >_{AS}]/\sqrt{|\alpha_+|^2 + 3|\alpha_-|^2} \tag{25}$$

ou, en développant les états antisymétriques,

$$|\psi_1 > = |2a, \phi >_{AS} \tag{26}$$

où $|\phi>$ est un état monoatomique défini comme

(27) $$|\phi>=[\alpha_+^*|1a'>-\sqrt{3}\alpha_-^*|1a>]/\sqrt{|\alpha_+|^2+3|\alpha_-|^2}$$

L'émission d'un photon sur la transition $1/2 \rightarrow 1/2$ laisse ensuite la paire d'atomes dans un état qu'on peut écrire

(28) $$|\psi_2>=|3a,\phi>_{AS}$$

Cet état peut de nouveau rayonner sur la transition $3/2 \rightarrow 1/2$: la polarisation u' de ce rayonnement est imposée par la forme de l'état monoatomique $|\phi>$ et l'on trouve

(29) $$u' = -\alpha_-^* e_{+1} + \alpha_+^* e_{-1}$$

Si l'on considère les sous–systèmes a et b comme indépendants, c'est à dire si on ignore l'interférence entre les transitions Zeeman de même polarisation, la polarisation de l'écho de subradiance est donc orthogonale à celle de la première impulsion.

8.2 Cas d'une cascade $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$

On considère, comme dans le paragraphe 4.3 de l'article présenté dans le chapitre 5, le cas d'une cascade $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ dont le niveau supérieur $j = 3$ est initialement peuplé dans un mélange statistique complet, à poids égaux, de tous ses sous–niveaux Zeeman. Comme on a

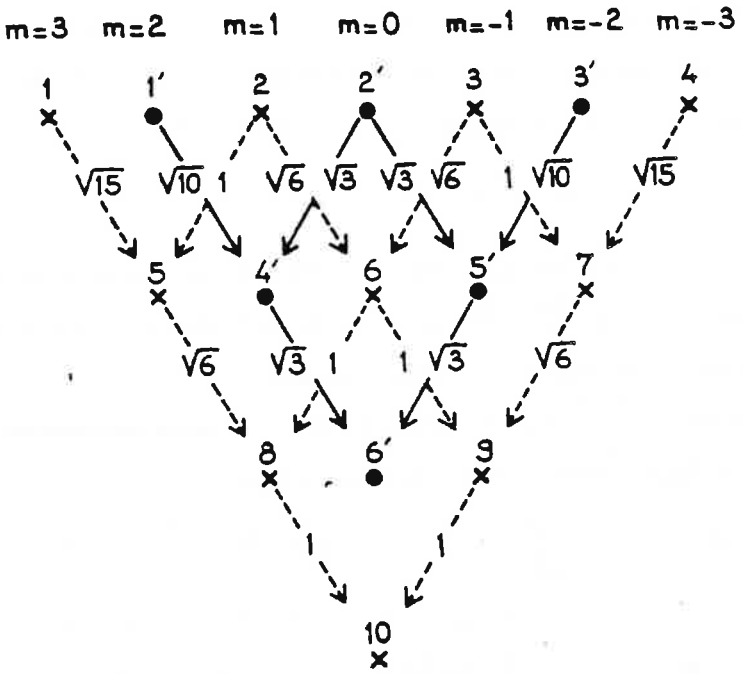


figure 16: sous–niveaux Zeeman des sous–systèmes a (×) **et** b (●) **pour la cascade** $3 \rightarrow 2 \rightarrow 1 \rightarrow 0$ **et amplitudes de probabilité relatives des diverses transitions Zeeman**

vu, l'état collectif du système peut être alors représenté par un produit de fonctions d'onde complètement antisymétriques à quatre atomes de classe a et à trois atomes de classe b. L'évolution de chaque quatuor (resp. trio) d'atomes comporte l'émission de trois (resp. deux) pho-

tons sur la transition $3 \rightarrow 2$ puis de deux (resp. un) photons sur la transition $2 \rightarrow 1$, photons qui appartiennent respectivement à deux impulsions superradiantes émises successivement sur les deux premières transitions. En raison de la nature quasi–classique du champ émis, la polarisation de chacune de ces impulsions est bien définie, mais on montre (Crubellier et al 1986) qu'elle est aléatoire d'une impulsion à l'autre. Nous n'étudions pas ici la distribution statistique de ces polarisations et nous supposons simplement que pour une suite d'impulsions donnée la polarisation des deux premières impulsions est caractérisée par des vecteurs complexes quelconques, $\mathbf{u}_1$ et $\mathbf{u}_2$, définis par

$$\mathbf{u}_i = \alpha_i \mathbf{e}_{-1} + \beta_i \mathbf{e}_{+1} \quad (i = 1, 2) \tag{30}$$

où $|\alpha_i|^2 + |\beta_i|^2 = 1$ et où $\mathbf{e}_{\pm 1}$ sont les vecteurs unitaires associés aux polarisations $\sigma_\pm$ (éq. 22). On peut alors déterminer qualitativement l'évolution des quatuors et des trios d'atomes.

Les operateurs "échelle" correspondant aux deux premières transitions et aux deux classes a et b s'écrivent (figure 16):

$$\begin{aligned}
\mathbf{R}_a^-(3 \rightarrow 2) &= \sum_{\alpha=1,\mathcal{N}} (\sqrt{15}|5>_{\alpha\alpha}<1| + \sqrt{6}|6>_{\alpha\alpha}<2| + |7>_{\alpha\alpha}<3|)\mathbf{e}_{-1} \\
&\quad + (|5>_{\alpha\alpha}<2| + \sqrt{6}|6>_{\alpha\alpha}<3| + \sqrt{15}|7>_{\alpha\alpha}<4|)\mathbf{e}_{+1} \\
\mathbf{R}_a^-(2 \rightarrow 1) &= \sum_{\alpha=1,\mathcal{N}} (\sqrt{6}|8>_{\alpha\alpha}<5| + |9>_{\alpha\alpha}<6|)\mathbf{e}_{-1} \\
&\quad + (|8>_{\alpha\alpha}<6| + \sqrt{6}|9>_{\alpha\alpha}<7|)\mathbf{e}_{+1} \\
\mathbf{R}_b^-(3 \rightarrow 2) &= \sum_{\alpha=1,\mathcal{N}} (\sqrt{10}|4'>_{\alpha\alpha}<1'| + \sqrt{3}|5'>_{\alpha\alpha}<2'|)\mathbf{e}_{-1} \\
&\quad + \sqrt{3}|4'>_{\alpha\alpha}<2'| + \sqrt{10}|5'>_{\alpha\alpha}<3'|)\mathbf{e}_{+1} \\
\mathbf{R}_b^-(2 \rightarrow 1) &= \sum_{\alpha=1,\mathcal{N}} (|6'>_{\alpha\alpha}<4'|)\mathbf{e}_{-1} + |6'>_{\alpha\alpha}<5'|)\mathbf{e}_{+1}
\end{aligned} \tag{31}$$

L'état initial d'un quatuor s'écrit

$$|\psi_o> = |1234>_{AS} \tag{32}$$

Son état après l'émission de trois photons de polarisation $\mathbf{u}_1$ est donné par

$$|\psi_1 > \propto (\mathbf{R}_a^-(3 \to 2) \cdot \mathbf{u}_1^*)^3 |\psi_o > \tag{33}$$

et on trouve, après un calcul analogue à celui du paragraphe 8.1.3, que

$$|\psi_1 > = |567\phi_1 >_{AS} \tag{34}$$

où $|\phi_1 >$ est un état monoatomique du niveau $j = 3$ défini, à un facteur de normalisation près, par

$$|\phi_1 > \propto -(\alpha_1^*)^3|1 > +\sqrt{15}(\alpha_1^*)^2\beta_1^*|2 > -\sqrt{15}\alpha_1^*(\beta_1^*)^2|3 > +(\beta_1^*)^3|4 > \tag{35}$$

De la même façon l'état du quatuor après émission de deux photons de polarisation $\mathbf{u}_2$ sur la transition $2 \to 1$ s'écrit

$$|\psi_2 > = |89\phi_2\phi_1 >_{AS}$$

où $|\phi_2 >$ est un état monoatomique du niveau $j = 2$ défini, à un facteur de normalisation près, par

$$|\phi_2 > \propto (\alpha_2^*)^2|7 > -\sqrt{6}\alpha_2^*\beta_2^*|6 > +(\beta_2^*)^2|5 > \tag{36}$$

Une fois cet état atteint, l'émission ultérieure d'un photon sur la transition $3 \to 2$, c'est à dire l'émission de l'écho de subradiance, dépend de l'action sur $|\psi_2 >$ de l'opérateur $(\mathbf{R}_a^-(3 \to 2) \cdot \mathbf{u'}_1^*)$, où $\mathbf{u'}_1$ est une polarisation pour le moment inconnue. On trouve aisément que

$$\begin{aligned} &(\mathbf{R}_a^-(3 \to 2) \cdot \mathbf{u'}_1^*)|\phi_1 > \\ &= \sqrt{15}(\alpha_1^*\beta_1'^* - \alpha_1'^*\beta_1^*)[(\alpha_1^*)^2|7 > -\sqrt{6}\alpha_1^*\beta_1^*|6 > +(\beta_1^*)^2|5 >] \end{aligned} \tag{37}$$

Cet état est identiquement nul si $\mathbf{u'}_1 \equiv \mathbf{u}_1$, c'est à dire que la polarisation de l'écho ne peut en aucun cas être identique à celle de la première impulsion. Il est en fait naturel de penser que la polarisation de l'écho

est celle qui maximise l'intensité, c'est à dire la polarisation orthogonale à u_1. Par ailleurs l'état défini par l'équation (36) ne dépend pas de la polarisation u'_1 et il s'écrit, à un facteur de normalisation près,

(38) $$|\phi'_2> \propto (\alpha_1^*)^2|7> -\sqrt{6}\alpha_1^*\beta_1^*|6> +(\beta_1^*)^2|5>$$

A cause de l'antisymétrie de la fonction d'onde, l'état du quatuor après émission de l'écho de subradiance est nécessairement de la forme

(39) $$|\psi_3> = |89\phi_2\phi''_2>$$

où $|\phi''_2>$ est un état monoatomique du niveau $j = 2$ orthogonal à $|\phi_2>$. La probabilité qu'a le quatuor, une fois dans l'état $|\psi_3>$, de rayonner à nouveau sur la transition $3 \rightarrow 2$ est donc proportionnelle, toutes choses égales d'ailleurs, à $1- <\phi_2|\phi'_2>$. Elle dépend donc des polarisations des deux premières impulsions émises; il n'y a en particulier pas d'écho si $|\phi_2>$ est identique à $|\phi'_2>$, c'est à dire si les polarisations u_1 et u_2 sont identiques; l'écho est au contraire maximum si $|\phi_2>$ et $|\phi'_2>$ sont orthogonales, c'est à dire si

(40) $$(\alpha_1^*)^2(\alpha_2^*)^2 + 6\alpha_1^*\alpha_2^*\beta_1^*\beta_2^* + (\beta_1^*)^2(\beta_2^*)^2 = 0$$

Le même raisonnement peut être fait pour les trios d'atoms. L'état d'un tel trio est initialement

(41) $$|\psi_0> = |1'2'3'>_{AS}$$

et il devient, après l'émission de deux photons sur la transition $3 \rightarrow 2$, avec la polarisation u_1,

(42) $$|\psi_1> \propto |4'5'\phi_1>_{AS}$$

où l'état $\phi_1>$ est un état monoatomique du niveau $j = 3$ défini, à un facteur de normalisation près, par

(43) $$|\phi_1> = \sqrt{3}(\alpha_1^*)^2|3'> -\sqrt{10}\alpha_1^*\beta_1^*|2'> +\sqrt{3}(\beta_1^*)^2|1'>$$

L'émission d'un photon sur la transition $2 \rightarrow 1$ avec la polarisation u_2 laisse alors le trio dans l'état

$$|\psi_2 >= |6'\phi_2\phi_1 >_{AS} \tag{44}$$

où

$$|\phi_2 >= \alpha_2^*|5' > +\beta_2^*|4' > \tag{45}$$

Là encore l'émission sur la transition $3 \rightarrow 2$ ne peut en aucun cas reprendre avec une polarisation u_1. La probabilité maximale est obtenue pour la polarisation orthogonale à u_1 et l'action de l'opérateur échelle sur $|\phi_1 >$ donne un état monoatomique $|\phi_2' >$ défini par

$$|\phi_2' >= \alpha_1^*|5' > -\beta_1^*|4' > \tag{46}$$

cet état est orthogonal à $|\phi_2 >$ et l'état du trio après émission de l'écho est donc

$$|\phi_3 >= |6'\phi_2'\phi_2 >_{AS} \tag{47}$$

En résumé on trouve pour les deux classes d'atomes que la polarisation de l'écho de subradiance ne peut jamais être identique à celle de la première impulsion et que la polarisation la plus probable est au contraire la polarisation orthogonale. De plus, on trouve en examinant le cas du sous-système a que l'importance de l'écho dépend de la polarisation des deux premières impulsions: il n'y a en particulier pas d'écho si les polarisations sont identiques. Ce résultat confirme et complète le raisonnement esquissé dans le paragraphe 4.3 de l'article présenté dans le chapitre 5.

d'interférence destructive due à la présence de transitions Zeeman de même polarisation, c'est à dire de transitions dégénérées, qu'il m'a paru intéressant d'étudier en détail. Le cas typique est celui d'une configuration à quatre niveaux reliés par deux transitions de même fréquence et polarisation, pour lesquelles nous avons pu montrer que la subradiance prévue dépend des différentes probabilités de transition aussi bien que des populations initiales des deux niveaux supérieurs. Nous avons aussi étudié les configurations où l'un des deux niveaux est commun aux deux transitions: ceci nous a notamment permis de montrer qu'un nouvel effet de subradiance est prévu en particulier dans le cas d'un système en cascade. Le dernier pas vers une expérience de subradiance a alors été l'analyse de configurations de niveaux encore plus compliquées pour lesquelles le phénomène, qui consiste en une inhibition de l'émission et qui serait très délicat à mettre en évidence directement, peut être rendu manifeste simplement. Nous avons choisi de considérer les configurations en cascade entre niveaux dégénérés, pour lesquelles nous avons pu montrer que la subradiance se manifeste par l'apparition de deux impulsions successives sur la transition où le phénomène est prévu.

Nous avons de plus réalisé la première mise en évidence de la subradiance à l'aide d'une cascade de niveaux de gallium: les résultats expérimentaux confirment parfaitement les prévisions qualitatives que l'on extrait de la symétrie de l'état atomique collectif. Nous avons parallèlement mis au point un programme numérique qui permet d'écrire et résoudre les équations d'évolution pour un système de niveaux dégénérés quelconques. Les calculs correspondant aux cas expérimentalement réalisés et à d'autres cas similaires sont en accord avec les résultats expérimentaux et les prévisions qualitatives. Ils montrent de plus que les polarisations des deux impulsions qui se produisent sur la transi-

tion où le phénomène de subradiance est prévu sont toujours orthogonales. Cette propriété d'orthogonalité des deux impulsions a été étudiée théoriquement, pour finir, dans le cas de l'expérience réalisée et dans le cas correspondant à la première mise en évidence de la superradiance. Dans les deux cas, des raisonnements concernant l'antisymétrie partielle de l'état collectif sont en mesure d'expliquer cette caractéristique de l'écho de subradiance.

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TITRE

Etude théorique et mise en évidence expérimentale du phénomène de subradiance

RESUME

L'interférence entre les atomes émetteurs qui caractérise l'émission spontanée coopérative peut être constructive ou destructive. Dans le second cas, on prévoit une inhibition de l'émission, qu'on appelle subradiance. Cette thèse est consacrée à l'étude théorique de ce phénomène, ainsi qu'à sa première démonstration expérimentale. La partie théorique s'intéresse notamment à la recherche des conditions nécessaires à l'apparition de la subradiance pour des systèmes macroscopiques. On y étudie les configurations à plusieurs niveaux, et, plus particulièrement, les transitions entre niveaux dégénérés. La mise en évidence a été réalisée sur la cascade $4d_{3/2} \rightarrow 5p_{1/2} \rightarrow 5s_{1/2}$ du gallium atomique.

MOTS CLES

Superradiance, subradiance, émission spontanée coopérative, interférence interatomique, indiscernabilité, symétrie par permutation