Experimental Evidence for Subradiance

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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 $j=\frac{3}{2}\rightarrow j'=\frac{1}{2}$ 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.

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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 $\frac{3}{2}\rightarrow\frac{1}{2}$ 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 $\frac{3}{2}\rightarrow\frac{1}{2}$ 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

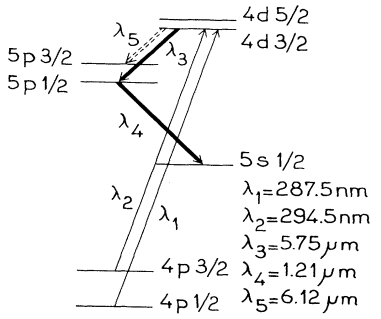

FIG. 1. Relevant level diagram of gallium.

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

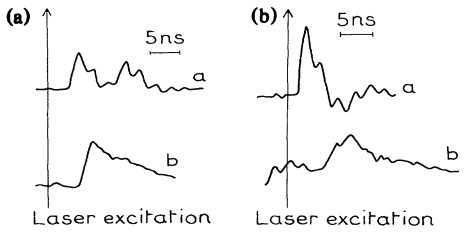

FIG. 2. Typical superradiant signals. (a) corresponds to the excitation with saturated linearly polarized light from the $4p_{3/2}$ level; (b), the $4p_{1/2}$ level. In both cases, traces *a* and *b* correspond respectively to $4d \rightarrow 5p$ and to $5p \rightarrow 5s$ transitions. The visible oscillations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

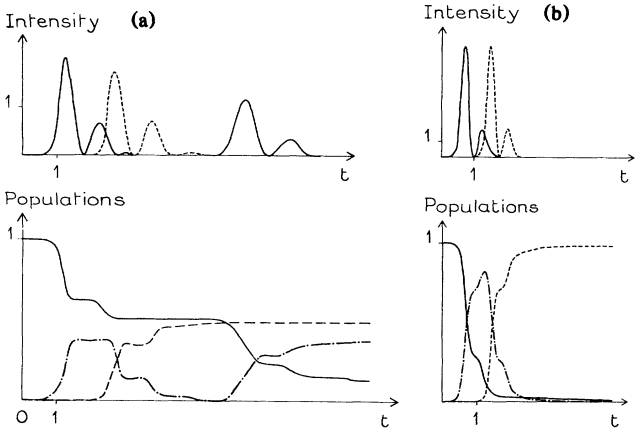

FIG. 3. Results of semiclassical calculations of cooperative spontaneous emission on a $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ cascade. In both cases (a) and (b) the upper curves show the total intensity radiated on $\frac{3}{2} \rightarrow \frac{1}{2}$ (solid line) and on $\frac{1}{2} \rightarrow \frac{1}{2}$ (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.

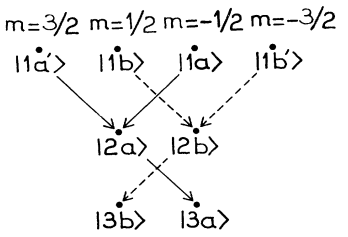

FIG. 4. Zeeman sublevels of the considered $\frac{3}{2} \rightarrow \frac{1}{2} \rightarrow \frac{1}{2}$ atomic cascade.

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.

Laboratoire Aimé Cotton, Centre National de la Recherche Scientifique, is a laboratoire associé à l'Université Paris-Sud.

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