Collective emission in a resonant cavity in the dependence on an external electric field

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(Received 25 February 1992)

Collective emission in a resonant cavity was observed between high-lying levels in Sr and Ba. These transitions react very sensitively to a static external electric field. The square root of the maximum of the intensity of the superfluorescent pulse decreases linearly with the second power of the electric field strength. The behavior of the maximum of the intensity, of the characteristic superfluorescent time, and of the delay time of the superfluorescent pulse was described in the framework of modified Maxwell-Bloch equations and explained by a change of the characteristic time constant of the superfluorescent pulse.

PACS number(s): 32.60.+i, 32.90.+a, 42.50.Fx

INTRODUCTION

Superfluorescence is a coherent spontaneous emission of an ensemble of atoms or molecules due to strong coupling by their common near-radiation fields. The superfluorescence is typically a pulse with peak intensity proportional to the square of the number of cooperating atoms, delayed from the time of simultaneous excitation [1–4]. This delay represents the time required for the individual atomic dipoles to come into phase lock as a result of the coupling between them.

Recently it was shown that superfluorescent transitions in free space between high-lying levels of Sr [5], Ba and Na [6] react very sensitively to an external electric field. The measurements showed that the square root of the peak intensity of the superfluorescent signal decreases linearly with the increase of the square of the electric field strength. The experimental result is confirmed qualitatively by calculations. Since the theory deals with two-level atoms, the experimental conditions have to be designed as closely as possible to a two-level atom. Superfluorescent cascades and branching have to be suppressed completely in order to simulate two-level atoms. Therefore we now observed the field dependence of the superfluorescent emission in a tunable cavity. The collective emission occurs only at transitions for which the cavity is resonant, thus avoiding unwanted cascading and branching [7]. The experiments were performed with Sr and Ba atoms. It was found that for all the investigated collective emissions between high-lying levels, the square root of the maximum of the intensity of the superfluorescent pulses decreases with the square of the electric field strength. The experimental results were described by the Maxwell-Bloch equations for a system of two-level atoms.

MEASUREMENTS

The experimental arrangement is shown in Fig. 1. A beam of alkaline earth atoms (Ba or Sr) ran through a cavity in a direction perpendicular to the axis of the cavity. The cavity was either a plane or a confocal Fabry-Pérot resonator with a small finesse of about 5. The surfaces of the mirrors covered with aluminum served as Stark plates, so that the direction of the electric field was parallel to the axis of the cavity. Using a HeNe laser as the monochromatic light source, the plane Fabry-Pérot resonator was adjusted optically and so the Stark plates were simultaneously aligned parallel with high accuracy. It was possible to vary the distance between the mirrors by a piezoelement.

In the center between the Stark plates, the beam density was approximately 10^{10} cm^{-3}. The atoms were excited stepwise via the $msmp\ ^1P_1$ level into a $msns\ ^1S_0$ or a $msn'd\ ^1D_2$ level ($m=5$ for Sr; $m=6$ for Ba) by two dye lasers simultaneously pumped by an excimer laser. The pulse lengths of the dye lasers were about 13 nsec full width at half maximum (FWHM).

The atomic population was initially inverted for a large

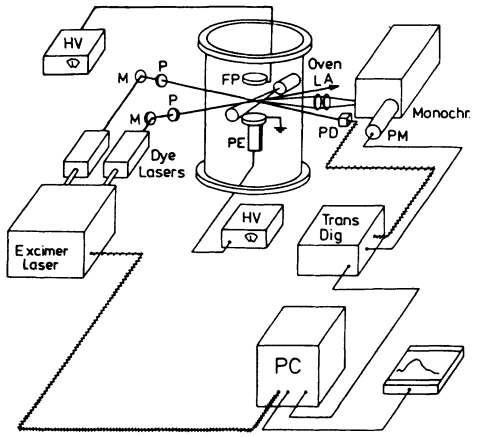

FIG. 1. Experimental setup. *M*, mirror; *P*, polarizer; *A*, analyzer; *L*, lens; FP, Fabry-Pérot; PE, piezo element; PM, photomultiplier; PD, photodiode; HV, high-voltage power supply; Trans Dig, transient digitizer; PC, personal computer.

number of optical transitions between the pumped $msns\ ^1S_0$ or $msn'd\ ^1D_2$ and lower $msn''p\ ^1P_1$ or $msn'''f\ ^1F_3$ levels. Tuning the cavity into resonance with one of these transitions led to superfluorescent emission only at one wavelength, since the collective emission occurs only at a transition for which the cavity is resonant. But this is not necessarily a transition between two levels, since complications might arise from the Stark effect. To select a sublevel as the initial level of the collective emission, the laser light of the two lasers was linearly polarized parallel to the electric field, so that only ($\Delta m = 0$) transitions were induced, which selectively populated the sublevel with $m = 0$ of the $^1D_2$ or $^1S_0$ level. Furthermore, the cavity can only sustain collective emission with a polarization direction perpendicular to the axis of the cavity, and therefore the final level of the superfluorescent transition is also determined ($|m| = 1$).

The collective emission was detected indirectly by observing the spontaneous decay of the superfluorescently populated level by means of a monochromator and a high-speed photomultiplier. The single-shot signal was recorded time resolved by a fast transient digitizer and stored by a personal computer. Each experimental run consisted of 500 single measurements, which were averaged to enhance the signal-to-noise ratio. Due to the indirect detection, one has to extract the superfluorescent pulse from the recorded signal before further investigations can be done. If one assumes that the superfluorescently populated level decays only by spontaneous emission with the time constant τ, the superfluorescent intensity $I(t)$ can be evaluated from the measured intensity $I_{obs}(t)$ [5]:

$$I(t) \propto (1/\tau) I_{obs}(t) + \dot{I}_{obs}(t) \ . \tag{1}$$

Figure 2 shows an example of an averaged decay curve and the superfluorescent signal derived by applying formula (1). To investigate the dependence of the superfluorescent emission on the electric field, the measurements were performed in one of the following infrared transitions at different electric field strengths:

Ba $6snd\ ^1D_2 - 6s(n-3)f\ ^1F_3 (n = 10, 11)$,

Sr $5s10s\ ^1S_0 - 5s9p\ ^1P_1$,

Sr $5snd\ ^1D_2 - 5snp\ ^1P_1$,

$5snd\ ^1D_2 - 5s(m-2)f\ ^1F_3 (m = 8, 9)$.

The frequency of the cavity was adjusted to the Stark shift of the superfluorescent transition frequency by varying the distance between the mirrors of the cavity. The electric field E caused a decrease of the spontaneous emission of the superfluorescently populated levels (see Fig. 3). Figure 4 shows the field dependence of the maximum of the intensity I_M of the superfluorescent pulses evaluated with Eq. (1) from the spontaneous emission. One can see that the square root of the maximum of the intensity decreases linearly with the square of the electric field strength. This is the same dependence that was found earlier in the case of free-space superfluorescence [5,6]. Furthermore, the superfluorescent pulses, which

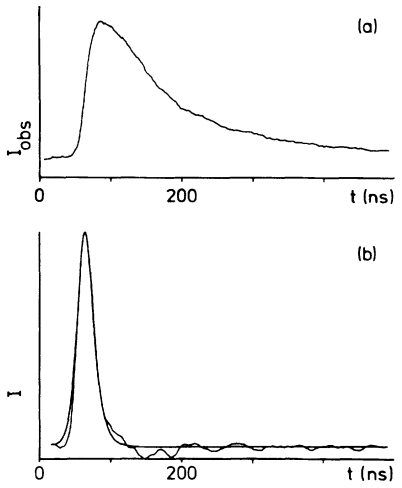

FIG. 2. (a) Experimental decay curve for the superfluorescently populated $6s8f\ ^1F_3$ level of Ba. Observed transition $6s8f\ ^1F_3 - 6s5d\ ^1D_2$. (b) Superfluorescent pulse of the transition $6s11d\ ^1D_2 - 6s8f\ ^1F_3$ derived from the decay curve compared with a numerical solution of the Maxwell-Bloch equations (for details see text).

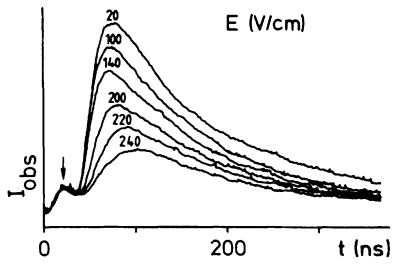

FIG. 3. Experimental decay curves for different electric field strengths observed in the transition $5s7f\ ^1F_3 - 5s4d\ ^1D_2$ of Sr following the superfluorescent transition $5s9d\ ^1D_2 - 5s7f\ ^1F_3$. The arrow denotes the fluorescence of the transition $5s5p\ ^1P_1 - 5s^2\ ^1S_0$ (first excitation step).

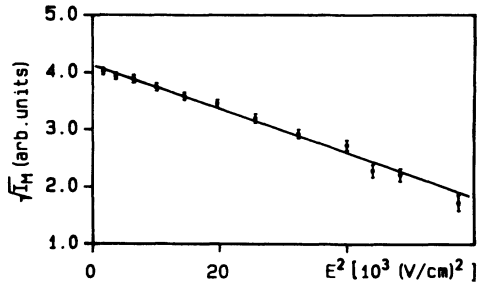

FIG. 4. Square root of the maximum of the intensity of the superfluorescent pulses plotted against the second power of the electric field strength, shown for the transition $5s9d\ ^1D_2 - 5s7f\ ^1F_3$ of Sr.

obviously change in the presence of an external electric field (Fig. 3), contain much more information than just about the decrease of the maximum intensity. The delay time and the width of the pulses increase with the increasing electric field strength. The connections between the maximum of the intensity, the delay time and the pulse width will be discussed in the following section with the Maxwell-Bloch equations.

DISCUSSION

For a theoretical discussion of the phenomenon, we describe the superfluorescence in a semiclassical way by the Maxwell-Bloch equations for a system of two-level atoms [8–10]. As was discussed above, only superfluorescent ($\Delta m = \pm 1$) transitions between Zeeman levels $m = 0$ and $|m| = 1$ were investigated. In this case, it is possible to introduce an alternative basis system, in which the dipole radiation vanishes in one transition [11], so that the superfluorescence can be described in a two-level system. When the axis of the cavity is chosen as the z axis, in the slowly varying envelope approximation the Maxwell-Bloch equations take the following form [8]:

$$\begin{aligned} \dot{\mathcal{P}}(z,t) &= (i|d|^2/\hbar)\mathcal{E}(z,t)n(z,t) , \\ \dot{n}(z,t) &= [i/(2\hbar)][\mathcal{E}^*(z,t)\mathcal{P}(z,t) \\ &\quad - \mathcal{E}(z,t)\mathcal{P}^*(z,t)] . \end{aligned} \tag{2}$$

$\mathcal{E}(z,t)$ is the electric radiation field in the cavity, $\mathcal{P}(z,t)$ the macroscopic polarization of the ensemble, and $n(z,t)$ the population inversion density. $|d|$ is the reduced electric dipole matrix element of the superfluorescent transition.

Since in the present investigations the delay of the superfluorescent pulses is of the same order of magnitude as the radiative lifetimes τ_i ($i = 1$ upper level, $i = 2$ lower level) of the states that are coupled by the superfluorescent emission, the decrease of the population densities n_i due to spontaneous emission, has to be considered in the Maxwell-Bloch equations. This decrease leads to a damping of the macroscopic polarization with a time constant T_2. An additional decrease of the macroscopic polarization with the time constant T_2^* is caused by the loss of phase coherence between the oscillating atoms mainly determined by the Doppler effect [12]. Moreover, the excitation process has to be considered in the Maxwell-Bloch equations, since the duration of the laser pulses is not negligibly small compared with the radiative lifetime τ_1. The pump rate, which populates the upper level, is given by the pulse shape of the pump laser, $\Lambda_P(t) = (t/\tau_P^2)\exp(-t/\tau_P)$ [7]. Equations (2) have to be completed by expressions which introduce the described relaxation and excitation mechanisms. In addition, for the numerical evaluation, the Maxwell-Bloch equations were converted in a system of rate equations consisting only of dimensionless quantities or quantities of dimension $\sec^{-1}$:

$$\begin{aligned} \dot{\rho}_1(z,t) &= \dot{n}_1(z,t)/n_0 \\ &= \Lambda_P(t) - (\tfrac{1}{2})\omega_R(z,t)v(z,t) - \rho_1(z,t)/\tau_1 , \\ \dot{\rho}_2(z,t) &= \dot{n}_2(z,t)/n_0 \\ &= (\tfrac{1}{2})\omega_R(z,t)v(z,t) - \rho_2(z,t)/\tau_2 , \\ \dot{v}(z,t) &= \dot{\mathcal{P}}(z,t)e^{-i\pi/2}/(n_0|d|) \\ &= \omega_R(z,t)[\rho_1(z,t) - \rho_2(z,t)] \\ &\quad - (1/T_2 + 1/T_2^*)v(z,t) . \end{aligned} \tag{3}$$

$n_0 = n_1 + n_2$ is the total population density. $\omega_R(z,t) = |\mathcal{E}(z,t)||d|/\hbar$ is the Rabi frequency of the transition.

The time necessary to establish a standing wave is about $\frac{1}{6}$ nsec in a cavity with a length of 50 mm. This time is much shorter than all the other evolution times in the problem, and especially the shortest time constant, the characteristic superfluorescence time T_R, which will be defined below. For the observed transitions, T_R has a minimum value of about 2 nsec. Therefore, $\mathcal{E}(z,t)$ and $\omega_R(z,t)$ can be described by a standing wave $\mathcal{E}(z,t) = \mathcal{E}_0(t)\cos(kz)$ and $\omega_R(z,t) = \omega_R(t)\cos(kz)$, respectively. It is sufficient to complete the above equations by an equation describing the time evolution of the radiation field $\mathcal{E}_0(t)$, and $\omega_R(t)$. An easy way to do this is to express the energy conservation for the combined atoms and field system [8]. In the notation introduced, this yields

$$\begin{aligned} \dot{\omega}_R(t) &= [1/(4T_R T_c)]\int v(z,t)\cos(kz)dz/\delta z \\ &\quad - \omega_R(t)/(2T_C) . \end{aligned} \tag{4}$$

$T_R = \epsilon_0\hbar\pi c/(8\omega|d|^2 n_0 f\delta z)$ is the characteristic time constant of the superfluorescent emission, where δz is the length of the active medium in the direction of the cavity axis, f the finesse of the cavity, and $\omega = kc$ the frequency of the superfluorescent transition. T_c is the damping time of the energy in the cavity. The set of Eqs. (3) and (4) allows one to calculate the coupled evolution of $n_1(z,t)$, $n_2(z,t)$, $\mathcal{E}(z,t)$, and $\mathcal{P}(z,t)$. The experimental conditions of the system at time $t = 0$ are given by

$$\begin{aligned} &n_1(z,t=0) = n_2(z,t=0) = \omega_R(t=0) = 0 , \\ &v(z,t=0) = b(n_0\delta z)^{-1/2}\cos(kz) . \end{aligned} \tag{5}$$

Expression (5) describes the *ad hoc* initial polarization simulating the random fields impinging on the system at time $t = 0$ [8,9]. Since the superfluorescent system is not initially inverted by the exciting laser pulse, a parameter b has to be introduced [13]. b was chosen in such a way that at a small static external electric field a good agreement was obtained between measured and calculated superfluorescent pulses (see Fig. 2). This was possible for all the investigated transitions with different values of the

parameter b. In a definite transition, b was a constant for all the values of the electric field strength.

In order to describe the experimental results of the collective emission in a static homogeneous electric field by the Maxwell-Bloch equations, we assume that the influence of the external electric field can be attached mainly to the characteristic time constant T_R. This is known from the investigation of the free superfluorescence in a homogeneous electric field [5,6].

A sequence of pulses of decreasing intensities was now calculated in the following way: Eqs. (3) and (4) were solved with the parameter set given by the experimental conditions and by the specified transition without an external electric field. Since the characteristic time constant increased with increasing electric field strength, the characteristic time constant was increased stepwise, Eqs. (3) and (4) were solved for each step. The simulated pulse data of the maximum of the intensity or of the pulse width were plotted versus the delay time (Fig. 5). To give a visual impression of the result, a solid line was drawn through the calculated points. The experimental values were also entered in the figure. The evaluations were done in the described way for all the investigated transitions in the plane resonator. The result is that the data of the simulated pulses obtained by changing the characteristic time constant T_R reproduce the course of the experimental pulse data in all the studied superfluorescence transitions very well. This result is not unexpected, since in a superfluorescent system T_R determines the features of the emission process. To discuss now the dependence of $T_R=\epsilon_0\hbar\pi c/(8\omega|d|^2 n_0 f\delta z)$ on the static electric field, the change in ω, $|d|^2$, and n_0 has to be considered.

To study the dependence of ω on the electric field strength, the Stark shift of the two levels that are connected by the superfluorescent pulse has to be calculated. The energy shift $\Delta W(JM)$ of a fine-structure level of a free atom in an electric field is expressed in terms of the scalar and tensor polarizabilities α_0 and α_2:

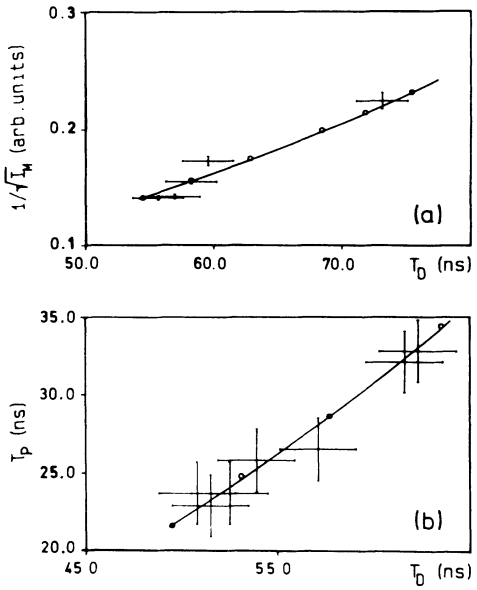

FIG. 5. Experimental (+) and simulated (○) data (see text) of the collective emission in a plane resonator. Simulated data were obtained by solution of the Maxwell-Bloch equations for increasing values of the characteristic time constant T_R (from left to right). (a) $I_M^{-1/2}$ plotted vs T_D for the transition $5s9d\ ^1D_2-5s9p\ ^1P_1$; (b) T_P vs T_D for the transition $5s9d\ ^1D_2-5s7f\ ^1F_3$.

$$\Delta W(JM)=-\{\alpha_0(J)+\alpha_2(J)[3M^2-J(J+1)]/[J(2J-1)]\}E^2/2\ . \tag{6}$$

Since the electric field admixes eigenstates of different parity, one has to use perturbed wave functions $|\overline{\gamma J}\rangle$ to calculate the change of $|d|^2$ with the electric field strength. The wave function is then written as

$$|\overline{\gamma J}\rangle=\left[|\gamma J\rangle+\sum_i\epsilon_i|\gamma_i J'\rangle+\sum_k\beta_k|\gamma_k J''\rangle\right]\Big/N\ , \tag{7}$$

with

$$N=\left[1+\sum_i\epsilon_i^2+\sum_k\beta_k^2\right]^{1/2},$$

$$\epsilon_i=(\langle\gamma_i J'|dE|\gamma J\rangle)/[W(\gamma J)-W(\gamma_i J')]\ ,$$

$$\beta_k=(\langle\gamma_k J''|dE|\gamma J\rangle)/[W(\gamma J)-W(\gamma_k J'')]\ .$$

As an example, the perturbed wave function of a $5snd\ ^1D_2$ level is given by (7) with $|\gamma J\rangle=|5snd\ ^1D_2\rangle$, $|\gamma_i J'\rangle=|5sip\ ^1P_1\rangle$, and $|\gamma_k J''\rangle=|5skf\ ^1F_3\rangle$.

The population density n_0 is proportional to the line strength of the exciting transition. The line strengths also have to be determined with the perturbed wave functions (7).

Using for the elements Sr and Ba multiconfigurational eigenfunctions [14–18], the dependence of ω, $|d|^2$, and n_0 on the electric field can be derived. The change of the characteristic time constant T_R with the electric field in the transition $6s10d\ ^1D_2-6s7f\ ^1F_3$ in Ba is then given by $T_R(E)=T_R(E=0)(1+0.018E^2\rho^2)$, in the transition $5s9d\ ^1D_2-5s7f\ ^1F_3$ in Sr by $T_R(E)=T_R(E=0)(1+0.022E^2\rho^2)$, and in the transition $5s10s\ ^1S_0-5s9p\ ^1P_1$ in Sr by $T_R(E)=T_R(E=0)\ [1+0.26\times10^{-3}E^2\rho^2)$, with E in kV/cm and $\rho=(\text{kV/cm})^{-1}$. Since in the experiments the electric field strengths were smaller than 1 kV/cm, the terms with power higher in E than E^2 were neglected because they are several orders of magnitude smaller. T_R increases with increasing electric field strengths, as was found in the experiments. These calculations confirm the experimental results qualitatively as in the case of the

free-running superfluorescence [5,6].

In order to study what happens if the external static electric field is not homogeneous, the collective emission was observed in an inhomogeneous electric field. The experimental data were obtained by observation of the collective emission in a confocal resonator. Since the curved mirrors were used as Stark plates, the static electric field is inhomogeneous. The maximum of the intensity of the superfluorescent emission shows the same qualitative behavior as in the case of the plane resonator. The square root of the maximum of the intensity decreases linearly with the square of the electric field strength. But this decrease cannot be reproduced by a change of the characteristic time constant T_R (Fig. 6) as in the case of a homogeneous static electric field (Fig. 5).

In the case of an inhomogeneous static electric field, spatial variations of the field strengths over the cross section of the cavity will result in different Stark shifts of atoms at different points. Therefore, the atomic dipoles radiate at different wavelengths, leading to a loss of phase coherence between the emitting dipoles. This leads to a damping of the macroscopic polarization and to a change of the relaxation time T_2' ($1/T_2' = 1/T^* + 1/T_2$). A sequence of pulses of decreasing intensity was now calculated by solution of Eqs. (3) and (4), where the relaxation time T_2' was decreased stepwise. The data of the simulated pulses reproduce the experimentally observed dependence between the maximum of the intensity and the delay time of the pulses very well (Fig. 6). These simulations show that the calculated parameters of the collective emission for the used inhomogeneous electric field depend more sensitively on the relaxation time T_2' than on the characteristic time constant T_R. If the inhomogeneity of the electric field is smaller, then one has to change T_2' and T_R simultaneously in order to describe the collective emission.

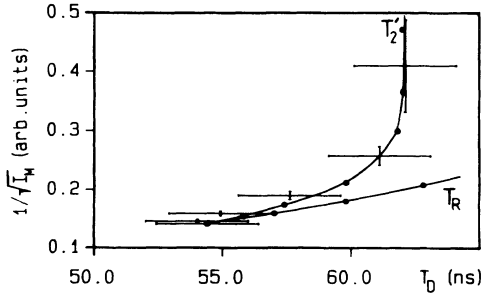

FIG. 6. Experimental (+) and simulated (○) data (see text) of a collective emission in a confocal resonator for the transition $6s10d\ ^1D_2 - 6s7f\ ^1F_3$ of Ba. Simulated data were obtained by solution of the Maxwell-Bloch equations for increasing values of the characteristic time constant T_R or decreasing values of the relaxation time T_2' (from left to right).

In conclusion, it can be stated from the experimental results that an external static electric field can inhibit superfluorescent emission in a cavity. The square root of the maximum intensity decreases linearly with the square of the electric field strength. This is the same dependence as in the case of the free-running superfluorescence [5,6]. In a homogeneous electric field this decrease is most likely due to a change of the characteristic time constant of the emission process, whereas in an inhomogeneous electric field it is due to the relaxation time.

ACKNOWLEDGMENTS

We would like to thank Professor Dr. A. Steudel for his interest in this work and for helpful discussion. We are indebted to the Regionales Rechenzentrum Hannover for making computer facilities available.

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