Observation of two-photon noise-initiated fluctuations in far-infrared Dicke's superradiance

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The anticorrelated fluctuation of the intensity of a driver beam pumping a Λ-like three-level system in the first transition from the initial to the intermediate level and that of the intensity of the radiation generated in the second transition from the intermediate to the final level initiated by noise is observed in an experimental arrangement designed for the investigation of far-infrared Dicke's superradiance. The fluctuation is interpreted as the result of a two-photon back-and-forth transition between the initial and the final level, emitting (absorbing) and absorbing (emitting) photons in the first and the second transition depending on the direction of the two-photon transition. The influence of this two-photon back-and-forth transition on the Dicke's superradiance is discussed.

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I. INTRODUCTION

For the observation of the cooperative radiation of the ensemble of atoms, the ensemble has either to be excited simultaneously by a δ-function pulse [1] or the excitation has to propagate along the axis of the cylindrically shaped volume containing the ensemble of atoms such that the superradiance pulse propagating along the axis meets always newly excited atoms. The latter arrangement results in the so-called swept-gain superradiance [2].

The experimental realization requires that the atoms are excited by a pulse of resonant radiation of finite duration with the trailing edge sharply cut, propagating along the axis of the volume [3]. The area of the exciting pulse is usually a multiple of π. Because of the difficulty of the excitation of a two-level system, three-level atomic or molecular systems are used by exciting the common upper level in one transition and the superradiance is observed on the other transition from the common level.

The excess area of the exciting pulse and the possibility of two-photon transitions in the three-level system causes a complication in the preparation of the atomic ensemble for Dicke's superradiance since the tipping angle of the Bloch vector at the beginning of the superradiance process depends on the duration of the exciting pulse [4]. Furthermore, the Raman-like fluctuation of the three-level system between the initial and the final states involving two photons, where one is emitted and one is absorbed, cannot be avoided in the case of the swept-gain superradiance by long-pulse excitation with a pulse area greater than π because of the simultaneous presence of the superradiant pulse and the driving radiation.

The superradiance process can be applied to ultrashort-pulse generation especially in the far-infrared (FIR) region [5,6] of the spectrum. This, however, requires a detailed study of the *per se* interesting processes mentioned above and their influence on the short-pulse formation.

The observation of the two-photon fluctuations in Dicke's superradiance experimental arrangement and the investigation of its features are the subjects of this paper.

II. TWO-PHOTON FLUCTUATIONS

It has been known for a long time that a Λ-like three-level system (Fig. 1) under the influence of two intense electromagnetic beams will make a two-photon transition [7] from the initial (1) to the final level (2) by absorbing one photon from the first beam (ω) and emitting one photon into the second beam (ω'). In the case of a long duration of the interaction with the two beams, the system returns to the initial state with the reverse process absorbing one quantum from the second beam (ω') and emitting one quantum into the first beam (ω). For an even longer duration of interaction the system repeats the cycle periodically. This type of process is also well known for a system where the energies of the initial and the final states are identical. In this case the process continues without any absorption from the exciting beams and leads to a complete mixing of the initial and final states. This phenomenon is called dark resonance [8].

For preparing an ensemble of atomic three-level systems for Dicke's superradiance, one excites the systems by a single strong electromagnetic wave (ω) to the intermediate energy level (0) in a time short compared to the relaxation time of the intermediate level and to the time of the formation of Dicke's superradiance. The relaxation time is determined either by the natural radiation de-

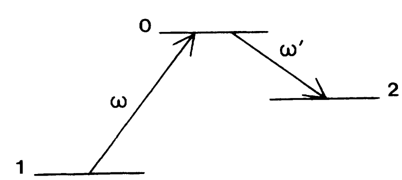

FIG. 1. The scheme of a three-level system interacting with two strong electromagnetic fields.

cay or by intermolecular collisions.

In the experiments performed previously [3,5,6] the duration of the excitation was usually longer than the formation time of Dicke's superradiance. This duration was in the range of the collisional-relaxation time of the intermediate level. The exciting radiation with typically 50-mJ pulse energy and 60-ns pulse duration was so strong that the system performed many Rabi cycles between the initial and the intermediate levels as in the case of NH_3, where the dipole moment of the transition is 0.2 D.

In order to observe Dicke's superradiance the exciting beam is cut in a time short in comparison to the time constants listed above. The duration of the cut versus the phase of the Rabi oscillation of the excitation determines whether the ensemble of the system is left in the fully or only partially excited state at the end of the excitation. This is implied by the fact that the tipping angle of the optical Bloch vector of the second transition from the intermediate level to the final level at the beginning of the transition, i.e., at the beginning of the formation of the superradiant pulse, depends crucially on the duration of the exciting pulse [4].

At higher density, i.e., at higher pressure of the molecules, the superradiant pulse initiated by noise appears simultaneously to the exciting pulse if the formation time of the superradiant pulse becomes smaller than half of the duration of the exciting pulse. In this case the two-photon fluctuation of the system between the initial (1) and the final level (2) of the system under the simultaneous influence of the exciting driver beam and the noise-initiated superradiance induced field has to be taken into account. This fluctuation can be observed as anticorrelated fluctuations in the driver and the induced fields. The rate of the fluctuation between the initial and the final states, which is some type of relaxation oscillation, can be calculated by using Javan's theory [9] of the three-level maser for the two-photon transition because the second induced field is comparatively weak, partly because of the wavelength difference between the driving and the induced fields and partly due to the comparatively low efficiency of the superradiant-pulse formation which is stopped at the start of the two-photon fluctuation process. The two-photon transition rate is always of the same order of magnitude as or less than the homogeneous linewidth multiplied by π, i.e., the relaxation rate of the levels. The two-photon transition rate was calculated on the basis of the results of the above-mentioned theory and the measured intensities of both the driving and the induced fields, whose intensities are averaged over the surface perpendicular to the propagation direction of the driving beam. Therefore the application of the results of Javan's theory seems to be justified, at least as an approximation.

The transition probability (W_{2p}) can be calculated with the aid of the expression for the gain (G) of the second (ω') field published by Javan [9] and later by Seligson *et al.* [10] for the case when both transitions are pressure broadened as

$$W_{2p}=\frac{GcE'^2}{h\omega' N}\ , \tag{1}$$

where c, E', h, and N are the velocity of light, the field strength of the second field, Planck's constant, and the number of the three-level systems in the initial state before the interaction of the fields with the systems. On the basis of the expression for the gain formulated by Seligson *et al.* [10] we find for the transition rate from the initial state (1) to the final state (2) and back

$$W_{2p}=\frac{2\pi\alpha^2\beta^2\gamma}{a^2}\left[\frac{1}{2}\left\{\frac{1}{\gamma^2+(a+b)^2}+\frac{1}{\gamma^2+(a-b)^2}\right\}+M\right] \tag{2}$$

and

$$M=\frac{(a^2-b^2)(2a^2+\gamma^2)-\gamma^4}{(4a^2+\gamma^2)[\gamma^2+(a+b)^2][\gamma^2+(a-b)^2]}\ ,$$

where $\alpha=\mu E/h$ and $\beta=\mu' E'/h$ are the Rabi frequencies for the two transitions while μ and μ' are the dipole moments of the first and second transitions; $a=(\Delta^2+\alpha^2)^{1/2}/2$, and $b=\frac{1}{2}\Delta-\Delta'$. In these equations $\Delta=\omega-\omega_{10}$ and $\Delta'=\omega'-\omega_{20}$ represent the detunings of the frequencies of the two fields from the atomic transition frequencies ω_{10} and ω_{20}. The relaxation rates γ of the levels are identical.

III. EXPERIMENTAL ARRANGEMENT

The experimental arrangement (Fig. 2) is similar to those generally applied to the investigation of the superradiant-pulse formation [3]. The vapor of NH_3 molecules in a cell of 3-m length is excited by the beam of a hybrid transversely excited atmospheric pressure (TEA) CO_2 laser whose wavelength is tuned to the $10R6$ emission nearest to the absorption line of the NH_3 molecule by a diffraction grating which serves as the back mirror of the laser cavity. The pulse of the CO_2 laser is abruptly cut by a plasma shutter triggered by the laser-triggered spark gap (LTSG) before the beam enters the cell [5,6].

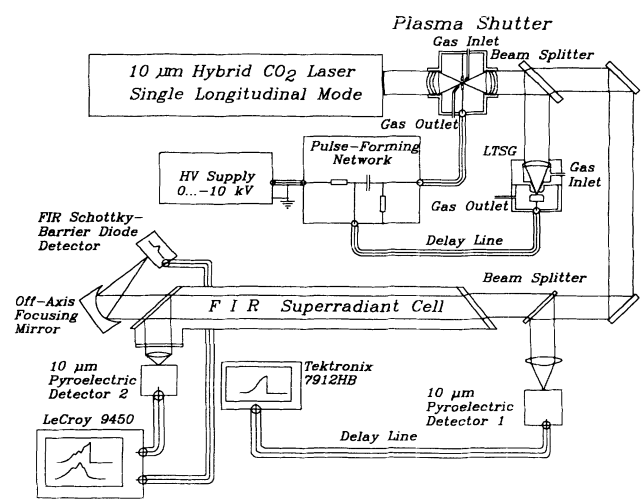

FIG. 2. The experimental setup.

The shape of the driver pulse is registered by fast pyroelectric detectors just before the beam enters the cell and immediately after leaving the cell. The radiation emitted in the second transition of the Λ-like system (see Fig. 1) with a wavelength of 291 μm is separated from the transmitted driver beam by a quartz plate mounted at 45°. It transmits the far-infrared radiation and reflects the driver beam through a salt window to the detector. The far-infrared pulse is detected by a fast Schottky diode. The signals of the different detectors are registered simultaneously by fast digital wave-form analyzers.

During the measurement the pressure of the molecular vapor is varied in the cell and the three signals, i.e., the pulse shape of the driver beam before the cell, that of the transmitted driver pulse, and that of the far-infrared pulse are registered as functions of the molecular pressure.

IV. RESULTS AND DISCUSSIONS

One record of the wave-form analyzers is shown in Fig. 3 for a comparatively low pressure of 0.1 Torr. The far-infrared pulse is formed long after the end of the driver pulse. The duration of this pulse and the delay from the end of the driver pulse depend linearly on the inverse pressure. This relation can be interpreted [5] as an indication of a superradiance pulse.

In order to shorten this pulse the gas pressure is increased above 1.2 Torr. Thus the superradiant pulse forms at the very end of the driver pulse or even merges into it. From theory it is expected that the superradiance pulse approaches the center of the driver pulse [11] if the vapor pressure is increased.

However, the merging of the superradiant pulse into the driver pulse is connected with the appearance of a simultaneous fluctuation of both the intensity of the far-infrared radiation and the intensity of the transmitted driver beam (Fig. 4). These fluctuations are anticorrelated. The sudden drop in the intensity of the CO_2 radiation drives a pulse of the FIR radiation which peaks at the time of a minimum of the intensity of the driver beam. In a next step the FIR radiation absorbed by the process of back transition and consequently, the intensity of the CO_2 radiation increases. The local maximum of the intensity of the CO_2 radiation coincides again with the minimum of the intensity of the FIR radiation. This anticorrelated fluctuation can be interpreted as the result of a flip-flop transition of the three-level system between the initial and the final levels of the two-photon transition

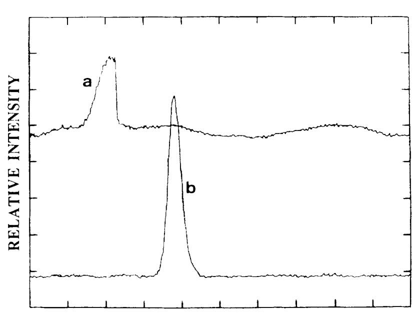

FIG. 3. The shape of the driving pulse (a) and that of the superfluorescence pulse (b) at a pressure of 0.1 Torr for the molecular vapor in the cell.

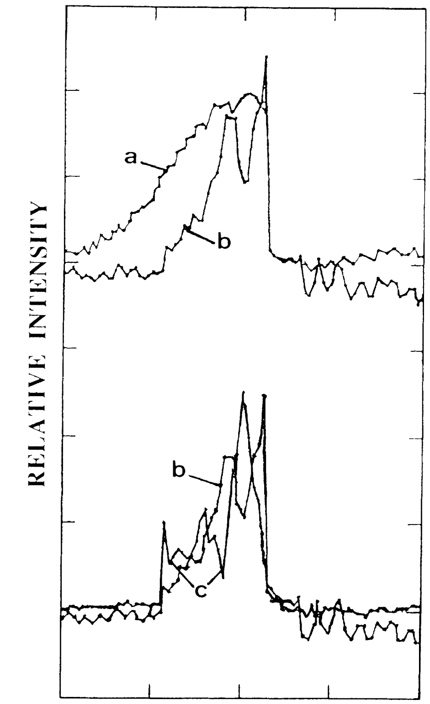

FIG. 4. The pulse shape of the driving beam before entering the cell (a); the pulse shape of the driving beam after propagation through the cell (b); and the pulse shape of the far-infrared superradiance pulse (c) if simultaneous interaction of the induced far-infrared radiation and the driving beam takes place with a Λ-like three-level system. The NH_3 pressure in the cell is 1.7 Torr.

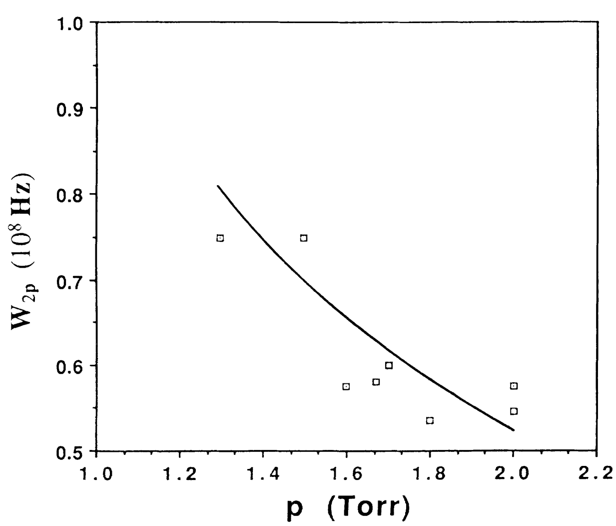

FIG. 5. The frequency of the fluctuation in dependence on the pressure of the vapor in the cell. The solid curve is the result of the calculation according to Eq. (2). The squares are the results of the measurement.

under the influence of two strong fields where one, i.e., the FIR field, is generated from noise in a superradiant process.

The rate of the fluctuation is calculated by Eq. (2) and the measured intensities of the CO_2 and the FIR radiation. The result is compared with the frequencies of the fluctuations derived from figures similar to Fig. 4 at different vapor pressures in the cell. The result is illustrated in Fig. 5. The comparatively good agreement between the theoretical curve and experimental points justifies the interpretation of the observed fluctuation as a result of two-quantum back-and-forth transitions between the initial and the final levels of a Λ-like three-level system.

V. CONCLUSIONS

The anticorrelated fluctuation in the intensity of the infrared driver and the FIR induced field in the case of their simultaneous presence is interpreted as a result of periodic back-and-forth fluctuation of the Λ-like three-level system. This observation indicates a possible destructive role of this fluctuation in the swept-gain superradiance which is usually proposed for ultrashort-pulse generation especially in the FIR region of the electromagnetic spectrum.

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