Observation of Dicke Superradiance in Optically Pumped HF Gas*

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We have found that room-temperature HF gas at millitorr pressures, optically pumped to produce a total inversion at a rotational transition in the $v=1$ vibrational state, emits a superradiant pulse with ringing after a considerable delay (~ microseconds). A semiclassical analysis shows that for a high-gain system the pulse evolution is determined by a single parameter τ_R, and that inhomogeneous broadening is unimportant. Close agreement between theory and experiment is obtained.

This paper reports the first detailed study of Dicke superradiance[1,2] in the optical region.[3] In the experiments a long sample cell of low-pressure (~millitorr) HF gas is pumped by an intense short pulse from an HF laser operating on an R or P branch transition to the vibrational ground state ($\lambda \sim 2.5$ μm). This produces a nearly complete population inversion between two adjacent rotational levels in the $v=1$ state, corresponding to transitions in the 50- to 250-μm range.[5] After a considerable delay (~microseconds) a burst of radiation appears at the rotational transition (Fig. 1). The formation, delay, and ringing of this radiation pulse are of major interest in this study.

As pointed out by Dicke,[1] a totally inverted two-level system can give rise to a superradiant pulse. In the Bloch formalism the initial state corresponds to a vector pointing straight up, in analogy to the unstable equilibrium position of a rigid pendulum standing exactly on end. Though this state cannot radiate classically, a macroscopic polarization develops in the medium, triggered by incoherent spontaneous emission and background thermal radiation, the "perturbing field." In the absence of collisions the system gradually evolves into a superposition of superradiant states, corresponding to the Bloch vector pointing sideways, at which time the radiation pulse is emitted. This pulse can be considerably delayed with respect to the sudden switching on of the population inversion. In order to produce long delays it is necessary to excite the system without leaving a macroscopic polarization in the medium. This condition is difficult to achieve by coherent excitation of a two-level system but it is easily fulfilled by optically pumping the system via a coupled transition.

In numerous treatments of superradiance the radiation field is quantized and the atomic system is described in terms of collective Dicke states.[5] As pointed out by Arecchi *et al.*,[6] these atomic states can be used to construct a new set of states, called Bloch states, which have similar properties to Dicke states, but may be treated semiclassically by means of the coupled Maxwell-Schrödinger equations.[7] The semiclassical approach is not justified during the initial stage of pulse evolution, where stimulated emission is not yet dominant.[8] However, as seen below, the form of the initial fluctuations is unimportant. Adopting this approach, we use the coupled Maxwell-Schrödinger equations,[9] including homogeneous and inhomogeneous broadening, linear loss (diffraction), and level degeneracy, to treat a weak perturbing field, simulating spontaneous emission and blackbody radiation, incident on a rod-shaped medium which is inverted at $t=0$. An

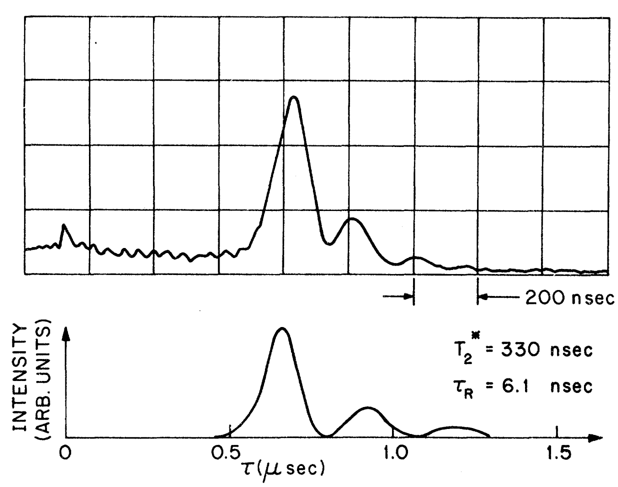

FIG. 1. Oscilloscope trace of superradiant pulse at 84 μm ($J=3\rightarrow 2$), pumped by the $R_1(2)$ laser line, and theoretical fit. The parameters are $I=1$ kW/cm^2, $p=1.3$ mTorr, and $kl=2.5$ for $l=100$ cm. The small peak on the scope trace at $\tau=0$ is the 3-μm pump pulse, highly attenuated.

important result of the analysis[10] is that the pulse evolution can be described simply in terms of only two parameters: τ_R, a characteristic time for the radiation damping of the collective system, and αl, the linear field gain;

$$\tau_R = 8\pi(N\lambda^2 l)^{-1}T, \tag{1}$$

and

$$\alpha l = T_2^*/\tau_R, \tag{2}$$

where λ is the wavelength, N is the net population-inversion density, T the radiation lifetime of an isolated atom, T_2^* is the dephasing time of the rotational transition,[11] and l is the length of the sample cell. In particular:

(1) The area of the output pulse is completely determined by the gain αl and the area of the perturbing field over the first few τ_R, $\theta(0)$, in accordance with the area theorem of McCall and Hahn,[12] valid for an inhomogeneously broadened nondegenerate[13] system. Since in our case $\theta(0) \sim 10^{-8}$, gains in excess of 20 are needed for appreciable pulse buildup. Such gains are available in HF at millitorr pressures.[4]

(2) For a given value of $\alpha l \gg 1$, the time scale of the pulse evolution (i.e., delays, pulse shapes) is completely determined by τ_R.

(3) For fixed αl and τ_R, inhomogeneous broadening, level degeneracy,[13] and a reasonable amount of linear loss change the results in minor ways.[14]

(4) The pulse delay τ_D depends linearly on τ_R,[10]

$$\tau_D = [\ln\theta(0)]^2\tau_R/4, \tag{3}$$

and this is confirmed in the experiments.

The change in delay caused by perturbing fields of different sizes is related to the problem of gently tipping over a rigid pendulum balanced on its end: The stronger the initial tap, the faster the pendulum will fall. Also, as with a pendulum, only the first few degrees of tipping progress slowly. Once the Bloch vector has tipped appreciably, it falls the rest of the way rapidly (in a few τ_R).

Numerical calculations, described below, confirm these predictions and also lead to the following conclusions:

(a) The effects of relaxation are unimportant as long as the homogeneous relaxation time T_2 exceeds the pulse delay. (At the millitorr pressures of the experiments, T_2, determined by collisions, is always much longer than the delays.)

(b) The effectiveness of the perturbing field is limited to the first few τ_R's. Furthermore, the system behavior is insensitive to the exact form of the initial perturbation. δ-function, step-function, Gaussian pulses, and pulse trains of intensity consistent with the magnitude of the background radiation field all give output pulses of the same shape and delay. Furthermore, the presence of random jumps in the phase of the input field to simulate spontaneous emission does not significantly affect the output pulses. This justifies our approach.

(c) The output pulses are insensitive to the specific time dependence of the population excitation.

The experimental arrangement consisted of an HF pump laser, a sample cell, and the detection system. The pump laser, a helical pin laser described previously,[4] produced R_1 (J) and P_1 (J) pulses of 200–400 nsec duration and peak powers of a few kilowatts per square centimeter. The stainless steel sample cells ranged in length from 30 to 100 cm, with inner diameters between 12 and 28 mm. They had silicon Brewster windows coated on the inside with a thin layer of Halocarbon stopcock grease to prevent corrosion. The HF gas was purified by freezing and distillation. The pressure in the sample cell was estimated from the linear attenuation of the pump laser output. To observe the superradiant pulses a helium-cooled In-Ge detector was used, followed by a fast preamplifier and a pulse amplifier. The overall rise time of the system was below 10 nsec, giving ample resolution of the pulse shapes.

The HF lines studied were the $J+1 \to J$ rotational transitions in the $v=1$ band, with J ranging from 0 to 4, corresponding to the wavelengths 252, 126, 84, 63, and 50 μm. All of these lines have been obtained previously by optical pumping at much higher pressures.[5] The sample cell was optically pumped in a single pass, leading to superradiant output pulses with peak intensities estimated to be in the 100 $\mu W/cm^2$ range and widths in the range of 50 to several hundred nanoseconds, depending on the gas pressure and pump laser power. This corresponds to pulse areas of order unity. Care was taken to ensure that no far-infrared radiation from the pump laser entered the sample cell.

Since both pump and rotational transitions are Doppler broadened, the gain is independent of the pump laser intensity provided the pump transition is saturated.[10] Therefore, in our experiments the gain is selected by fixing the sample cell pressure, and τ_R may then be adjusted by

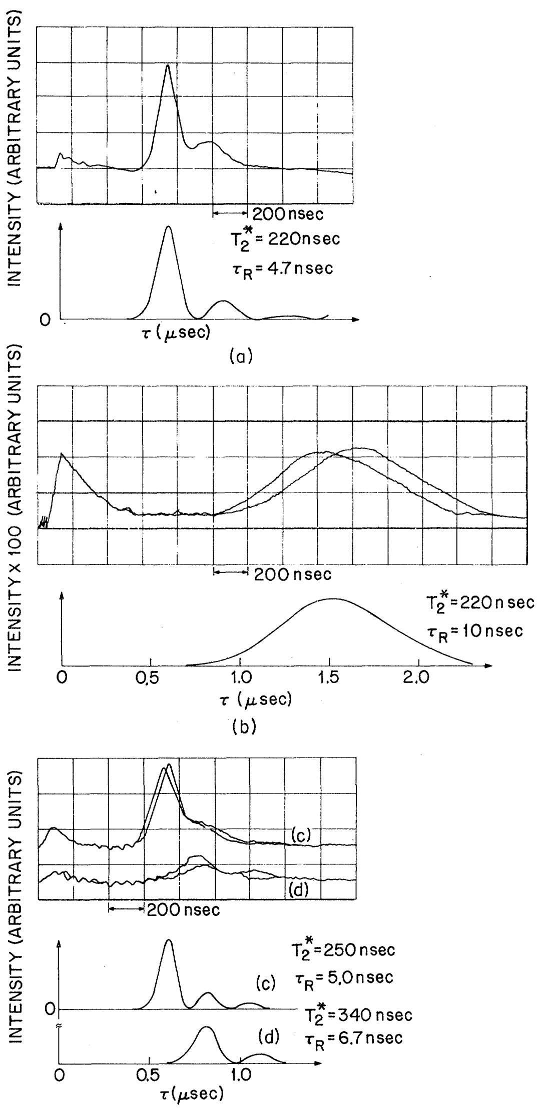

FIG. 2. Oscilloscope traces of superradiant pulses and computer fits. (a) $J=3\rightarrow 2$ transition at 84 μm pumped by $P_1(4)$ laser line. $I=2.2$ kW/cm^2, $p=4.5$ mTorr, $\kappa l=2.5$ ($l=100$ cm). (b) Same as (a) but $p=2.1$ mTorr. Note increased delay and broadening of pulse. (c) Same transition as in (a), but pumped by $R_1(2)$ laser line. $I=1.7$ kW/cm^2, $p=1.2$ mTorr, $\kappa l=3.5$ ($l=100$ cm). (d) Same as (c) except $I=0.95$ kW/cm^2. The same intensity scale is used in fitting curves (a) and (b), and (c) and (d). Note the reproducibility of the oscilloscope traces in double exposure.

varying the pump intensity.

Figures 1 and 2 show oscilloscope traces of superradiant pulses under various experimental conditions. At pressures below 5 mTorr the pulses were delayed by 500 to 2000 nsec past the beginning of the pump pulse. Decreasing the pressure increased the delays, broadened the pulses, and decreased their magnitude, in agreement with theoretical expectations [Figs. 2(a) and 2(b)].[15] 2-μsec delays, the longest observed, occurred at pressures below 1 mTorr. (At high pressures, i.e., above 10 mTorr, the pulses were single spikes 40 nsec in width and occurred during the pump pulse.) Furthermore, decreasing the pump power increased the pulse delay and width and decreased its amplitude [Figs. 2(c) and 2(d)], again in agreement with theory. The pulses often exhibited ringing with as many as four lobes (Fig. 1). Similar pulses were also seen in the backward direction, i.e., propagating antiparallel to the traveling-wave pump radiation.

To compare experiment with theory we have integrated the coupled Maxwell-Schrödinger equations given in Eq. (208) of the article by Icsevgi and Lamb,[9] modified to include level degeneracy, as is necessary in treating molecular systems. All parameters are expressed in terms of the three experimental variables, the pump field intensity I, the sample cell pressure p, and the incremental loss κ. These values, estimated from the experimental conditions, are given in the figure captions. In addition, it is necessary to specify the pump transition used. All the calculations assume a collision time of $T_2=7$ μsec at 1 mTorr. An input field envelope of constant amplitude is used. Its magnitude E_0, estimated from blackbody radiation and spontaneous emission intensity (~ 10^{-14} W/cm^2), depends on I and p and ranges from 10 to 50 sec^{-1}, in units of $\mu E_0/\hbar$. The rotational levels are inverted over a period of ~ 100 nsec to take account of the pump pulse duration. As can be seen in the figures good agreement is obtained throughout.

The presence of feedback in our high-gain system drastically shortens the pulse delays, as was verified by intentionally introducing feedback. Estimates show that for gains typical of our experiments feedback of the order of 10^{-4} to 10^{-5} is permissible. Scattering off the cell windows into the gain volume is estimated to be at least a factor of 10 smaller than this. Other sources of feedback are even less important. The absence of regeneration was verified by placing a second HF cell after the first one, with a common window between them oriented at Brewster's angle. Both were excited by the same pump laser beam. The first cell was filled to produce long delays, and the second cell was filled to produce large signals and short delays. The pulses of the first cell were observed through a side window positioned to intercept the weak reflection off the common window. The delay and appearance of these pulses was not affected by the presence of the second cell, demonstrating the absence of significant amounts of feedback.

In conclusion, we would like to stress that a superradiant pulse can evolve in an inhomogeneously broadened extended sample. This is because in an amplifier, in contrast to an absorber, dephasing is counteracted by high gain, giving rise to an effective dephasing time[10] $\alpha l T_2^*$. This time is always considerably longer than the pulse evolution time τ_D. Furthermore, the bandwidth of of the initial perturbation is larger than $1/T_2^*$.[10] Therefore, the time scale of the pulse evolution (determined by τ_R) depends on the *total number* of excited molecules, irrespective of the extent of inhomogeneous broadening. These points are verified in the experiments.

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[3]A number of coherent optical effects closely related to the concepts used to treat superradiance have been observed. These are reviewed by E. Courtens, in *Laser Handbook,* edited by F. T. Arecchi (North-Holland, Amsterdam, 1972), p. 493.

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[5]See R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A 4, 302 (1971), and references contained therein.

[6]F. T. Arecchi, E. Courtens, R. Gilmore, and H. Thomas, in *Fundamental and Applied Laser Physics: Proceedings of the Esfahan Symposium,* edited by M. S. Feld, N. A. Kurnit, and A. Javan (Wiley, New York, 1973).

[7]F. T. Arecchi and E. Courtens [Phys. Rev. A 2, 1730 (1970)] have also pointed out that superradiance may be treated semiclassically.

[8]This stage extends over a period $\sim\tau_R$, as can be seen by calculating the time necessary for the first photon to be emitted into the diffraction mode determined by the sample geometry. Also note that the pulses evolve over a period of $\sim 100\tau_R$ [cf. Eq. (3)].

[9]See, for example, A. Icsevgi and W. E. Lamb, Jr., Phys. Rev. 185, 517 (1969).

[10]I. P. Herman, J. C. MacGillivray, N. Skribanowitz, and M. S. Feld, to be published.

[11]The inhomogeneously broadened linewidth of the rotational transition is $\Delta = 1/T_2^*$. For a Doppler-broadened transition $\Delta = ku/\pi^{1/2}$, where ku is the half-width of the Doppler profile at the $1/e$ point in units of circular frequency.

[12]S. L. McCall and E. L. Hahn, Phys. Rev. 183, 457 (1969).

[13]The consideration of C. K. Rhodes, A. Szöke, and A. Javan [Phys. Rev. Lett. 21, 1151 (1968)] applied to an amplifier show that level degeneracy does not inhibit pulse evolution and that pulses of increasing energy but stable area near π can evolve.

[14]These approximations lead to an analysis similar to that of D. C. Burnham and R. Y. Chiao [Phys. Rev. 188, 667 (1969)] applied to an amplifier rather than to an absorber.

[15]At high pressures the signal size also decreased with decreasing pump intensity. At intermediate pressures (~10 mTorr), however, decreasing the pump power actually increased the signal amplitude.

Collective Excitations in Superfluid Helium

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Collective excitations of the phonon gas in liquid helium-4 are considered. It is proposed that below 0.5°K there are other propagating collective excitations in addition to second sound.

In liquid helium-4 below the λ point it is well known that temperature waves (second sound) can propagate through the phonon-roton gas.[1] Temperature waves propagate because both energy and momentum are conserved in collisions between excitations in superfluid helium. It has recently been proposed[2-4] that the dispersion curve for phonons in helium-4 is anomalous in