Superfluorescence and Amplified Spontaneous Emission of 29-cm^{-1} Phonons in Ruby

M. H. F. Overwijk, J. I. Dijkhuis, and H. W. de Wijn
Faculty of Physics and Astronomy, and Debye Research Institute, University of Utrecht, P.O. Box 80000, 3508 TA Utrecht, The Netherlands
(Received 24 July 1990)

Superfluorescence and amplified spontaneous emission of 29-cm^{-1} phonons are observed following inversion of the $\bar{E}(^2E)$-$2\bar{A}(^2E)$ acoustic transition of Cr^{3+} in ruby. The conditions for both of these manifestations of phonon avalanches are selected via the Cr^{3+} concentration. Superfluorescence is described in terms of a pendulum equation of the acoustic Bloch vector. Rate equations of the level and phonon populations account for amplified spontaneous emission.

PACS numbers: 63.20.−e, 42.50.Fx

Superfluorescence and amplified spontaneous emission are well-known phenomena in the release by optical emission of energy stored in ensembles of inverted two-level systems.[1] By contrast, the acoustical analog, i.e., superfluorescent emission of phonons, has not been observed up until now, although its existence was suggested.[2] Evidence for incoherent phonon avalanches is available,[3] but only scarce as concerns phonons of frequencies beyond ultrasound (>30 GHz).[4–7] In this Letter, we report on the first observation of superfluorescence of high-frequency phonons, and further show that by suitable modification of the conditions the emission becomes incoherent.

The acoustic transition used in the present experiments is the $\bar{E}(^2E)$-$2\bar{A}(^2E)$ transition in optically excited ruby (Fig. 1). Thermal phonons are removed by immersion in liquid helium at 1.5 K. The inversion is achieved by optical excitation from the $^4A_2$ ground state to the $2\bar{A}(^2E)$ level, which is situated at a distance of 29 cm^{-1} (0.87 THz) above $\bar{E}(^2E)$. To this end, optical pulses of 30-μJ energy were delivered by an amplified synchronously pumped picosecond dye-laser system at a repetition rate of 30 Hz. An advantage of this pumping scheme is that initially the inversion is complete and free of transverse polarization. In terms of the well-known Bloch picture, this corresponds to an *acoustic* Bloch vector pointing upwards. The scheme further allows one to follow the ensuing phonon avalanche as it develops on picosecond time scales by monitoring the dephasing of the *optical* transition[8] with two-pulse photon echo, where the optical pulse inverting the $2\bar{A}(^2E)$-$\bar{E}(^2E)$ acoustic transition at the same time serves as the pump pulse of the photon-echo experiment. The technique of optical gating was applied to suppress signals from Rayleigh-scattered light.

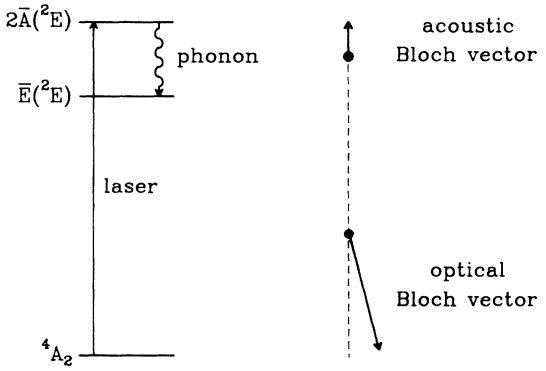

FIG. 1. Principle of inverting an acoustic transition by optical pumping, and the initial orientations of the Bloch vectors.

A criterion for the occurrence of (optical) superfluorescence has been devised by Schuurmans and Polder,[9] and verified to be quite sharp by Malcuit *et al.*[10] It states that the dephasing time T_2^* of the transition must satisfy

$$T_2^* > (T_R T_D)^{1/2}, \tag{1}$$

in which T_R is the superradiant lifetime and T_D is the time span until emission of the coherent pulse. The time T_R can be interpreted as the collective damping time of the radiating dipoles in a diffraction-limited pencil-shaped volume with a length equal to the coherence length. The criterion can, of course, only be applied in retrospect after values for T_R and T_D have been deduced from an analysis in terms of an evolving acoustic Bloch vector.

In order to fulfill the criterion for superfluorescence, large T_2^* are imperative. In very dilute ruby, the acoustic transition is of predominantly homogeneous nature, implying the largest feasible T_2^*. Dilution, however, has to be traded in to some extent for the sake of sufficient signal intensity. In the present sample, having a Cr^{3+} concentration of 1000 at. ppm, $T_2^* \approx 600$ ps.[11] Estimates for T_D and T_R deduced from the experiments below yield the sufficiently small value of $(T_R T_D)^{1/2} \approx 400$ ps. In order to pass on into the regime of amplified spontaneous emission while preserving sufficient signal intensity, we opted to reduce T_2^* by augmenting the primary concentration of Cr^{3+} centers. In 2500-at. ppm ruby, T_2^* is reduced to 270 ps,[11] but the in-

crease in the primary concentration also results in a reduction of $(T_R T_D)^{1/2}$ to about 300 ps at constant laser power. The experiments on the 2500-at.ppm sample were, therefore, repeated at half the laser power, in which case the conditions for amplified spontaneous emission are satisfied with more certainty.

For the case of 1000-at.ppm ruby, a typical decay of the photon echo of the $2\bar{A}(^2E)$-$^4A_2$ transition is given in Fig. 2 as a function of the delay t_{12} between the pump and probe pulses. Note that the time at which the echo appears is $2t_{12}$. The laser beam runs parallel to the crystalline c axis, and is focused in the sample. At short times, Rayleigh scattering dominates. From the echo intensity of the $2\bar{A}(^2E)$-$^4A_2$ transition in the absence of a pump pulse (lower trace in Fig. 2), it is seen that beyond $t_{12} \approx 150$ ps Rayleigh scattering ceases to contribute. The most salient feature of the further development of the decay is that at first it does not markedly speed up in comparison with the spontaneous decay, which has a time constant $T_1 = 700 \pm 50$ ps,[12] but that a significant acceleration sets in only after a delay $t_{12} \sim 400$ ps. Such a behavior is indeed typical for superfluorescence.[1] In connection with the experiments, it is finally noted that the photon-echo intensity of the $\bar{E}(^2E)$-$^4A_2$ transition decays only minutely in the present time window, which demonstrates that the optics remains correctly aligned during the scan of the optical delay line.

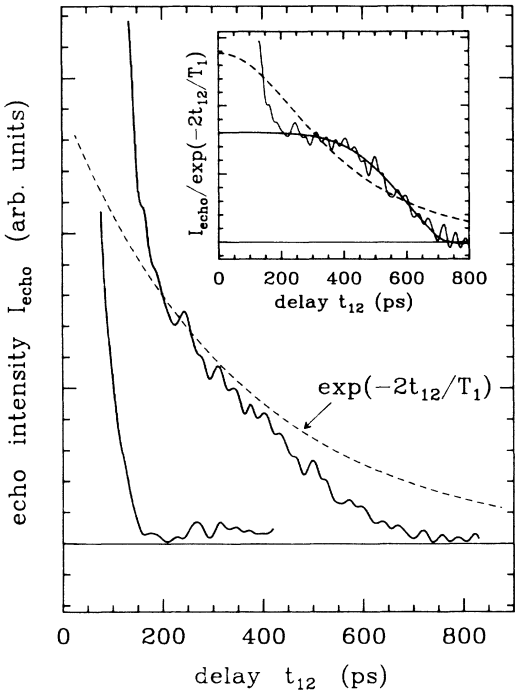

FIG. 2. The photon-echo intensity vs t_{12} for the $2\bar{A}(^2E)$-$^4A_2$ transition in ruby with 1000-at.ppm Cr^{3+}. Lower trace represents Rayleigh scattering of the probe beam only. Inset: Photon-echo intensity over $\exp(-2t_{12}/T_1)$ vs t_{12}, where the solid curve is based on the pendulum equation, and the dashed curve on rate equations.

If all decay times are long compared to T_R, the tumbling over the initially inverted acoustic Bloch vector specifying the ensemble of two-level systems can, for the simple case of strictly homogeneous broadening, be described by the pendulum equation[2]

$$\frac{d^2\psi}{dw^2} + \frac{1}{w}\frac{d\psi}{dw} = \frac{\sin\psi}{T_R L_c}, \tag{2}$$

in which w is defined by $w = 2(xT)^{1/2}$, with x the spatial coordinate and T the retarded time, and the dependent variable ψ is the angle the acoustic Bloch vector makes with respect to the vertical. The inversion density ΔN and the phonon polarization P are the longitudinal and transverse projections of the Bloch vector, respectively. If the spontaneous decay time cannot be ignored in comparison with T_R, as is the case for phonon superfluorescence, a correction has to be made for the removal of coherent sites from the superradiant ensemble. Slightly generalizing the heuristic approach of MacGillevray and Feld,[1] we substitute the single-exponential decay in real time $\Delta N_0 e^{-t/T_1}$ for the initial inversion density ΔN_0, in order to obtain a modified (time-dependent) superradiant lifetime and modified projections. A second property of phonons is their small velocity v, which implies that the extent of the coherence is much more restricted than in the case of optical superfluorescence. For transverse phonons in ruby $v_t \approx 6.5$ km/s, and so the coherence length[13] $L_c \approx v_t T_D$ amounts to only a few μm. Accordingly, L_c is substantially smaller than the dimensions of the optically pumped zone. In other terms, there is no preferential mode as a result of the geometry,[14] but rather each spontaneously emitted phonon defines a mode from which a one-dimensional ray can develop.

The polarization associated with the echo scales with the square root of the number of optically coherent centers still residing in $2\bar{A}(^2E)$, i.e., centers that have not yet undergone dephasing by phonon-associated transitions to $\bar{E}(^2E)$. The total optical polarization then is obtained by integrating the spatial coordinate x over the coherence length. The intensity of the echo emerging at time $2t_{12}$ after applying the pump pulse thus reads

$$I_{\text{echo}} \propto \left[\int_0^{L_c} dx \{1 + \cos[\psi(2t_{12} - x/v_t)]\}^{1/2}\right]^2 e^{-2t_{12}/T_1}, \tag{3}$$

where the cosine should be set to unity for $x \geq 2v_t t_{12}$. Equation (3) has been fitted to the data beyond $t_{12} = 150$ ps with ΔN_0 as an adjustable parameter. The result of the fit is presented in the inset of Fig. 2, where we have divided the decay by the trivial time dependence e^{-2t_{12}/T_1} occurring in Eq. (3). The best agreement is found for $\Delta N_0 = 6.5 \times 10^{17}$ cm^{-3}. The superradiant lifetime at $t = 0$ is then $T_R \approx 110$ ps, and the corresponding $L_c \approx 3.8$ μm. All the features familiar from optical superfluorescence clearly emerge, particularly the char-

acteristic delay preceding coherent emission. While the agreement between experiment and Eq. (3) is gratifying, it should be noticed that Eq. (3) was derived for a single optical pump pulse at $t=0$. The probe pulse at $t=t_{12}$, of course, further enhances the inversion density, which, in turn, reduces T_R. We have investigated these effects on the assumption that the second pulse does not affect the transverse components of the acoustic Bloch vector. The results indicate that the initial ΔN_0 must be chosen somewhat larger, and that the true superfluorescent decay is slightly steeper than according to Eq. (3).

If the conditions for superfluorescence of phonons are not met, as in the case of 2500-at. ppm ruby under the present conditions, the decay of the ensemble of two-level systems may still be faster than the spontaneous decay as a result of amplified spontaneous emission. In this case, the inhomogeneous dephasing is so effective, i.e., the transverse components of the acoustic Bloch vector decay so rapidly, that no macroscopic polarization builds up that is large enough to bring about superfluorescence. Instead, the Bloch vector remains near vertical, and shortens until equilibrium is reached. In this process, the occupation number of the 29-cm^{-1} phonons rises, and, in fact, becomes appreciable if the inversion density ΔN is at least of the order of the density of phonon modes $\rho\Delta\nu$ resonant with the $2\bar{A}(^2E)$-$\bar{E}(^2E)$ transition. In the incoherent limit, the development of the occupation numbers of the $\bar{E}(^2E)$ and $2\bar{A}(^2E)$ states, and the occupation number $p(t)$ of the phonons resonantly interacting with this two-level system, are adequately described by rate equations.[3]

In Fig. 3, we present two representative examples of the two-pulse photon-echo intensity of the $2\bar{A}(^2E)$-$^4A_2$ transition as a function of the delay t_{12} for 2500-at. ppm ruby. The laser beam is perpendicular to the c axis.[15] Here, as we have pointed out, T_2^* associated with the phonon transition is short enough to anticipate an incoherent avalanche of phonons. The two traces differ by a factor of 2 in pumping power. Indeed, a marked acceleration is observed with respect to spontaneous decay, i.e., according to $\exp(-2t_{12}/T_1)$. The inset of Fig. 3 shows the photon-echo trace pertaining to the $\bar{E}(^2E)$-$^4A_2$ transition, the minor decay of which again confirms correct optical alignment. The oscillatory dependence of all these traces originates from the ground-state splitting.[12]

The initial conditions are similar to those in Fig. 2: The first pulse of the two-pulse sequence brings about an initially complete inversion of the $2\bar{A}(^2E)$-$\bar{E}(^2E)$ phonon transition, and phonons are initially absent. For incoherent avalanches, the intensity of the echo appearing at time $2t_{12}$ after applying a probe pulse at time t_{12} is

$$I_{\text{echo}} \propto \exp\left[-\int_0^{2t_{12}} \frac{1+p(t)}{T_1}\,dt\right], \tag{4}$$

where $p(t)$ is derived from the rate equations of the level

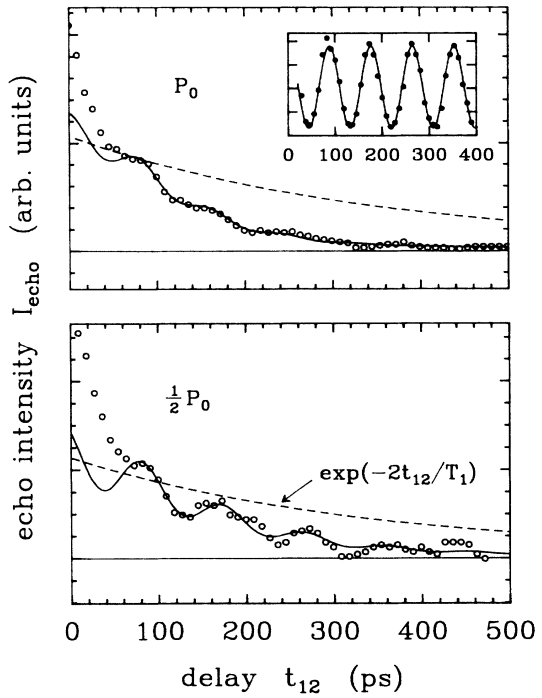

FIG. 3. The photon-echo intensity of the $2\bar{A}(^2E)$-$^4A_2$ transition vs t_{12} for ruby with 2500-at. ppm Cr^{3+} at maximum (upper frame) and half-maximum (lower frame) laser power. Solid curves represent fits to the data, and dashed curves spontaneous emission only. Inset: Photon-echo intensity vs t_{12} for the $\bar{E}(^2E)$-$^4A_2$ transition.

and phonon occupation numbers, given the initial inversion density ΔN_0. In calculating $p(t)$, the effect of the probe pulse is incorporated by properly augmenting the inversion density at time t_{12}. The probe pulse, being of double the intensity, in good approximation triples the number of optically excited centers. Equation (4) times an oscillatory dependence accounting for the beat was fitted to the data beyond 80 ps, by which time the Rayleigh scattering has died out (solid curves in Fig. 3). We thus find $\Delta N_0/\rho\Delta\nu \approx 3.3 \pm 0.2$ for maximum and 1.9 ± 0.3 for half-maximum laser power. Within the errors, these values scale linearly with the laser power, as they should. At maximum laser power, this corresponds to $\Delta N_0 \sim 3\times 10^{17}$, and to a phonon occupation at the end of the decay amounting to approximately 5.

It is important to realize that the decays caused by superfluorescence and amplified spontaneous emission are notably different in character. In the case of superfluorescence, acceleration does not occur until sufficient time has passed for acoustic transverse polarization to build up. In the case of amplified spontaneous emission, by contrast, the decay speeds up as soon as a minute phonon population is present. The description in terms of rate equations did indeed prove incompatible with the "superfluorescence" data (dashed curve in the inset of Fig. 2), and, similarly, the pendulum equation is

inconsistent with the "amplified-spontaneous-emission" data. When a quantitative comparison of the two descriptions is made, however, minor discrepancies remain. More specifically, we find that ΔN_0 for superfluorescence in 1000-at.ppm ruby is larger than ΔN_0 for amplified spontaneous emission in the 2500-at.ppm specimen at the same laser power. The two descriptions we have relied on are limiting cases. Although the sharpness of the criterion for superfluorescence[10] provides some justification for this approach, it is unlikely that coherence is entirely absent in the case of amplified spontaneous emission. Furthermore, these limiting descriptions, by necessity, contain a number of simplifying approximations. In the description of superfluorescence, the excited zone is thought of as consisting of independently radiating rays, the emission in each ray is assumed to take place in a single mode, spontaneous decay is incorporated via an exponential shortening of the acoustic Bloch vector, and the effect of the probe pulse is ignored. In the rate equations used, the linewidth is assumed to be constant, and the transition is taken to be of square form. Furthermore, in both descriptions anisotropy and anharmonic phonon decay are ignored. All these assumptions, however, are not expected to detract from the main conclusions of this Letter, and their effects can indeed be accommodated in the parameters.

We acknowledge partial financial support by The Netherlands foundation "Fundamenteel Onderzoek der Materie (FOM)" and the "Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO)."

[1]M. S. Feld and J. C. MacGillevray, in *Nonlinear Optics,* edited by M. S. Feld and V. S. Letokhov (Springer-Verlag, Berlin, 1980), p. 7; J. C. MacGillevray and M. S. Feld, Phys. Rev. A **23**, 1334 (1981).

[2]C. Leonardi, J. C. MacGillevray, S. Liberman, and M. S. Feld, Phys. Rev. B **11**, 3298 (1975).

[3]W. J. Brya and P. E. Wagner, Phys. Rev. **157**, 400 (1967).

[4]W. E. Bron and W. Grill, Phys. Rev. Lett. **40**, 1459 (1978).

[5]P. Hu, Phys. Rev. Lett. **44**, 417 (1980).

[6]D. J. Sox, J. E. Rives, and R. S. Meltzer, Phys. Rev. B **25**, 5064 (1982).

[7]J. G. M. van Miltenburg, G. J. Jongerden, J. I. Dijkhuis, and H. W. de Wijn, in *Phonon Scattering in Condensed Matter,* edited by W. Eisenmenger, K. Lassmann, and S. Döttinger (Springer-Verlag, Berlin, 1984), p. 130.

[8]R. S. Meltzer and R. M. Macfarlane, Phys. Rev. B **32**, 1248 (1985).

[9]M. F. H. Schuurmans and D. Polder, Phys. Lett. **72A**, 306 (1979); M. F. H. Schuurmans, Opt. Commun. **34**, 185 (1980).

[10]M. S. Malcuit, J. Maki, D. J. Simkin, and R. W. Boyd, Phys. Rev. Lett. **59**, 1189 (1987).

[11]M. J. van Dort, M. H. F. Overwijk, J. I. Dijkhuis, and H. W. de Wijn, Solid State Commun. **72**, 237 (1989).

[12]M. H. F. Overwijk, C. R. de Kok, J. I. Dijkhuis, and H. W. de Wijn, J. Lumin. **45**, 440 (1990).

[13]F. T. Arecchi and E. Courtens, Phys. Rev. A **2**, 1730 (1970).

[14]J. Mostowski and B. Sobolewska, Phys. Rev. A **28**, 2943 (1983); **30**, 1392 (1984).

[15]No ground-state associated beat occurs in the $2\bar{A}(^2E)$-$^4A_2$ transition if the laser beam runs parallel to the c axis, as in Fig. 2.