Transition from Superfluorescence to Amplified Spontaneous Emission

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The cooperative emission process in $KCl:O_2^-$ has been studied as a function of the dephasing rate of the transition dipole. As the temperature of the sample is increased from 10 to 30 K, the emission evolves continuously from that characteristic of superfluorescence to that of amplified spontaneous emission. These results are in qualitative agreement with the predictions of current theories, but quantitative agreement is obtained only when current theories are modified so that the noise source that initiates the emission process is allowed to act continuously during the superfluorescent buildup.

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It is well known that cooperative effects involving many atoms can profoundly modify the nature of the spontaneous emission process.[1] Two such effects are superfluorescence,[2-10] that is, cooperative spontaneous emission involving many atoms, and amplified spontaneous emission (ASE),[11-13] in which the spontaneous emission from a single atom is amplified as it propagates through an inverted atomic medium. Extensive theoretical and experimental studies of these two processes have been performed. However, for the most part these processes have been studied independently, and relatively little work has been done in characterizing the nature of the emission process in the regime[14-17] intermediate between that of superfluorescence and that of ASE. In this Letter, we describe an experiment in which the nature of the cooperative emission process involving the super-oxide ion in potassium chloride ($KCl:O_2^-$) is studied as a function of temperature and, hence, as a function of the dephasing rate of the transition dipole. We selected $KCl:O_2^-$ for this experiment because it is the only solid-state system in which superfluorescence has been reported,[18] and because the dephasing rate of a dipole imbedded in a solid increases rapidly with temperature. We find that the nature of the emission evolves gradually from that of superfluorescence to that of ASE as the dipole dephasing rate is increased.

The simplest manifestation of superfluorescence occurs in the limiting case in which dephasing is negligible. All N excited atoms within the interaction volume participate, and the macroscopic dipole moment of the collection of atoms is approximately N times larger than the atomic dipole transition moment. As a result, the emission is highly directional and is emitted in the form of a pulse whose peak intensity scales as N^2 and whose characteristic duration is $\tau_R = 8\pi\tau_{sp}/3\rho\lambda^2 l$, where τ_{sp} is the single-atom spontaneous decay time, λ is the emission wavelength, ρ is the number density of atoms, and l is the length of the medium. The emitted pulse is delayed with respect to the exciting radiation by an interval[19] $\tau_D = \tau_R[\ln(2\pi N)^{1/2}]^2/4$, which is typically 10 to 100 times longer than τ_R. This delay represents the time required for the individual atomic dipoles to come into phase lock due to the coupling between them.

The presence of dephasing processes modifies this behavior by inhibiting the formation of the macroscopic dipole moment. If the dephasing rate is not too large, superfluorescence can still occur, but the delay time and the pulse duration are increased and the peak intensity is decreased.[4,16,17] These characteristics have been observed experimentally by Okada, Ikeda, and Matsuoka[14] and by Brechignac and Cahuzac.[15] For sufficiently large dephasing rates, no macroscopic dipole moment ever develops; each atomic dipole simply responds to the instantaneous value of the radiation field. If the number density of excited atoms is sufficiently large, this emission is still much more rapid than single-atom spontaneous emission, because the spontaneous emission from any one atom is amplified as the radiation propagates through the inverted medium. In this case, the emission is referred to as ASE. As a result of geometrical effects, the radiation is still highly directional, but there is essentially no time delay and the peak intensity no longer scales as N^2. Schuurmans and Polder[16] have calculated the value of the dephasing rate $\Gamma_2 = T_2^{-1}$ at which the transition between these two types of behavior occurs. They show that the emission is characteristic of superfluorescence if $T_2 \gg (\tau_R\tau_D)^{1/2}$, and is characteristic of ASE if $\tau_R \ll T_2 \ll (\tau_R\tau_D)^{1/2}$. Our experiment, which we describe below, verifies this prediction.

We have performed our experiment on a KCl crystal cleaved to dimensions of approximately $7\times7\times4$ mm^3 containing $\simeq 2\times10^{18}$ O_2^- molecular ions/cm^3. The crystal was mounted in a temperature-regulated cryostat cooled by a closed-cycle helium refrigerator. Since the dipole dephasing rate increases with the third power of the temperature,[20] this rate can be increased from its value[18] of 10^{10} s^{-1} at 10 K to $\simeq 3\times10^{11}$ s^{-1} by variation of the temperature between 10 and 30 K. The crystal was excited by a 30-ps pulse containing up to 60 μJ of energy from a frequency-quadrupled Nd-doped yttrium-aluminum-garnet laser. The pulse was focused into the crystal with a cylindrical lens to provide an interaction region approximating a cylinder of diameter 80 μm and a length of 7 mm and, hence, having a Fresnel num-

ber of unity. For excitation energies above $\simeq 10$ μJ, highly directional emission at the 6294-Å vibronic transition in O_2^- was emitted from both ends of the crystal. All of our experiments were performed using laser energies below the threshold for two-color superfluorescence.[18,21] The emission was detected by means of a streak-camera system capable of providing a temporal resolution better than 2 ps.

Figure 1 shows typical output pulses obtained under identical excitation conditions for several different values of the crystal temperature. The displayed pulse shapes have been scaled in such a manner that the peak intensities are equal, although actually the energy of each pulse is approximately the same. As the temperature is raised, the output pulse shape is seen to evolve continuously from that characteristic of superfluorescence to that characteristic of ASE. At the lowest temperature shown, the emission is superfluorescence with a pulse length of approximately 60 ps and a time delay of approximately 160 ps. As the temperature of the crystal is increased slightly [cases (b)–(d)], the emitted pulse broadens and the time delay increases. As the temperature is increased still further [case (e)], the pulse continues to broaden but the time delay begins to decrease. At the highest temperature shown, the emission is characteristic of ASE: The time delay is almost immeasurably small, the output pulse is very noisy, and the pulse duration has increased still further. This pulse duration is, however, considerably shorter than the 80-ns spontaneous emission lifetime of the transition. In all cases shown, the unsaturated line-center single-pass gain $g_0 l = T_2/\tau_R$ is much greater than unity.

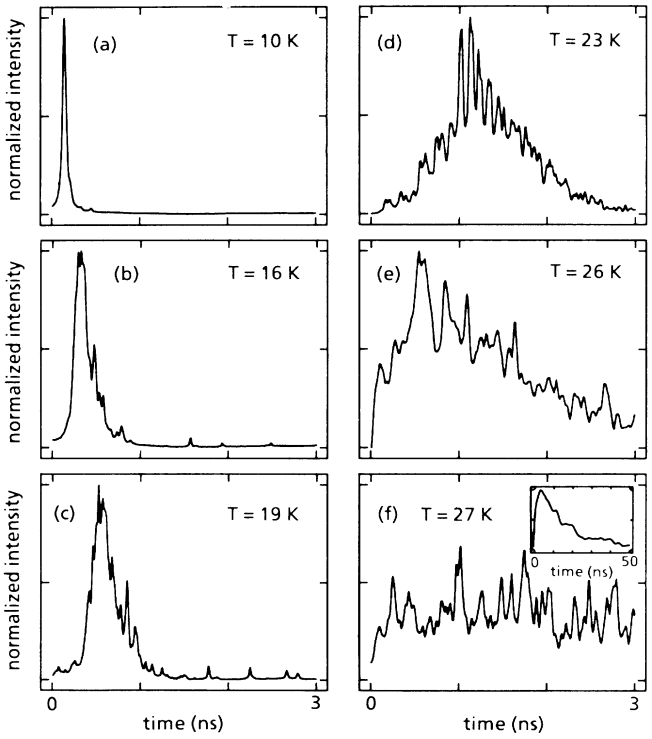

FIG. 1. Typical experimental realizations of the temporal evolution of the emission from $KCl:O_2^-$ for several different temperatures. At the lowest temperature, the emission is characteristic of superfluorescence, whereas at the highest temperature the emission is characteristic of amplified spontaneous emission. The inset to case (f) shows the evolution of the emission on a greatly enlarged time scale.

Figure 2 shows how the time delay and peak intensity vary with temperature. Because of the statistical nature of the emission process, many shots are collected at each temperature so that the mean values and variations of the time delay and the peak intensity could be determined. The circles give the mean values and the vertical bars indicate the range of values at each temperature. As the temperature of the crystal and, hence, the dephasing rate is increased, the mean time delay is seen to

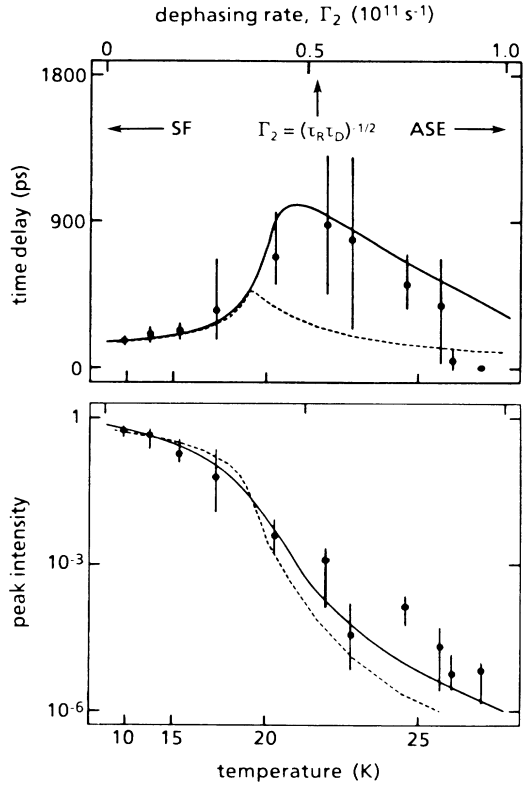

FIG. 2. Time delay and peak intensity of the emission plotted vs temperature and vs dephasing rate. Circles represent the mean values of the experimentally observed quantities, and the vertical lines represent the range of observed values. According to the criterion of Schuurmans and Polder, the emission is characteristic of superfluorescence (SF) for $\Gamma_2 \ll (\tau_R\tau_D)^{-1/2}$ and is characteristic of ASE for $\Gamma_2 \gg (\tau_R\tau_D)^{-1/2}$. The dashed curve represents the predictions of a theory that treats the effects of noise only as an initial condition, whereas the solid curve represents the predictions of a model that includes a temporally fluctuating noise source.

increase from $\simeq 160$ ps at our lowest temperature to a maximum of $\simeq 800$ ps and then to decrease monotonically as the temperature is increased still further. In addition, the fluctuations in the time delay are seen to increase in the transition region. From Fig. 2(b) we see that the peak intensity decreases monotonically over 6 orders of magnitude as the temperature is increased.

We have compared our results given in Fig. 2 with the predictions of current theories that treat the initiation of superfluorescence in the presence of dephasing.[4,16,17] We use the theory as formulated by Haake *et al.*[17] because this treatment leads to predictions that are readily compared to our experimental results. This theory assumes that the transition is inhomogeneously broadened, whereas in our experiment the transition is largely homogeneously broadened. It has been shown that the nature of the broadening mechanism does not affect the initiation of superfluorescence[4]; therefore, we assume that the theory of Haake *et al.* can be used to interpret our experimental results. The theory characterizes the emission in terms of the probability distribution of the first passage time,[22] which is defined to be the time at which the emitted intensity reaches some reference intensity. If this reference intensity is equal to the mean value of the peak intensity, the first passage time closely approximates the time delay of the emission. Haake *et al.* have obtained analytic expressions for the probability distributions of the first passage time in the limit in which the excited-state population can be assumed constant and under the assumption that the effects of quantum fluctuations enter only as an initial condition for the polarization. In the superfluorescence limit (i.e., the limit of little dephasing) these assumptions are well justified and lead to theoretical predictions[19,23] that are in excellent agreement with experimental results.[24] Haake *et al.* predict that for a Lorentzian line shape the time evolution of the mean intensity is given by

$$I(t)=\frac{4}{Nl}\int_0^l dz\,\theta\left(t-\frac{z}{c}\right)\left\{\exp\left(-\frac{2(t-z/c)}{T_2^*}\right)I_0\left(2\left[\frac{z}{l\tau_R}\left(t-\frac{z}{c}\right)\right]^{1/2}\right)^2 + \frac{2}{T_2^*}\int_0^{(t-z/c)} dt'\exp\left(-\frac{2t'}{T_2^*}\right)I_0\left(2\left(\frac{zt'}{l\tau_R}\right)^{1/2}\right)^2\right\}, \quad (1)$$

where $\theta(t)$ is the unit step function, $I_0(x)$ is the modified Bessel function of zeroth order, and T_2^* is the dipole dephasing time. In applying this theory to our experimental conditions, we take the reference intensity to be that intensity at which the upper-level population drops to half of its initial value, and we take the delay time to be the time at which the intensity first equals the reference intensity. We define the time delay in this manner because in the ASE limit the mean intensity reaches an asymptotic value well before half of the upper-state population is depleted. The dashed curves in Fig. 2 give the results of this theory assuming a Lorentzian line of width equal to the homogeneous width $\Gamma_2=5\times10^6T^3$ and using the values $N=3\times10^9$ and $\tau_R=2.7$ ps. It is seen that the predicted time delays agree with the experimental observations only when the dephasing rate is not too large; in the transition region the predicted time delay is considerably shorter than that observed experimentally.

In the transition regime many dephasing collisions occur during the buildup of the macroscopic dipole moment, and as a result the effects of noise *during* the emission process are important. The effects of noise, thus, cannot be described solely as an initial condition, as they are in most current quantum electrodynamic theories. However, it is not well understood how to include the effects of both noise and dephasing collisions into a quantum electrodynamic treatment of superfluorescence. In order to explain our experimental results, we have therefore developed a semiclassical theory to investigate the manner in which noise initiates the emission process in the transition regime. Semiclassical treatments[25] of superfluorescence in the limit of little dephasing are known to lead to accurate predictions for the time delay and peak intensity if the quantum noise is modeled as an initial condition, for example, by the assumption that each atomic dipole has an initial "tipping angle." In order to allow for the effects of noise during the initiation of the superfluorescence, our model assumes that a weak, continuous noise field is incident upon the inverted medium. We have integrated the on-resonance Maxwell-Bloch equations[4,5,7] for different input fields. When the input field is assumed constant in time, the predictions of the theory are essentially the same as those of the theory of Haake *et al.* shown by the dashed line in Fig. 2. However, the predictions are markedly different for the case of a randomly fluctuating input noise field. We model this field as a classical thermal noise field with a coherence time equal to twice the dephasing time of the transition.[26] The mean intensity is chosen such that in the limit of no dephasing the results of the simulation agree with the predictions of quantum electrodynamic theories. For this simulation many realizations are studied for each dephasing rate, and the mean value of the time delay and peak intensity are thereby obtained. The results of this simulation are shown by the solid curves in Fig. 2. These results are in excellent agreement with our experimental results, even in the intermediate regime. This agreement suggests that when collisions are sufficiently frequent, quantum noise cannot be modeled solely as an initial condition, because under these conditions the temporal evolution of the system is not determined solely by

these initial conditions.

In conclusion, we have studied cooperative emission in the regime intermediate between that of superfluorescence and ASE. The results are in good qualitative agreement with predictions of current theories, but quantitative agreement is obtained only when the current theories are extended to include a noise source that can act during the superfluorescent buildup.

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[1]R. H. Dicke, Phys. Rev. **93**, 99 (1954).

[2]N. E. Rehler and J. H. Eberly, Phys. Rev. A **3**, 1735 (1971).

[3]R. Bonifacio and L. A. Lugiato, Phys. Rev. A **11**, 1507 (1975), and **12**, 587 (1975).

[4]M. F. H. Schuurmans, Q. H. F. Vrehen, D. Polder, and H. M. Gibbs, in *Advances in Atomic and Molecular Physics,* edited by D. Bates and B. Pederson (Academic, New York, 1981), p. 167.

[5]M. S. Feld and J. C. MacGillivray, in *Coherent Nonlinear Optics,* edited by M. S. Feld and V. S. Letokhov (Springer-Verlag, Berlin, 1980), p. 7.

[6]M. Gross and S. Haroche, Phys. Rep. **93**, 301 (1982).

[7]Q. H. F. Vrehen and H. M. Gibbs, in *Dissipative Systems in Quantum Optics,* edited by R. Bonifacio (Springer-Verlag, Berlin, 1982), p. 111.

[8]H. M. Gibbs, Q. H. F. Vrehen, and H. M. J. Hikspoors, Phys. Rev. Lett. **39**, 547 (1977).

[9]F. P. Mattar, H. M. Gibbs, S. L. McCall, and M. S. Feld, Phys. Rev. Lett. **46**, 1123 (1977).

[10]D. J. Heinzen, J. E. Thomas, and M. S. Feld, Phys. Rev. Lett. **54**, 677 (1985).

[11]L. Allen and G. I. Peters, Phys. Rev. A **8**, 2031 (1973).

[12]U. Ganiel, A. Hardy, G. Neumann, and D. Treves, IEEE J. Quantum Electron. **11**, 881 (1975).

[13]S. R. Wilk, R. W. Boyd, and K. J. Teegarden, Opt. Commun. **47**, 404 (1983).

[14]J. Okada, K. Ikeda, and M. Matsuoka, Opt. Commun. **26**, 189 (1978), and **27**, 321 (1978).

[15]C. Brechignac and P. Cahuzac, J. Phys. B **14**, 221 (1981).

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

[17]F. Haake, J. Haus, H. King, G. Schroder, and R. Glauber, Phys. Rev. Lett. **45**, 558 (1980), and Phys. Rev. A **23**, 1322 (1981).

[18]R. Florian, L. O. Schwan, and D. Schmid, Solid State Commun. **42**, 55 (1982), and Phys. Rev. A **29**, 2709 (1984); L. O. Schwan, R. Florian, D. Schmid, E. Betz, W. Schrof, B. Walker, and H. Port, Phys. Status Solidi (b) **124**, 741 (1984).

[19]D. Polder, M. F. H. Schuurmans, and Q. H. F. Vrehen, Phys. Rev. A **19**, 1192 (1979).

[20]D. B. Fitchen, in *Physics of Color Centers,* edited by W. B. Fowler (Academic, New York, 1968), p. 293.

[21]F. Haake and R. Riebold, Phys. Rev. A **29**, 3208 (1984); P. Schwendimann, Opt. Acta **31**, 107 (1984).

[22]R. Roy, A. W. Yu, and S. Zhu, Phys. Rev. Lett. **55**, 2794 (1985); S. Zhu, A. W. Yu, and R. Roy, Phys. Rev. A **34**, 4333 (1986).

[23]R. Glauber and F. Haake, Phys. Lett. **68A**, 29 (1978); F. Haake, H. King, G. Schroder, J. Haus, R. Glauber, and F. Hopf, Phys. Rev. Lett. **42**, 1740 (1979); F. Haake, H. King, G. Schroder, J. Haus, and R. Glauber, Phys. Rev. A **20**, 2047 (1979).

[24]Q. H. F. Vrehen, in *Laser Spectroscopy IV,* edited by H. Walther and K. W. Rothe (Springer-Verlag, Berlin, 1979).

[25]J. C. MacGillivray and M. S. Feld, Phys. Rev. A **14**, 1169 (1976).

[26]R. Loudon, *Quantum Theory of Light* (Clarendon, Oxford, 1973), p. 81.