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Time-resolving experiments on Dicke superfluorescence of O_2^- centers in KCl. Two-color superfluorescence

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Single crystals of KCl containing O_2^- centers (8×10^{16}–7×10^{17} cm^{-3}) were excited at low temperatures ($T<30$ K) using frequency-quadrupled single pulses of a mode-locked Nd-doped yttrium aluminum garnet laser. At excitation intensities above 20 GW/cm^2 a spontaneous emission of coherent light was observed which could be identified as Dicke superfluorescence. This superfluorescence was observed on two of the zero-phonon lines in the vibrational progression of the O_2^- fluorescence spectrum (0-10 and 0-11 at 592.8 and 629.1 nm, respectively). We have studied the time dependence, the statistics of the delay times, and the intensities. The observed delay times vary between 0.5 and 10 ns, the observed pulse widths between 0.5 and 6 ns, and the coherence time was found to be longer than 100 ps. The observed linewidths of the superfluorescence (0.016 nm$\cong$14 GHz) is a factor of 2.5 smaller than that of the spontaneous fluorescence (0.042 nm at 4.2 K). The superfluorescence is observed only below a critical temperature which depends on the O_2^- concentration (8 to 28 K for concentrations between 8×10^{16} and 7×10^{17} cm^{-3}).

I. INTRODUCTION

In ensembles consisting of a large number of two-level systems which initially are all in the upper state there may exist a cooperative interaction which yields a collective spontaneous emission of radiation. Starting with the pioneering work of Dicke[1] this phenomenon has attracted intense theoretical interest.[2–10] In spite of these activities there is comparatively little experimental information on the properties of superfluorescence although it has been studied successfully in various laboratories.[11–19] Several reviews have been published describing both the theoretical and the experimental achievements in this field.[19–22]

In a recent note[23] we have reported preliminary results on the observation of superfluorescence of O_2^- centers in KCl. We have now completed a series of more extensive experiments. Concerning the interpretation of some of the results of these experiments, in particular in connection with the observed two-color superfluorescence (see Sec. IV C), a theoretical approach has already been presented by Haake and Reibold.[24] Another approach is currently about to be completed by Schwendimann.[25] In this paper we do not intend to invoke either of these models for an explanation of the observations, but only to summarize the observed experimental facts.

II. THE O_2^- CENTER

The O_2^- center in KCl was discovered in 1937 by Honrath.[26] Its structure[27,28] and its optical spectra[29,30] are well known. The absorption spectrum consists essentially of a broad band, peaked at about 250 nm (40 000 cm^{-1}). The O_2^- molecular ion substitutes a halide ion. At low temperatures in the ground state its molecular axis is oriented along a ⟨110⟩ axis.

The fluorescence spectrum (as shown in Fig. 1 for $T=4.2$ K) is dominated by a progression of narrow lines ($\Delta\bar{\nu}_{1/2}\cong1$ cm^{-1}) which results from transitions from the relaxed excited state to vibrationally excited states of the electronic ground state of the O_2^- molecular ion. In Fig. 1 some of the participating vibrational levels are indicated as well as the wavelength of the resulting zero-phonon lines. The spectrum presented in Fig. 1 is not corrected for the spectral sensitivity of the detection scheme. The highest oscillator strength of all the zero-phonon lines is observed for the 0-9 transition at (559.8±0.1) nm. (All wavelengths quoted in this paper are the wavelengths measured in air at standard conditions.) The wavelengths given in Fig. 1 are in good agreement with those quoted by Rolfe *et al.*[29]

III. EXPERIMENTAL

Most of the crystals used in this work were grown in air from melts containing approximately 5×10^{-3} mole KO_2

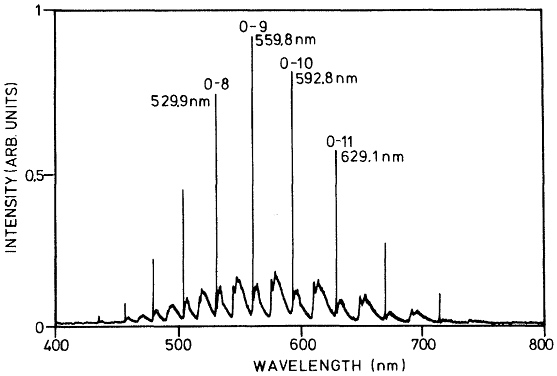

FIG. 1. Fluorescence spectrum of O_2^- centers in KCl at 4.2 K. Spectrum is not corrected for the spectral sensitivity. Ratio of the corrected oscillator strengths of the zero-phonon lines at 529.9, 559.8, 592.8, 629.1, and 669.6 nm is 0.88:1:0.86:0.61:0.39.

per mole KCl. When using this procedure the crystals were always contaminated by significant amounts of OH^-. However, by comparing the results obtained with these crystals to those obtained with samples grown in an inert-gas atmosphere it was found that the OH^- did not influence the O_2^- superfluorescence, unless the intense uv irradiation resulted in a formation of F centers. In this way O_2^- concentrations between 8×10^{16} and 7×10^{17} cm^{-3} were obtained. The O_2^- center concentration obtainable simply by growing the crystal in an atmosphere of oxygen was too low to yield the superfluorescence results reported in the following sections.

The crystals were cleaved to samples of about $5\times5\times10$ mm^3 and mounted in a variable-temperature cryostat (4—300 K, Leybold-Heraeus). The spectrum shown in Fig. 1 was obtained using the 253.7-nm line of a mercury low-pressure arc-discharge lamp for excitation. However, since the main emphasis of this work was placed on the time dependence of the fluorescence, in most of the experiments we used single pulses of the fourth harmonic of a mode-locked Nd:YAG laser (JK, model 2000) with the following specifications: $\lambda=266$ nm, pulse duration $\cong 30$ ps, maximum energy of a single pulse 100 μJ, and peak intensity at the sample surface $\cong 40$ GW/cm^2. The lower harmonics were removed from the exictation pulse using appropriate filters.

Figure 2 presents an extremely simplified block diagram of the experimental setup. The excitation pulses were focused to a spot of about 0.1-mm diameter at the sample surface using a 0.5-m-focal-length lens. The absorption coefficient of the samples at the excitation wavelength varied between 1 and 10 cm^{-1} which warranted a reasonably homogeneous penetration of the excitation light in the lightly doped samples, whereas in the heavily doped samples more than 99% of the excitation light was absorbed. In this way we obtained a thin pencil-shaped excitation volume in the sample.

The fluorescence light was monitored with a fast vacuum photodiode (Valvo, UVHC 20) or a microchannelplate photomultiplier (Hamamatsu, model R 1294 U-03) using suitable filters to protect the detector from the excitation light. The signals were registered using a 1-GHz oscilloscope (Tektronix, model 7104) and a digitizing camera (Thomson CSF, model TSN 1150-10) and recorded using a microcomputer (Kontron, PSI 80). The overall response time of the entire setup was about 500 ps. In order to improve the time resolution we determined the response function of the entire detection scheme using the second harmonic of the Nd:YAG laser as a reference signal. This response function could then be deconvoluted from the recorded signals yielding an estimated time resolution of 100 and 150 ps for detection with the photodiode and the photomultiplier, respectively. For the registration of the spectra we used a 0.6-m monochromator (Jobin-Yvon, model HRP) with a holographic grating (1800 lines per mm). For the determination of the exact position and the linewidth of the zero-phonon lines a 1.5-m monochromator was used (Jobin-Yvon, model THR) with a holographic grating (2400 lines per mm).

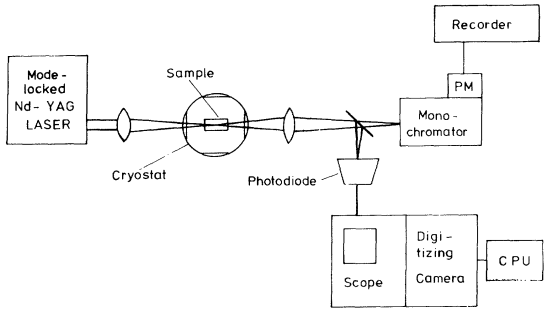

FIG. 2. Simplified block diagram of the experimental setup. See text for detailed description.

IV. EXPERIMENTAL RESULTS

A. Observation of superfluorescence

The top part of Fig. 3 illustrates the principle of the basic experiment for the observation of the superfluorescence: At excitation pulse energies below 20 μJ one observes the well-known isotropic spontaneous emission of the O_2^- centers with a decay time of 91 ± 2 ns at 4.2 K, which decreases to (50 ± 3) ns at 300 K. If the excitation is increased to pulse energies above a threshold of typically 30 μJ (>10 GW/cm^2), the emission of the zero-phonon line at 629.1 nm becomes highly anisotropic being collinear with the pencil-shaped active volume inside the sample. [Outside the sample there are deviations between the direction of the excitation pulse (266 nm) and the fluorescence light (629.1 nm) due to the refraction at the sample surface.] At the same time the fluorescence inten-

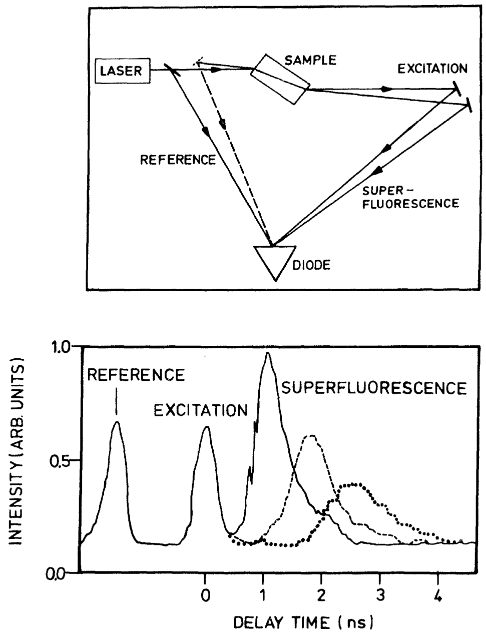

FIG. 3. Top: experimental setup for measuring intensities and delay times of the superfluorescence pulse. Bottom: experimental results for the forward superfluorescence in three independent shots at identical excitation conditions. The backward superfluorescence can be monitored using the arrangement indicated by the dashed line in the top part of the figure.

sities, both in the forward and the backward direction, increase by a factor of more than 10^4. The Fresnel number $F=S/\lambda L$ is close to unity for this radiation, S being the cross section of the active volume, L the length of the sample, and λ the wavelength.

The bottom part of Fig. 3 illustrates the time dependence of this radiation, which in the following will be identified as Dicke superfluorescence. A small fraction of the excitation pulse is reflected by a beam splitter to the photodiode to monitor the excitation pulse prior to the excitation. It is seen on the left-hand side of this figure (reference). The second pulse (excitation) originates from the fraction of the excitation pulse which penetrates through the sample and which is properly attenuated in front of the detector. It gives the time reference for the excitation event. The third pulse (superfluorescence) monitors the light emitted from the sample. It is recorded in Fig. 3 for three different shots at identical excitation conditions. The density of initially excited O_2^- centers was estimated to be in the order of 10^{17} cm^{-3}.

In spite of the identical excitation the fluorescence varies strongly from shot to shot. The pulse intensities fluctuate by more than a factor of 10, the pulse widths between 0.5 and 6 ns (full width at half maximum) and pulse delay times between 0.5 and 10 ns have been observed.

Such fluctuations of pulse intensities and pulse delay times are phenomena predicted for the superfluorescent process[7,8] since the initiation of each pulse is an intrinsically random process induced by the microscopic fluctuations of the initial dipole moments. The observation of these fluctuations therefore gives a first hint that the observed pulses are due to superfluorescence. It will be corroborated by the observations reported in the following sections. It should be mentioned that the occurrence of this radiation does not depend on the orientation of the sample. This rules out an accidental laser activity caused by unintended specular reflections.

An important quantity of interest is the coherence time of the superfluorescence pulses, since it gives information about fluctuations of the phase of the electromagnetic field. By sending the pulse through a Michelson interferometer with different lengths of the two arms it was possible to estimate the coherence time to be longer than 100 ps.

Coincident with the forward superfluorescence pulse recorded in Fig. 3 there always occurs a backward superfluorescence which can be monitored simultaneously using the arrangement sketched in dashed lines in the top part of Fig. 3. In spite of the large fluctuations in the results of different shots the forward and the backward pulse are always identical in pulse intensity, delay time, and pulse width within the limit of error (±10% for the intensity, ±100 ps for the delay time and pulse width).

Using an inteferometer arrangment it could be demonstrated that the wavelengths of the forward and the backward pulse were identical and that they were coherent with respect to each other.

B. Statistics of superfluorescence pulses

One of the most dominant features of the observed coherent emission is the fluctuation of both the pulse intensities and the delay times. In Fig. 4 we have collected the results of 300 individual experiments for which the excitation conditions were identical within less than ±10%. Both the intensities and the delay times vary by almost a factor of 10 and there is an obvious relation beween the pulse intensities and the delay times: For short delay times high pulse intensities (combined with short pulse widths) are more likely. In the top part of Fig. 4 the pulse intensities are plotted versus the accompanying delay times. In the histogram in the bottom part of Fig. 4 the number of pulses in a 100-ps time interval is plotted as a function of the delay time. The behavior illustrated by this figure is in qualitative agreement with the theoretical results obtained by Haake *et al.*[7,8] However, no quantitative comparison has yet been performed.

C. Two-color superfluorescence

In his original paper[1] Dicke has treated the coherent spontaneous emission of an ensemble of two-level systems. Clearly the O_2^- center is a much more complicated system. The observed fluorescence spectrum (Fig. 1) is readily explained in the conventional configurational-coordinate diagram[31] (Fig. 5): Following the optical excitation the system is in a nonequilibrium state and relaxes towards a new equilibrium position. The ensemble of the relaxed excited O_2^- centers is now in a totally inverted

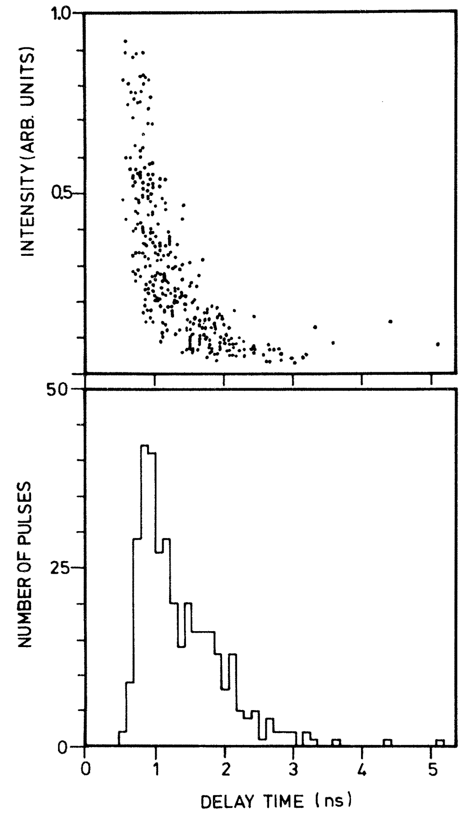

FIG. 4. Top: intensity of the superfluorescence pulse as a function of the delay time for 300 different shots at identical excitation conditions. Bottom: probability density for the observed delay times.

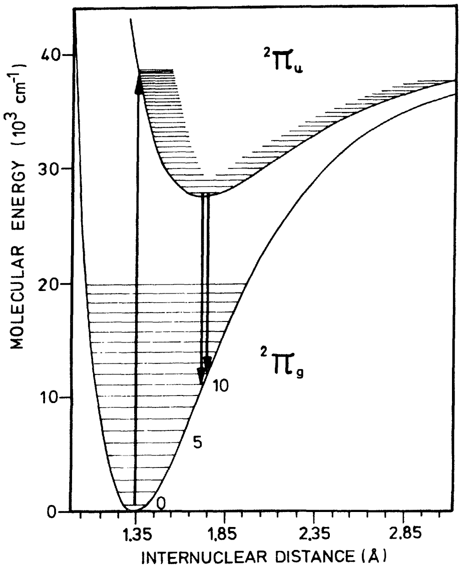

FIG. 5. Configurational-coordinate diagram for the O_2^- center in KCl. Superfluorescence was observed on the two transitions indicated in the figure (0-10 and 0-11).

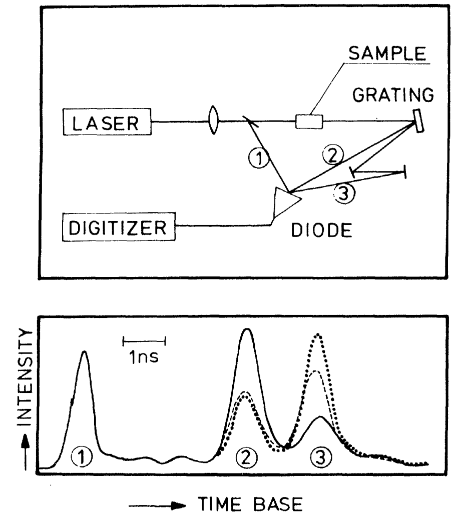

FIG. 6. Top: experimental setup to study the superfluorescence at 592.8 and at 629.1 nm, separately. 1, reference pulse; 2, yellow superfluorescence; 3, red superfluorescence. Bottom: experimental result for three independent shots at identical excitation conditions. Time difference between the two pulses on the right-hand side of the figure reflects the difference in the optical paths.

state, since the ground-state vibronic levels to which optical transitions can occur are unoccupied. However, as indicated by Fig. 1 there are several vibronic levels of almost equal importance. In Secs. IV A and IV B results were presented only for the 0-11 transition in the vibronic progression. This transition does not even yield the most intense line in the spontaneous fluorescence spectrum. The question now arises, whether it is possible to observe superfluorescence also on other transitions. The experimental answer to this question is yes: If the excitation density is further increased, we observe also superfluorescence on the 0-10 transition at 592.8 nm. The evidence for this statement is given by Fig. 6: If the excitation pulse energy is increased above a threshold of about 70 μJ the resulting superfluorescence pulse changes its color. The spectral content of the pulse can be analyzed using a reflection grating as indicated in the top part of Fig. 6. Above the threshold we observe a simultaneous emission of superfluorescence at 592.8 and at 629.1 nm.

In order to study the pulse intensities and delay times separately we have added an additional optical delay for the pulse at 629.1 nm. Thus the two pulses arrive at the photodiode at different times, even if they originate at the same time. The bottom part of Fig. 6 gives an example for the results. Pulse number 1 monitors the excitation pulse. It was identical within the limits of error for the three individual experiments recorded in this figure. Pulse number 2 is a recording of the superfluorescence pulse at 592.8 nm (yellow), pulse number 3 corresponds to the superfluorescence at 629.1 nm (red). In order to simplify the presentation for this figure we have selected three experiments for which the delay times of the red pulse were almost identical. The time difference between pulses 2 and 3 in Fig. 6 reflects exactly the difference in their optical path length. Thus we conclude that the red and the yellow pulse originate at the same instant within the limits of error (±100 ps).

Contrary to the simultaneous origin the relative pulse intensities fluctuate statistically with a qualitative trend for a preference for the yellow pulse at very short delay times, whereas in events with relatively long delay times the red pulse usually dominates. This statement is illustrated in Fig. 7 which gives the information analogous to that given in Fig. 4. We have tried to distinguish between the two cases by marking events with the yellow pulse dominating by small vertical bars, whereas events with the red pulse dominating are indicated as dots (top part of Fig. 7). Similarly, in the histogram in the bottom part of Fig. 7 we have distinguished between the two cases.

We do not intend to give an explanation for these observations. Indepedent attempts for a theoretical interpretation have already been proposed by Haake and Reibold[24] and by Schwendimann.[25]

D. Additional experimental observations

All the experiments reported so far in this paper have been done at 4.2 K. Superfluorescence is observed only below a critical temperature, which in turn depends on the concentration of the O_2^- centers. This is indicated in Fig. 8. The critical temperature at the highest available concentration of about 7×10^{17} cm^{-3} is about 27 K. It drops to about 8 K at a concentration of 8×10^{16} cm^{-3}. The O_2^- concentration was determined by measuring the optical absorption coefficient.[32]

In the same range of temperature both the wavelength and the width of the zero-phonon lines in the spontaneous emission spectrum depend slightly on the temperature. Figure 9 gives an example for the yellow line: Its wavelength shifts from (592.774±0.01) nm at 4.2 K to 592.84

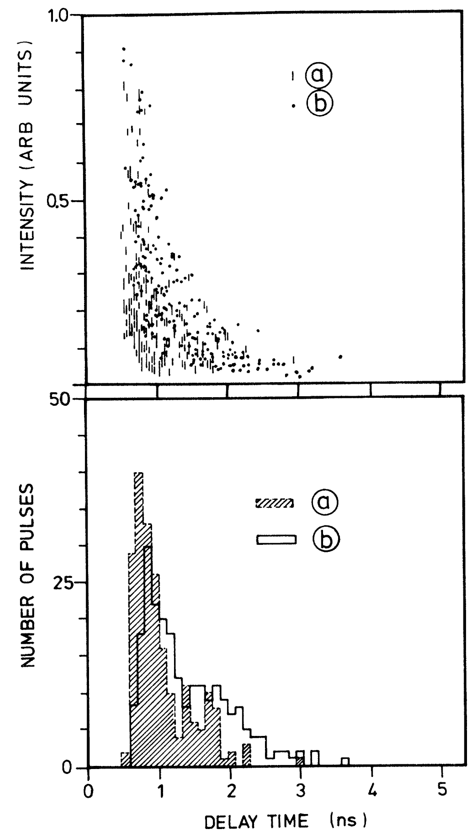

FIG. 7. Top: intensity of the superfluorescence pulses as a function of the delay time. Dots indicate pulses with the red contribution exceeding the yellow one (a), bars indicate experiments in which the yellow pulse dominated (b). Bottom: probability density for the observed delay times.

nm at 27 K, whereas its linewidth (full width at half maximum) increases in the same range of temperature from 1.2 to 4.5 cm^{-1}, respectively. Rebane and Rebane have quoted linewidths which qualitatively show the same temperature dependence, but are wider roughly by a factor of 2. We believe that in some cases the zero-phonon lines are inhomogeneously broadened, possibly due to internal strains. This assumption is based on the observation that in some samples we also find broader zero-phonon lines up to a factor of roughly 2.

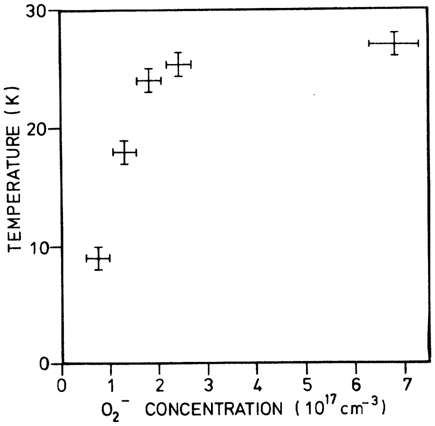

FIG. 8. Critical temperature as a function of the O_2^- concentration.

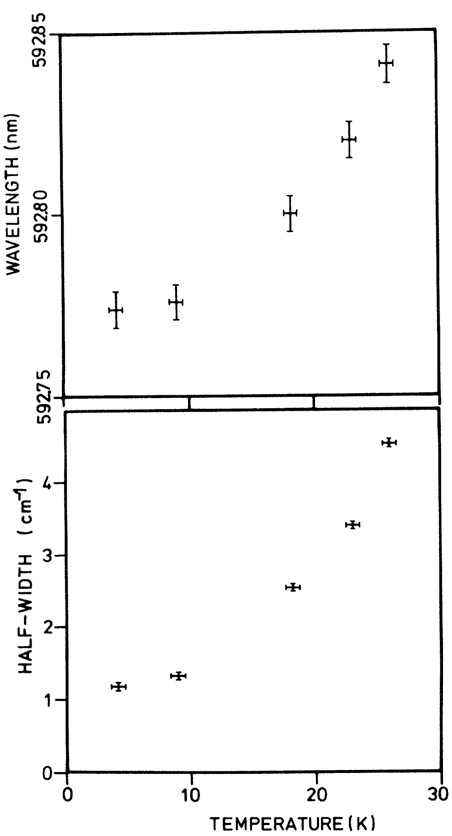

FIG. 9. Peak position of the 0-10 fluorescence line (top) and its half-width (bottom) in the spontaneous fluorescence spectrum as a function of temperature.

In Fig. 10 the spectral properties of the superfluorescence pulse at 4.2 K are compared to those of the spontaneous emission. Within the limits of error its maximum occurs at the same wavelength as that of the spontaneous emission, but its half-width shrinks to about 0.016 nm. On a frequency scale this width ($\cong$14 GHz) is at least in an order-of-magnitude agreement with the width expected for a lifetime-broadened line with the measured coherence time ($>$100 ps, see Sec. IV A) being the relevant time constant. In the latter case one expects a linewidth of about 3.2 GHz. Some of the additional broadening in the observed spectral linewidth may be due to the fact that it is the result of a large number of individual shots, where the unavoidable fluctuations in the pulse amplitude had to be corrected for. This indicates that the linewidth of the superfluorescence is at least close to the case of lifetime broadening. Experiments on the red superfluorescence yielded equivalent results.

Whereas in the experiments reported so far the active volume was always pencil-shaped with a Fresnel number close to unity, in a number of experiments the exciting pulse was focused using an additional cylindrical lens to yield an active volume in the form of a thin sheet. In this case a two-dimensional coherent emission was observed with the electric-field vector in the plane of the active

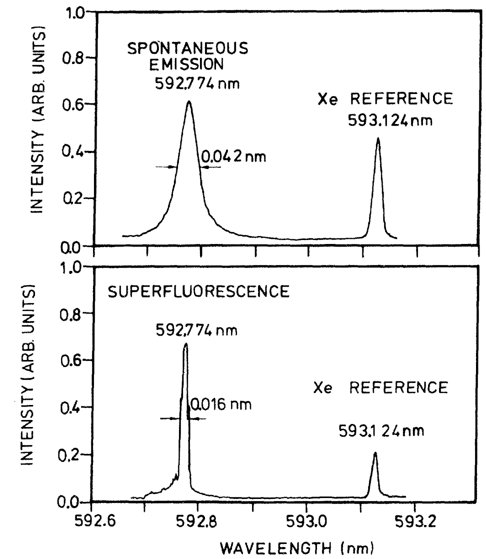

FIG. 10. Line shape of the 0-10 fluorescence line for spontaneous fluorescence (top) and superfluorescence (bottom). For calibration purpose a reference line from a Xe calibration lamp is also shown.

volume. (In the one-dimensional experiments the superfluorescence was linearly polarized with statistical directions of the electric-field vector.) Details about these experiments will be published in a forthcoming paper. A theoretical description of this case has been presented by Drummond and Eberly[33] and by Watson *et al.*[34]

Finally, we tried to get information about the relaxation time in the excited state by looking at the rise time of the spontaneous fluorescence. However, this rise time was too fast to be measurable with the available experimental equipment ($t_r < 150$ ps).

V. DISCUSSION

As stated in the Introduction, there are many different approaches to arrive at a complete theoretical understanding of the phenomenon of superfluorescence, in particular of the initiation process, since it yields a macroscopic tool to study quantum fluctuations.[2–10] It is not the purpose of this contribution to make a decision in favor of any particular of the proposed concepts, but it is hoped that the reported experimental results may help to test the validity of the assumptions in different models.

In view of the available experimental evidence in our opinion it is clear that the coherent radiation observed in this work is, in fact, due to a superfluorescence process. Particularly, this is supported by the following observations.

(1) The statistics of the pulse intensities and delay times.

(2) The coincidence, coherence, and intensity ratio of the forward and the backward pulse.

(3) The observed coherent two-color emission.

(4) The observed coherent two-dimensional emission.

A few comments on some of the observations follow.

(1) At first glance the disappearance of the superfluorescence in a very narrow range of temperature around the critical temperature is quite surprising, since the spectrum of the spontaneous fluorescence does not change significantly in this range of temperature. Electron spin resonance experiments,[27,28] however, have established that the O_2^- molecular ion begins to jump between the different low-temperature equilibrium orientations ($\langle 110 \rangle$ directions) in this range of temperature. Such a reorientation should, in turn, impede the formation of the collective coherent state responsible for the superfluorescence. It can, therefore, explain the occurrence of the critical temperature.

(2) The rise time of the spontaneous fluorescence is shorter than 150 ps indicating that the relaxation in the excited state takes place faster than this. On the other hand, the delay times observed for the formation of the superfluorescence were always longer than 500 ps. This can be understood by recognizing the fact that in the course of the relaxation of an O_2^- center more than 1 eV of energy has to be transferred to the lattice-phonon bath. This may cause a local heating and therefore prevent the formation of a superfluorescent state until the local temperature decreases again below the critical temperature.

(3) In his original paper[1] Dicke introduced the concept of superfluorescence for an ensemble of two-level systems. Whereas in thermal equilibrium the off-diagonal elements of the density matrix vanish, the superfluorescent state is characterized by large nonvanishing off-diagonal elements of the density matrix. Crudely speaking this means that both the upper and the lower level of the systems of the ensemble must be occupied partially with a definite phase relation.

The situation for the O_2^- centers is somewhat more complex: Following the excitation the centers first relax into the state which is the upper state involved in the radiation process. It is this relaxation process which guarantees that there are no unwanted phase relations in the initial state, since any phase relations, which might have been created accidentally when preparing the ensemble in the excited state with the laser pulse, will certainly be destroyed during the relaxation.

The lower state in turn is not simply the ground state but one of the vibrationally excited states (see Fig. 5) from where the centers again return to the ground state. For the ensemble to be able to form a superfluorescent state this relaxation rate must not be too fast, because otherwise the off-diagonal elements of the density matrix decay too fast, since the lower-state population is too small. It is straightforward to write down the rate equations using the Pauli operators for the ensemble[1] and to include a term taking account of the lower-state relaxation. We have solved these rate equations numerically for various initial conditions. As expected the solutions yield a superfluorescence event only if the relaxation process is not too fast. From the experimentally available initial concentration of excited-state centers ($\sim 10^{17}$ cm^{-3}) we can estimate a lower bound for the relaxation time: The lifetime of the 10th and the 11th vibrationally excited level must be longer than about 50 ps, since otherwise the solutions of the rate equations do not yield superfluorescence.

Of course, this procedure yields only a very crude estimate for the relaxation time, but more sophisticated calculations performed by Haake and Reibold[35] and by Schwendimann[36] yield relaxation times of the same order of magnitude. Since these calculations will be published soon, no details of the rough estimate described above are presented here.

VI. CONCLUDING REMARKS

A number of additional experiments have been performed on the subject. Among them are the following.

(1) A study of the polarization of the superfluorescence, which may give the information about the reorientation of the centers.

(2) Experiments on the behavior upon repeated pumping using a pulse train from a mode-locked Nd:YAG laser. These experiments should give information about the relaxation process.

(3) Experiments on "triggered superfluorescence," where a small fraction of the original superfluorescence pulse is coupled back into the sample to stimulate another superfluorescent pulse. Again we expect information about the relaxation and the reorientation processes.

Since these experiments are not yet quite completed and require detailed explanation they will be presented in a subsequent publication.

In this work superfluorescence was observed only on the 0-10 and the 0-11 transitions at 592.8 and 629.1 nm, respectively. We have not been able to observe it on any other transition, e.g., the 0-9 or the 0-12 transition at 559.8 and 669.4 nm (see Fig. 1). We believe that for the 0-12 transition the oscillator strength is too low, whereas for the short-wavelength transitions we have indications that absorptions to higher excited states take place and lower the obtainable gain below the threshold. Experiments concerning this matter are currently under way.

A final remark should be make concerning the reproducibility of the experiments. Due to the presence of OH^- impurities the intense uv radiation creates F centers which also absorb in the wavelength range of interest. However, this formation of F centers is slow enough to allow roughly 1000 experiments before moving the laser beam to a different spot at the crystal surface. We believe that this number can be increased by strengthening the effort to avoid the OH^- impurities.

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