Superradiance in a diphenyl crystal containing pyrene

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Dicke superradiance has been observed for the first time in an impurity-containing molecular crystal (diphenyl containing pyrene molecules) as a result of the $^1B_{2u} \to {}^1A_g$ transition in pyrene (3739.3 Å). The concentration dependence was investigated and found to be quadratic. The emission is highly directional as compared with the noncoherent spontaneous background and with the superluminescence process that occurs as a result of transitions from the $^1B_{2u}$ state to the vibrational sublevels of the lower state. The propagation of the superradiant emission was investigated in specimens of different length, and it was found that it took place under the conditions of self-induced transparency. The evolution of the superradiant pulse in a solid matrix was also studied. A theoretical interpretation of the experimental data is given.

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§1. INTRODUCTION

Cooperative emission processes and especially Dicke superradiance, i.e., coherent spontaneous emission,[1] have been attracting considerable attention. However, publications reflecting this scientific and practical interest have been largely confined to different questions in super-radiance theory (see, for example, published reviews[2,3] and monographs[4,5]). A very small number of experiments on superradiance have been carried out in gaseous media,[6–10] in which the model conditions on which the theory is founded are more readily satisfied and one can avoid the use of cryogenic techniques that are essential when such experiments are performed with crystals. On the other hand, very dense packing of active particles can be achieved in activated and impurity-containing crystals, and this facilitates self-induced correlations between them. Finally, the definite disposition of active particles in a solid matrix should lead to specific superradiant features connected with polarization and propagation properties. All this means that experiments on superradiance in crystals are exceedingly topical, and we now report our experiments in this field.

The working specimen used in our experiments was an impurity-containing molecular crystal (diphenyl containing pyrene molecules) whose spectroscopic and relaxation parameters and properties have been investigated in the literature.[11–13] In this paper, we examine the spatial, density, polarization, and spectroscopic properties of optical superradiance in this crystal.

§2. EXPERIMENTAL DATA ON OPTICAL SUPERRADIANCE IN A DIPHENYL CRYSTAL CONTAINING PYRENE

The diphenyl crystal belongs to the monoclinic space group C_{2h}^5 with two molecules at inversion sites in each unit cell. At liquid-helium temperatures, the pyrene molecule in the diphenyl crystal can have that of three stabilized orientations which ensures that it is close to a real vacancy.

Figure 1 shows the level scheme of a pyrene molecule in diphenyl. The molecule was excited nonresonantly by the third harmonic of an $Y_3Al_5O_{12}$:Nd laser producing a 10-ns pulse via the 1→3 transition. Because of the fast 3→2 radiationless transition, the pyrene molecules are in state 2, from which they undergo transitions both to state 1 (0-0 transition) and to state 4 that corresponds to the 1408 cm^{-1} fully-symmetric, intramolecular vibration. Absorption via the 0-0 transition is possible only when the exciting light is polarized along the *b* axis of the crystal (Fig. 1), whereas the emitted luminescence has the same intensity and polarization along the *b* and *c'* axes. We note that, as the intensity of the pump (1→3) increases, the emission spectrum of the pyrene molecules at 4.2 K is found to vary from a weakly polarized line emission (corresponding to spontaneous emission[11]) to a final spectrum containing only two lines. These lines correspond to the 0-0 resonance transition (2→1) with $\nu_{21} = 26\,734$ cm^{-1} and the 2→4 vibronic transition with $\nu_{24} = 25\,326$ cm^{-1}. Above certain particular threshold excitation intensities ($W_{13} = 5 \times 10^5$ W/cm² for the 2→1 emission and $W_{13} = 10^6$ W/cm² for the 2→4 emission), there was a sharp reduction in the transition time for both cases, i.e., from 110 ns (Ref. 14) to 5–6 ns for the 2→1 transition and to

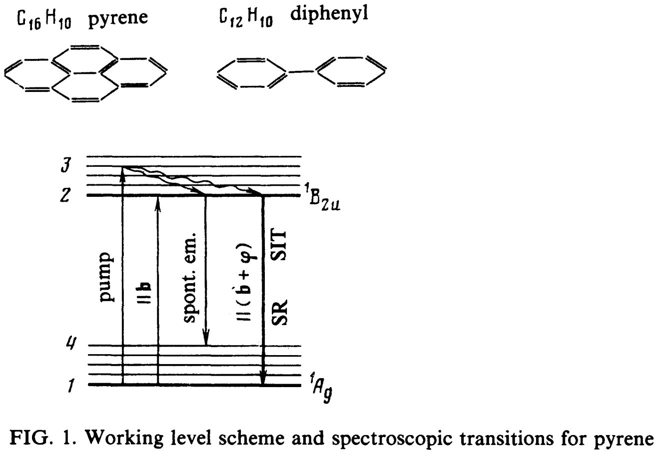

FIG. 1. Working level scheme and spectroscopic transitions for pyrene molecules in the diphenyl matrix.

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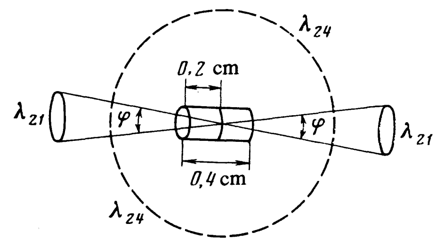

FIG. 2. Geometry of 2→1 and 2→4 emission by pyrene: λ_{21}—superradiance, λ_{24}—superluminescence, $\varphi = 0.1$ sr.

10–12 ns for the 2→4 transition. We note that the evolution of the emission pulses has been studied[13] with the fast ÉLU-FT photomultiplier and the I2-7 time interval meter; the pulse lengths were measured with the FÉK-22 coaxial photocell.

Experiments show that the 2→1 emission is highly directional (solid angle 0.1 sr, Fig. 2) and its intensity exceeds the intensity of noncollective spontaneous emission by a factor of 1000. The emission resulting from the vibronic 2→4 transition was found to be uniformly distributed within the entire solid angle, and its intensity exceeded the intensity of spontaneous emission by a factor of only 30–50. It was proposed that the 2→1 emission could be identified with superradiance, whereas the 2→4 transition produced superluminescence, since the vibrational state 4 was short-lived ($\tau_{41} = 10^{-11}$ s).

According to Dicke,[1] superradiance (SR) is a highly directional coherent emission of light by an ensemble of particles that have been excited to a collective (Dicke) state. The intensity of this emission is proportional to the square of the number of particles. This phenomenon is connected with self-induced correlation between emissions of the individual particles making up the ensemble, where the self-induced correlation time τ_c must be less than the times of longitudinal (T_1) and transverse irreversible (T_2) relaxation. Initially, none of the molecules in the inverted system has an electric dipole moment in state 2 (if it is nondegenerate). Spontaneous transition to state 1 of any of the molecules in the system through the radiation field that is common to all the molecules will lead to the transition of the remaining molecules from state 2 to the superposition state $\psi_c = a_1\psi_1 + a_2\psi_2$ where ψ_i is the wave function describing the i-th state and $|a_1|^2$ is the probability of finding the particle in the i-th state ($i = 1,2$). It is readily verified that, in the superposition state, the molecules do have an electric dipole moment (because the electric dipole moment of the 1→2 transition is not equal to zero). This produces an acceleration of the spontaneous emission process, so that the admixture of the ψ_1 state grows at an increasing rate which, in turn, eventually results in a burst of radiation whose intensity is proportional to the square of the number of active particles.

In view of this, we have carried out an experimental study of the density dependence of the 2→1 emission intensity. It is shown in Fig. 3, where N is the number of active pyrene molecules participating in the 2→1 emission process. The dependence is seen to be quadratic in character. On the other hand, the 2→4 emission intensity, which we have also investigated, was not found to be a quadratic function.

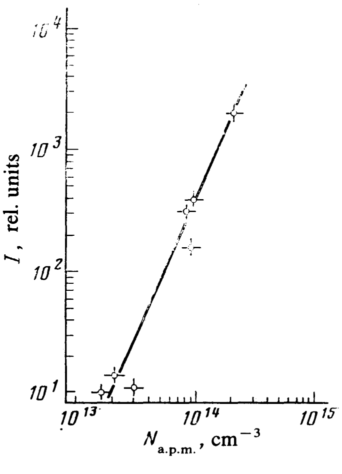

FIG. 3. Density dependence of the 2→1 emission intensity.

In our experiment, we investigated the polarization properties of 2→1 and 2→4 emission, and found that a low degree of polarization persisted for the 2→4 emission ($P = 0.4$), whereas the 2→1 emission was almost entirely linearly polarized ($P = 0.95$) under the conditions corresponding to a sharp threshold-type reduction in the transition time. Moreover, we found that the plane of polarization of the 2→1 emission was at an angle $\varphi \approx 100$–$110°$ to the b axis of the crystal. We found only two variants of the mutual disposition of the crystallographic axes of the diphenyl crystal and of the plane of polarization of the exciting radiation (Fig. 4) for which there was a sharp reduction in the transition time at the resonance frequency and the 2→1 emission

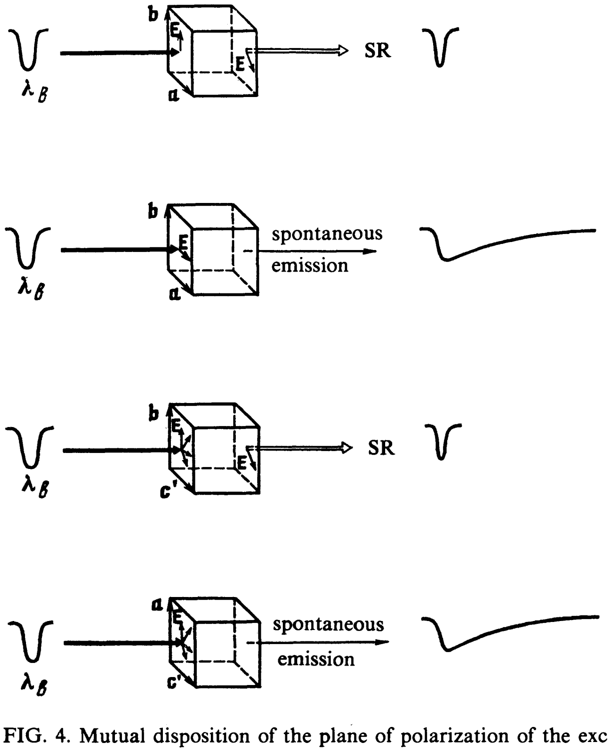

FIG. 4. Mutual disposition of the plane of polarization of the exciting wave and of the axes of the diphenyl crystal containing pyrene.

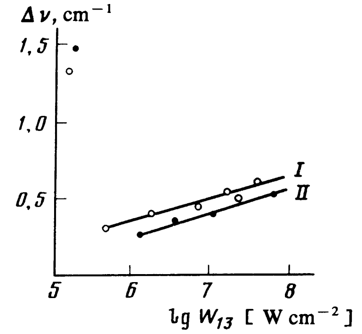

FIG. 5. The 2→1 (I) and 2→4 (II) emission linewidths as functions of the exciting intensity.

was highly directional. In the first of these variants, the excitation was at right-angles to the *ab* plane of the crystal and, in the second, at right-angles to the *bc'* plane.

We also investigated the width of these spectral lines, and found that it increased with increasing post-threshold pump flux density W_{13} (Fig. 5), in accordance with superradiance theory.

Thus, the sharp reduction in the 2→1 transition time with the large increase in its intensity, the highly directional nature of the emission, the well-defined polarization, the broadening of the spectrum, and the quadratic concentration dependence of the intensity enable us to consider that the 2→1 signal was, in fact, the Dicke superradiance.

§3. THEORETICAL ANALYSIS OF EXPERIMENTAL RESULTS

The evolution and subsequent propagation of the superradiant pulse in an extended medium can be interpreted in terms of the four-level system shown in Fig. 1. We note that superradiance and self-induced transparency in multilevel systems have been examined theoretically in Refs. 15 and 16.

Let us begin by considering the conditions necessary for producing inverted population of level 2 in this type of system. The kinetic equations for the level populations N_n (n is the level number) have the following form:

$$\frac{dN_n}{dt}=\sum_{n'}[P_{n'n}N_{n'}-P_{nn'}N_n]+\tilde{\omega}_{nn'}(N_{n'}-N_n), \tag{1}$$

where $P_{nn'}$ is the probability of a transition from level $|n'\rangle$ to $|n\rangle$,

$$\left.\begin{aligned}&\tilde{\omega}_{nn'}=\hbar^{-2}\mathscr{P}^2_{nn'}g(\omega),\\&\mathscr{P}_{nn'}=-\mathbf{d}_{nn'}\mathbf{E}_0\exp\left(-\frac{\alpha_{nn'}z}{2}\right),\end{aligned}\right\}$$

$g(\omega)$ is the normalized line-shape function, $d_{nn'}$ is the electric dipole moment of the nn' transition, E_0 is the electric field in the pump pulse, and $\alpha_{nn'}$ is the resonance absorption coefficient for the $|n'\rangle\to|n\rangle$ transition.

Since the length of the exciting laser pulse produced by the $|1\rangle\to|3\rangle$ transition ($\Delta t_{13}=10$ ns) is much greater than the irreversible relaxation times between the vibrational sublevels ($\tau_{32}\approx\tau_{41}\approx10^{-11}$ s), the excitation process may be regarded as quasistationary. The solution of (1) then yields the following ratio of populations of levels $|2\rangle$ and $|1\rangle\Delta N_{21}$ and of levels $|2\rangle$ and $|4\rangle\Delta N_{24}$ to the total number of active particles N:

$$n_{21}=\frac{\Delta N_{21}}{N}$$

$$=\frac{\tilde{\omega}_{31}\tau_{21}\tau_{24}-(\tau_{24}+\tau_{21})(\tau_{32}/\tau_{31}+\tilde{\omega}_{31}\tau_{32}+1+\tau_{32}/\tau_{34})}{\tilde{\omega}_{31}\tau_{21}\tau_{24}+(\tau_{24}+\tau_{21})(2\tilde{\omega}_{31}\tau_{32}+\tau_{32}/\tau_{31}+1+\tau_{32}/\tau_{34})+\tilde{\omega}_{31}\tau_{41}\left[\frac{\tau_{32}}{\tau_{34}}(\tau_{24}+\tau_{21})+\tau_{21}\right]},$$

$$n_{24}=\frac{\Delta N_{24}}{N}=\frac{N_2}{N}\,\frac{\tau_{34}\tau_{21}\tau_{24}-[\tau_{32}(\tau_{24}+\tau_{21})+\tau_{34}\tau_{21}]\tau_{41}}{\tau_{34}\tau_{21}\tau_{24}}. \tag{2}$$

Since $\tau_{21}\approx10^{-7}$ s, $\tau_{31}\approx10^{-8}$ s, $\tau_{34}\approx10^{-8}$ s, $\tau_{24}\approx10^{-7}$ s, and $\tau_{41}\approx10^{-11}$ s, we have

$$n_{21}\approx-\text{th}\left(\frac{\alpha_{13}z+\gamma}{2}\right), \tag{3}$$

$$n_{24}\approx\frac{\tilde{A}e^{-\alpha_{13}z}(1-B)}{\tilde{A}e^{-\alpha_{13}z}+C}, \tag{4}$$

where

$$\begin{gathered}\gamma=-\ln A,\qquad A=\frac{\hbar^{-2}d_{13}{}^2E_0{}^2\tau_{23}\tau_{21}\tau_{24}}{4\pi(\tau_{24}+\tau_{21})},\\ \tilde{A}=\frac{1}{4\pi\hbar^2}d_{13}{}^2E_0{}^2\tau_{23}\tau_{31},\qquad B=\tau_{41}/\tau_{24},\\ C=\frac{\tau_{31}}{\tau_{21}}+\frac{\tau_{31}}{\tau_{24}},\end{gathered} \tag{5}$$

and z is the coordinate of the leading edge of the pulse in the direction of propagation.

Numerical estimates performed for the threshold pump intensity $W_{13}=5\times10^5$ $W\cdot$cm^{-2} show that, when the pyrene density is $N\simeq3\times10^{16}-8\times10^{16}$ cm^{-3} and the measured absorption coefficient is $\alpha_{13}=10$–15 cm^{-1}, the conditions for inverted population difference, i.e., $n_{21}>0$, $n_{14}>0$, are satisfied when the effective length of the specimen is 0.1–0.2 cm. Since the length of the specimen used in our experiment was 0.4 cm, the propagation in the specimen of the 2→1 and 2→4 signals must be described by the coupled set of Maxwell equations and Bloch-type kinetic equations. Proceeding as in Ref. 13, and using (3) and (4), it can be shown that the pulse areas

$$\Theta_{ij}=\frac{2d_{ij}}{\hbar}\int_{-\infty}^{\infty}E(z,t')\,dt';$$

for the 2→1 and 2→4 transitions are given by

$$\Theta_{21}(z)=2\,\text{arctg}\left\{\text{tg}\left(\frac{\Theta_{0(21)}}{2}\right)\left(\text{ch}\frac{\gamma}{2}\right)^{\beta}\left[\text{ch}\left(\frac{\alpha_{13}z+\gamma}{2}\right)\right]^{-\beta}\right\}, \tag{6}$$

$$\Theta_{24}(z)=2\,\text{arctg}\left\{\left[\text{tg}\left(\frac{\Theta_{0(24)}}{2}\right)\right]\left(\frac{\tilde{A}+C}{C}\right)^{-\sigma}\left(\frac{Ce^{\alpha_{13}z}}{\tilde{A}+Ce^{\alpha_{13}z}}\right)^{-\sigma}\right\}, \tag{7}$$

$$\beta=\frac{\alpha_{12}}{\alpha_{13}},\qquad\sigma=\frac{\alpha_{24}}{2\alpha_{13}}(1-B),$$

where $\Theta_0(ij)$ is the initial pulse area for $z=0$ is the case of the $|i\rangle\to|j\rangle$ transition.

It follows from (6) that the propagation of the superradiant pulse in the case of the 2→1 transition is accompanied by selfinduced transparency (SIT), in accordance with Ref. 13, where the superradiant pulse passes through a partially inverted medium under these conditions. On the other hand, it follows from (7) that, when $C>\tilde{A}$ and z is large enough, so that $Ce^{\alpha}_{13}{}^{z}/(\tilde{A}+Ce^{\alpha}_{13}{}^{z})\sim 1$, we have $\Theta \lesssim \Theta_0$, i.e., we have the analog of free transmission by the medium under nonresonance saturation in the case of the 2→4 transition.

Even this analysis is sufficient to make clear the difference between the character of the 2→1 and 2→4 emissions. The hierarchy of relaxation times between different sublevels of the four-level system with a particular initial level population difference has ensured that the nonlinear coherent interaction processes (superradiance and self-induced transparency) turn out to be possible only for the 2→1 transition.

It follows from (3) and (4) that $n_{21}\to -1$ and $n_{24}\to 0$ as z increases. In this connection, it is useful to recall here that MacGillivray and Feld[17] predicted "ideal" superradiance in the case of a specimen with persisting inversion (as in our experiment).

One of the conditions for the detection of superradiance is

$$L/c<\tau_c<T_1,\ T_2, \tag{8}$$

where L is the length of the specimen, c is the velocity of light in the medium, τ_c is the self-induced correlation time, given by

$$\tau_c=8\pi\tau_1/3N\lambda_{21}{}^2L,\ \tau_1\approx 3\hbar c^3/4\omega_{21}{}^3d_{12}{}^2, \tag{9}$$

and λ_{21} and ω_{21} are, respectively, the wavelength and frequency of the 2→1 emission.

Since, in our experiment, $L=0.4$ cm, $N=10^{16}-10^{17}$ cm^{-3}, $d_{12}\simeq 0.8\times 10^{-20}$ esu, and $T_2^*\simeq 10^{-10}$ s, we have $\tau_c=0.23\times 10^{-9}-2.3\times 10^{-9}$ s. The time taken by a photon to traverse the medium, $L/c=0.3\times 10^{-11}$ s, is much less than τ_c which, in turn, is much less than the relaxation times $T_2=8$ ns and $T_1=10^{-7}$ s. Thus, the inequality (8) was satisfied in our experiment.

We have already noted that the conditions for inverted population difference can be realized in a specimen 0.1–0.2 cm thick. It is therefore logical to suppose that superradiance will also be produced in a layer of comparable thickness but, elsewhere in the specimen, it will propagate through a partially inverted medium and experience the reaction of the resonance medium. This enables us to estimate the ratio of the output flux density to the pump flux density ($W_{13}=10^{25}-4.8\times 10^{25}$ photons/cm$^2\cdot$s):

$$W_{13}/W_{21}\approx(0.6-2.87)\cdot 10^3,$$

which agrees with the experimental data (see also Ref. 13). The estimated energy of the 2→1 output pulse is 10^{-5} J, which is in agreement with experiment.

It is known that enhanced spontaneous luminescence is also possible in the inverted system, and the following inequality is satisfied[2,18]:

$$T_2<\tau_{\rm sl}=(2\pi Nd_{12}{}^2\omega_{21}/\hbar T_2{}^{-1})^{-1}\ll L/c, \tag{10}$$

which signifies that, in this case, the photons remain in the resonant medium for the time L/c, which is sufficient for the single-pass amplification of spontaneous luminescence that occurs at time $\tau_{\rm sl}$. When $N\simeq 10^{16}$ cm^{-3}, $T_2\simeq 8\times 10^{-9}$ s, $d_{12}\simeq 0.8\times 10^{-20}$ esu, and $\tau_{\rm sl}\simeq 3.97\times 10^{-12}$ s, we have $\tau_{\rm sl}<L/c\simeq 10^{-11}$ s. However, the left-hand half of the inequality given by (10) is not satisfied because the irreversible transverse relaxation time T_2 can be shorter than the photon transit time in the resonatorless system. On the other hand, analogous estimates for the 2→4 transition show that superluminescence will occur.

According to Ref. 17, the length of the superradiant pulse leaving the extended resonant medium can be estimated from the expression

$$\Delta t_{21}\approx\tau_c\Phi, \tag{11}$$

where

$$\Phi=\ln\frac{2\pi}{R},\quad R=\left[\frac{4}{N}\ln(2\pi N)^{1/8}\right]^{1/2},$$

and the delay of the superradiant pulse in the resonant medium can be calculated from the formula

$$\tau_3\approx\tau_c\Phi^2/4. \tag{12}$$

In our experiment, when $N\simeq 10^{16}$ cm^{-3}, $\Phi=13.52$, $R=8.46\times 10^{-6}$, and $l_{\rm SR}=0.1$–0.2 cm, we have $\Delta t_{21}=2.16\times 10^{-9}$ s and $\tau_3=7.3\times 10^{-9}$ s, which is in satisfactory agreement with experimental data ($\Delta t_{21}\simeq 5$–6 ns, $\tau_3\approx 9$ ns).

We note that the existence of 2→1 superradiance is also confirmed by the measured line halfwidth $\Delta\nu_{21}$ as a function of the pump intensity W_{13} (Fig. 5). It follows from (9) that the effective working length of the superradiant specimen increases with increasing post-threshold pump intensity. This produces a reduction in τ_c and a broadening of the spectrum of the output pulse. Figure 5 shows that, when W_{13} is reduced from $10^{5,7}$ W/cm^2 to $10^{6,3}$ W/cm^2, the linewidth changes by 0.05 cm^{-1}. If we estimate the width of the superradiance spectrum, we find that $\Delta\omega\simeq 7.5\times 10^8-29.9\times 10^8$ s^{-1}, i.e., the calculated change in the width $\Delta\omega_1-\Delta\omega_2\simeq 22.36\times 10^8$ s^{-1} is in agreement with the experimental result $\Delta\omega_1-\Delta\omega_2\simeq 15\times 10^8$ s^{-1}. On the other hand, in the case of amplified spontaneous luminescence, we have the following expression for the spectrum width[19]:

$$\Delta\omega_{\rm sl}\approx\Delta\omega/(W_{\rm th}\,t)^{1/2}, \tag{13}$$

where $W_{\rm th}$ is the intensity at the threshold for resonator-free generation and t is the generation time. Hence, it follows that, in the case of superluminescence, the spectrum should become narrower as the pump intensity increases.

We draw attention to the fact that the time Δt_i for inversion to be reached must satisfy the inequality[17]

$$\Delta t_i<\Phi^2T_2{}^*/4(1-f)\alpha_{12}l_{\rm SR}, \tag{14}$$

where f lies in the range $\frac{1}{2}\leqslant f<1$. This is actually equivalent to a restriction on the length $l_{\rm SR}$ of the superradiant part of the specimen:

$$l_{\rm SR}<\frac{\Phi^2(T_2{}^*/\Delta t_i)}{4\alpha_{12}(1-f)}. \tag{15}$$

In our experiment, $\alpha_{12} = 6\ \text{cm}^{-1}$ and $t_i \simeq 8 \times 10^{-9}$ s, and we have $l_{SR} = 0.19$ cm, which is in agreement with the above estimates. Elsewhere in the specimen, the superradiant pulse propagates through a partially inverted medium, which reacts to it and produces a specific pulse deformation.[13]

§4. CONCLUSIONS

Our experimental data and theoretical analysis lead us to the conclusion that, for the first time[20] in a molecular crystal containing an impurity, namely, a diphenyl crystal containing pyrene molecules, we have observed the phenomenon of superradiance propagating under the conditions of self-induced transparency. The process exhibits all the features observed in experiments on superradiance in gaseous media,[6-10] but there are also new features (both polarization and spatial) connected with the crystal structure.

The proposed method of excitation is also of methodological interest because it enables us to investigate the evolution of collective processes under the conditions of nonresonance excitation, which is important for spectroscopic studies of a wide range of molecular crystals containing impurities.

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

[2] A. V. Andreev, V. I. Emel'yanov, and Yu. A. Il'inskiĭ, Usp. Fiz. Nauk **131**, 653 (1980) [Sov. Phys. Usp. **23**, 493 (1980)].

[3] F. C. MacGillivray and M. S. Feld, Contemp. Phys. **22**, 299 (1981).

[4] A. Allen and J. H. Eberley, Optical Resonance and Two-Level Atoms, Wiley-Interscience, 1975 [Russ. transl. Mir, Moscow, 1978, Chap. 8].

[5] J. Macomber, The Dynamics of Spectroscopic Transitions Illustrated by Magnetic Resonance and Laser Effects, Wiley-Interscience, 1975 [Russ. transl., Mir, Moscow, 1979].

[6] N. Skribanowitz, J. P. Hermann, and F. C. MacGillivray, Phys. Rev. Lett. **30**, 309 (1973).

[7] A. Flusberg, T. Mossberg, and S. R. Hartmann, Phys. Lett. A **58**, 373 (1976).

[8] M. Gross, C. Fabre, P. Pillet, and S. Haroch, Phys. Rev. Lett. **36**, 1035 (1976).

[9] A. T. Rosenberger, T. A. Detple, C. M. Bowden, and C. C. Sung, J. Opt. Soc. Am. **68**, 700 (1978).

[10] Q. H. F. Vrehen, M. M. J. Hikspoors, and H. M. Gibbs, Phys. Rev. Lett. **38**, 764 (1977).

[11] A. Bree and V. B. Vilkos, Spectrochim. Acta **27**, 2333 (1971).

[12] J. B. Birks and J. N. Munro, Progr. React. Kinetics **4**, 239 (1967).

[13] P. V. Zinov'ev, G. G. Zaĭtseva, Yu. V. Naboĭkin, V. V. Samartsev, N. B. Silaeva, and Yu. E. Sheĭbut, Zh. Eksp. Teor. Fiz. **77**, 1519 (1979) [Sov. Phys. JETP **50**, 761 (1979)].

[14] Yu. V. Naboĭkin and V. V. Samartsev, Izv. Akad. Nauk SSSR Ser. Fiz. **46**, 1491 (1982).

[15] I. M. Lizin, T. M. Makhviladze, and L. A. Shelepin, "Coherent cooperative processes," Tr. Fiz. Inst. Akad. Nauk SSSR **87**, 21–37 (1976).

[16] M. J. Konopnicki and J. H. Eberly, Phys. Rev. A **24**, 2567 (1981).

[17] F. C. MacGillivray and M. S. Feld, Phys. Rev. A **14**, 1169 (1976); Phys. Rev. A **23**, 1334 (1981).

[18] R. F. Malikov, V. A. Malyshev, and E. D. Trifonov, in: Teoriya kogerentnykh kooperativnykh effektov v izluchenii (Theory of Coherent Cooperative Effects in Radiation), Leningrad State Pedagogical Institute, 1980, pp. 3–32; Zh. Eksp. Teor. Fiz. **76**, 65 (1979) [Sov. Phys. JETP **49**, 33 (1979)].

[19] E. A. Tikhonov and M. T. Shpak, Nelineĭnye opticheskie yavleniya v organicheskikh kristallakh (Nonlinear Optical Phenomena in Organic Crystals), Naukova Dumka, Kiev, 1979, p. 144.

[20] P. V. Zinov'ev, S. V. Lopina, Yu. V. Naboikin, N. B. Silaeva, and Yu. E. Sheĭbut, Tezisy 2-go Vsesoyuz. simp. po svetovomu ekho (Abstracts of Second All-Union Symposium on the Light Echo) Kazan', 1981, p. 52.

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