

S. N. Andrianov, Yu. V. Naboïkin, V. V. Samartsev, N. B. Silaeva, and Yu. E. Sheïbut. *Optical superradiance in a diphenyl crystal with pyrene.* Up to now the overwhelming majority of experiments on optical superradiance (coherent spontaneous emission) has been performed with gases,¹ since in gases it is technically simpler to provide the model conditions built into the theory. The first experiments on optical superradiance (SR) in solids, namely, in extrinsic crystals, were reported in Refs. 2 and 3. The high density of impurity centers per unit volume, the diversity of the mecha-

nisms by which excitation is transferred between centers and into the lattice, and the optical anisotropy of the crystals associated with their symmetry are by no means a complete list of factors determining the characteristics of SR in crystals. This report is devoted to the discussion of the basic characteristics of the SR in a mixed molecular crystal—diphenyl with pyrene molecules²—as well as the analysis of the results of application of SR to the investigation of relaxation processes.⁴

The inverted population difference between the work-

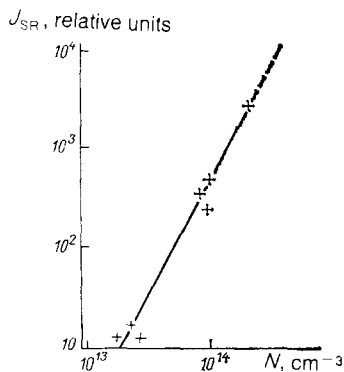


FIG. 1. Concentration dependence of superradiance intensity in a diphenyl crystal with pyrene at a temperature of 2.2 K. The width of the spectrum of pumping 1→3 and emission (2→1) is narrower than the inhomogeneous width of the line (10^{10} – 10^1 s⁻¹) by two to three orders of magnitude.

ing levels [¹A_g (1) and ¹B_{2u} (2)] required in order to realize superradiance was achieved through a third (electronic-vibrational, short-lived) level 3, narrow-band pumping of which was carried out at the wavelength 3530 Å with a YAG laser pulse with a duration of 8–10 nsec. As a result of the nonradiative transition 3→2 the electrons rapidly (within a time of $\approx 10^{-11}$ s) populated the state 2. Superradiance was formed on the transition 2→1 (0–0 transition) at the wavelength 3739.3 Å. When definite threshold powers for the pulse pumping the level 3 (equal to $5 \cdot 10^5$ W/cm²) was achieved, the deexcitation time for the 0–0 transition decreased markedly from 110 ns to 5–6 ns. Superradiance was characterized by the sharp spatial directedness (with a solid angle of 0.1 sr) on both sides, and its intensity J_{SR} was one thousand times higher than that of the incoherent spontaneous background. It turned out to be highly polarized (the degree of linear polarization equalled 0.95), and the polarization plane of the SR formed an angle of 100–110° with the *b* axis of the crystal, along which the pyrene absorption is polarized. Checking the concentration dependence of SR, shown in Fig. 1, presented the greatest technical difficulties. This dependence exhibited a quadratic character ($J_{SR} \propto N^2$, where N is the number of active impurity centers per cm³) in agreement with the theory of SR.¹ The delay time in the appearance of the SR pulse equals 2 ns.⁵ We note that the SR pulse formed in the effective part of the sample ($l_{eff} \approx 0.15$ cm with a total sample length l equal to 0.4 cm) propagated into the partially inverted resonance medium, subject to its “reaction.” A special investigation carried out in Ref. 6 showed that this resonance passage of the pulse occurred in the regime of self-induced transparency (SIT). This regime corresponded to the characteristic (for SIT) kinetics of deformation of the SR pulse shape (self-broadening, self-compression, self-division), whose analysis yielded the parameters of the resonance medium (the modulus of the electric dipole moment of the resonance transition 2–1 equals 10^{-19} CGSE units; the transverse irreversible relaxation time T_2 at a temperature of 2.2 K equals 10 ns). The delay time of the superradiance pulse owing to SIT equalled 7 ns.

The SR phenomenon was later used to investigate relax-

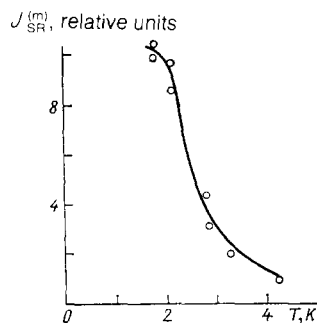


FIG. 2. Temperature dependence of the superradiance intensity at maximum in a diphenyl crystal containing pyrene molecules with a concentration of 0.25 mole %.

ation (with the participation of phonons) in a diphenyl crystal with pyrene.⁴ To this end, the temperature dependence of the SR intensity at maximum $J_{SR}^{(m)}$ in diphenyl crystals with different concentrations (C) of pyrene molecules was measured in the experiment. This dependence is presented in Fig. 2 for $C = 0.25$ mole %. The decay of the SR intensity with increasing temperature is caused by Orbach relaxation with the participation of pseudolocalized phonons, whose frequency lies in the continuous spectrum of the allowed phonon frequencies of the matrix. As a result of the absorption of the phonon energy the unpaired electron of the pyrene molecule is transferred from the state 2 into the electronic-vibrational state 2', in which the pyrene molecule undergoes librational oscillations. Returning rapidly to the state 2 (and, correspondingly, giving up a phonon to the lattice), the electron completely loses in the process of such a double transition information about the phase, as a result of which this molecule is “knocked out” of the SR process. The calculation of the dynamic variables, describing the behavior of the system as a function of time, was carried out by the method of the nonequilibrium statistical operator. It showed that the SR intensity at maximum $J_{SR}^{(m)}$ is proportional to the factor

$$\left[\frac{1}{\tau_c} - \frac{1}{\tau_{ph}} \exp\left(-\frac{\Delta E}{k_B T}\right) \right]^2,$$

where τ_c is the correlation self-induction time (I), defined by the expression $\tau_c = (4\pi A / \lambda^2) T_1 / N'$, T_1 is the spontaneous decay rate of one atom; A is the active part of the transverse cross section of a cylindrical sample; λ is the wavelength of SR; τ_{ph} and ΔE are the lifetime and energy of activation of pseudolocalized phonons; k_B is the Boltzmann constant; T is the absolute temperature of the crystal; and, $N' = N \cdot l_{eff} A$. Using the values of $J_{SR}^{(m)}$, corresponding to the three values of the temperatures in the graph of Fig. 2, it is possible to find two independent parameters: τ_{ph} and ΔE . In particular, for a diphenyl crystal with a pyrene concentration of 0.25 mole %, these parameters equal $\tau_{ph} = 1$ –15 ps and $\Delta E = 7 \pm 3$ cm⁻¹.

We note that in all these investigations the main condition for the formation of SR $l/c < \tau_c < T_1, T_2$ (where c is the velocity of light in the crystal) was satisfied. The question of whether or not this coherent emission is superluminescence

(amplification of spontaneous emission) was specially investigated. It was found that under the conditions of the present experiment it is optical superradiance that forms on the transition ${}^1B_{2u} \rightarrow {}^1A_g$ of pyrene in diphenyl.

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⁵P. V. Zinov'ev, S. V. Lopina, Yu. V. Malyukin, Yu. V. Naboïkin, N. B. Silaeva, and V. V. Samartsev, *Zh. Prikl. Spektrosk.* **43**, 683 (1985).

⁶P. V. Zinov'ev, G. G. Zaitseva, Yu. V. Naboïkin, V. V. Samartsev, N. B. Silaeva, and Yu. E. Sheïbut, *Zh. Eksp. Teor. Fiz.* **77**, 1519 (1979) [*Sov. Phys. JETP* **50**, 761 (1979)].