Effect of phonon disequilibrium on the optical Dicke super-radiance

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Optical Dicke super-radiance is investigated for the first time, using a pyrene-doped diphenyl crystal as the example, under conditions when the temperature of the system of impurity centers differs from the lattice temperature. The influence of lattice phonons and of pseudolocalized phonons on the course of this phenomenon is investigated. The effective temperature of the local heating is found ($T_{\text{eff}} = 4.45$ K) and the temperature increment produced by each succeeding pump pulse is determined (0.25 K). The optical Dicke super-radiance method is used to determine the propagation velocity of pseudolocalized phonons (10^4 cm/s) and the specific heat of a pyrene-doped diphenel crystal (1.15 kgf^{-1}J/deg). A theory of optical super-radiance in crystals is developed in the nonequilibrium statistical operator formalism.

1. INTRODUCTION

In Ref. 1 Dicke predicted optical super-radiance (SR), in which a system of excited particles emits spontaneously a narrow coherent photon beam whose intensity is proportional to the square of the number of photons. Super-radiance is the result of self-induced correlations between the emitters through their common radiation field. The overwhelming number of experimental observations of SR, which date back to 1973, were in gases (see the review by Andrew *et al.*[2]). Only a few recent papers are devoted to experimental study of SR in solids, viz., doped crystals.[3–5]

One of the main features of SR in solids is the influence that phonons exert on it. This was manifested in the temperature dependence of the SR intensity plotted in Ref. 4, where an equilibrium phonon subsystem was assumed. Frequently, howver, no such assumption can be made. Thus, in exact quantitative measurements of the SR intensity, the pumping pulse is applied to the crystal repeatedly. The excess energy (equal to the difference between the pump and super-radiance photons) goes to molecule vibrations at definite frequencies, and this in turn increases substantially the number of phonons having these frequencies. When the time intervals between the pump pulses is decreased, there is no time for the energy of these phonons to go over to other phonons, so that the phonon sytem is in disequilibrium. We shall refer to this character of crystal-lattice heating as local. The present paper is in fact devoted to the properties of SR formation in a pyrene-doped diphenyl crystal under conditions of local heating. In our investigation we determined the effective local-heating temperature, the specific heat of the investigated crystal, and the average propagation velocity of the pseudolocalized phonons that take part in the relaxation.

2. THEORY OF OPTICAL DICKE SUPERRADIANCE UNDER CONDITIONS OF LOCAL HEATING OF A MIXED MOLECULAR CRYSTAL

We use the energy scheme, shown in Fig. 1, of the levels that take part in the excitation and evolution of optical super-radiance in mixed molecular crystal. The pumping is via the energy transition $|1\rangle \to |3\rangle$. As a result of the rapid nonradiative transition (within a time 10^{-10}–10^{-11} s), the electrons are in the state $|2\rangle$ and under certain conditions the system of impurity molecules is inverted. The first electrons that break away in the spontaneous transitions $|2\rangle \to |1\rangle$ influence, via their radiation field, the electrons that remain in the state $|2\rangle$, causing them to go over next into state $|1\rangle$. The growth of the number of such transition is avalanche-like. If the time of self-induction of correlations in the emission of the impurity molecules is shorter than the

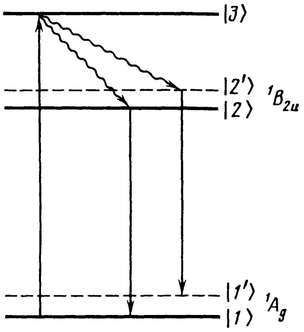

FIG. 1. Energy-level scheme of an impurity molecule in a molecular crystal.

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times of the longitudinal (T_1) and transverse (T_2) irreversible relaxations, Dicke super-radiance is realized in the transition designated $|2> \rightarrow |1>$ in Fig. 1 (the (0-0 transition). Depending on the experimental conditions, its formation is influenced by various relaxation processes. At very low temperatures (~ 1.5 K) intermolecular interactions predominate, as well as certain types of spectral diffusion. As the temperature is raised, relaxation processes with participation of phonons become activated. In mixed molecular crystals, at liquid-helium temperature, the principal role is played in the damping by the Orbach relaxation. In many such crystals, including pyrene-doped diphenyl,[4] pseudolocalized phonons with activation energy on the order of 4–16 cm^{-1} take part in this relaxation. Libration of the impurity molecules sets in, and the electrons previously in the $|2>$ state turn out to be in a short-lived electron-vibrational state $|2'>$. Although the electrons may return to the $|2>$ state after 10^{-11} s, nontheless the data on the phase are lost, and the impurity molecule itself is "knocked out" of the super-radiance process. This lead to a temperature decrease of the super-radiance intensity.[4] It follows from the foregoing that, from the large set of lattice vibrations, the impurity molecules are acted upon mainly by "resonant" phonons. Their frequency coincides with the molecule-libration frequency, which in turn is determined by the scatter of the spatial orientations of the impurity molecule in the matrix and by the nature of the molecules.

Under conditions of local heating of the impurity molecules, the energy of the nonradiative transitions $|3> \rightarrow |2>$ and $|3> \rightarrow |2'>$ is "dispersed" into many phonons, including pseudolocalized resonant ones. In view of their relatively slow motion through the crystal,[6,7] they transfer their energy to the helium heat bath much more slowly than the delocalized phonons. This energy is therefore transferred mainly by transformation of the pseudolocalized phonons into delocalized ones that escape subsequently through the crystal boundaries. If, however, the number of pseudolocalized phonons is large enough, i.e., at sufficiently high densities of the impurity molecules, before a delocalized phonon manages to reach the sample boundary it manages to become reabsorbed and reradiated many times and to interact with the pseudolocalized phonon modes, thereby hindering in principle the relaxation in this channel, too. Local heating of the sample is thereby made possible. If under conditions of local heating one applies to the heat bath not a single pump pulse but a sequence of pulses at intervals shorter than the resonant-phonon relaxation time, the heating will be increased by each new pulse, and the relaxation with participation of resonant phonons becomes stronger, thereby causing damping of the SR.

Thus, let us consider a system of N impurity molecules in a molecular matrix and investigate its behavior at times t longer than the correlation of self-induction in SR and the time of formation of the nonequilibrium pseudolocalized phonon mode. We shall regard the pseudolocalized oscillations and the continuum oscillaton of the lattice as two interacting subsystems (this interaction is manifested by the existance of anharmonic terms in the expression for the crystal potential energy[6]). We express the Hamiltonian of the system in the form

$$\mathscr{H}=\mathscr{H}_0+\mathscr{H}_1, \tag{1}$$

where

$$\mathscr{H}_0=\mathscr{H}_d+\mathscr{H}_l+\mathscr{H}_{\text{pn}}+\mathscr{H}_{\text{pn}}, \tag{2}$$

$$\mathscr{H}_1=\mathscr{H}_{l-\text{pn}}+\mathscr{H}_{d-\text{pn}}+\mathscr{H}_{d-\text{pn}} \tag{3}$$

is the perturbation Hamiltonian, $\mathscr{H}_d$ is the electron-energy operator of the system of impurity molecules (in the two-level approximation, $\mathscr{H}_l$ is the energy operator of the pseudolocalized phonons, $\mathscr{H}_{\text{pn}}$ is the energy operator of the lattice phonons, $\mathscr{H}_{\text{pt}}$ is the photon-field energy operator, $\mathscr{H}_{l\text{-pn}}$ is the operator of the resonant and nonresonant interactions between the pseudolocalized and delocalized (lattice) phonons[6–8] (allowance is made in the nonresonant interaction for only processes such as Raman scattering, in which the creation or annihilation of a pseudolocalized phonon is accompanied by creation of one lattice phonon and annihilation of another), $\mathscr{H}_{d\text{-pn}}$ is the electron-phonon interaction operator, responsible for the temperature dependence of the homogeneous linewidth of the optical transition and due to noncorrelated scattering by phonons,[7,8] and $\mathscr{H}_{d\text{-pt}}$ is the electron-photon interaction operator in the two-level model and in the dipole approximation.

To describe the dynamics of the evolution of the system in the super-radiance process, we use Zubarev's[9] nonequilibrium-statistical operator (NSO) method (as modified by Pokrovskiĭ[10]). We chose the following quasi-integrals of the motion:

$$R_z=2\sum_j S_j^z,$$

$$R_{\mathbf{k}_0}{}^2=\sum_{ij}\left\{\frac{1}{2}[\exp(i\mathbf{k}_0\mathbf{r}_{ij})S_i^+S_j^-+\text{H.c.}]+S_i^zS_j^z\right\}, \tag{4}$$

$$N_{js}=B_{js}^+B_{js}, \quad n_q=b_q^+b_q,$$

where $S^{\pm}=S^x \pm iS^y$; S_x, S_y, and S_z are the components of the energy spin $S=1/2$; B_{js}^+ and B_{js} are the creation and annihilation operators of a pseudolocalized phonon of mode s (of frequency Ω_{js}) and connected with the impurity site f; b_q^+ and b_q are the operators of creation and annihilation of a lattice phonon with wave vector $\mathbf{q}$ and polarization δ (we use a common subscript for both parameters); $\mathbf{r}_{ij}$ is the radius vector joining the locations of the ith and jth impurity molecules, and $\mathbf{k}_0$ is the super-radiance wave vector. These quasi-integrals of the motion have respectively the meaning of the operators of the collective population difference of the electron levels, the square of the Bloch vector, the number of pseudolocalized phonons of mode s, and the number of delocalized phonons of mode q.

In second-order perturbation theory in $\mathscr{H}_1$, the system of kinetic equations for the mean values of the quasiintegrals of the motion are written in the form

$$\frac{d\langle R_z\rangle}{dt}=-\frac{1}{\tau_1}(\langle R_z\rangle+N+2\mu\langle\mathscr{P}_{\mathbf{k}_0}\rangle), \tag{5}$$

$$\frac{d\langle\mathscr{P}_{\mathbf{k}_0}\rangle}{dt}=-\frac{1}{2}\langle R_z\rangle\frac{d\langle R_z\rangle}{dt}-\frac{1}{N}\sum_{jqs}\alpha_{qs}{}^j(\langle N_{js}\rangle\langle n_q\rangle$$

$$+\frac{\langle N_{js}\rangle+\langle n_q\rangle}{2}\Big)\langle\mathscr{P}_{\mathbf{k}_0}\rangle-\frac{1}{2\tau_1{}'}\Big\{\langle R_z\rangle(\langle R_z\rangle+N)+2\langle\mathscr{P}_{\mathbf{k}_0}\rangle\Big\},\quad(6)$$

$$\frac{d\langle N_{js}\rangle}{dt}=\sum_q\left(\beta_{qs}{}^j+\gamma_{qs}{}^j\frac{\langle R_z\rangle}{N}\right)(\langle n_q\rangle-\langle N_{js}\rangle)$$

$$+\sum_{q\neq q'}\xi_{qq'}^{js}\{\langle N_{js}\rangle(\langle n_q\rangle-\langle n_{q'}\rangle)+\langle n_q\rangle(\langle n_{q'}\rangle+1)\},\quad(7)$$

$$\frac{d\langle n_q\rangle}{dt}=\sum_{js}\left(\beta_{qs}{}^j+\gamma_{qs}{}^j\frac{\langle R_z\rangle}{N}\right)(\langle N_{js}\rangle)-\langle n_q\rangle)$$

$$-\frac{1}{\tau_{\mathrm{pn}}}(\langle n_q\rangle-\bar{n}_q)-\sum_{jq'S}\{(\xi_{qq'}^{js}+\xi_{q'q}^{js})\langle N_{js}\rangle(\langle n_q\rangle-\langle n_{q'}\rangle)$$

$$+(\xi_{qq'}^{js}-\xi_{q'q}^{js})\langle n_q\rangle\langle n_{q'}\rangle+\xi_{qq'}^{js}\langle n_q\rangle-\xi_{q'q}^{js}\langle n_{q'}\rangle\},\quad(8)$$

where

$$\langle\mathscr{P}_{\mathbf{k}_0}\rangle=\sum_{i=j}\langle S_i{}^+S_j{}^-\rangle\exp(i\mathbf{k}_0\mathbf{r}_{ij});$$

n_q is the equilibrium number of phonons with wave vector $\mathbf{q}$;

$$\frac{1}{\tau_1}=\frac{2\pi}{\hbar^2}\sum_{\mathbf{k}}|g_{\mathbf{k}}|^2\delta(\omega_{\mathbf{k}}-\omega_0);$$

τ_1 is the radiative lifetime of the isolated impurity center (and is due to the spontaneous transition)[11];

$$\mu=\frac{2\pi\tau_1}{\hbar^2}\sum_{\mathbf{k}}|g_{\mathbf{k}}|^2\Gamma(\mathbf{k}-\mathbf{k}_0)\delta(\omega_{\mathbf{k}}-\omega_0)$$

is the sample shape factor; ω_0 is the frequency of the resonant transition; $\omega_{\mathbf{k}}$ is the radiation frequency corresponding to the mode $\mathbf{k}$; $\sigma(\omega_{\mathbf{k}}-\omega_0)$ is the Dirac delta function;

$$\Gamma(\mathbf{k}-\mathbf{k}_0)=\left|\frac{1}{N}\sum_j\exp[i(\mathbf{k}-\mathbf{k}_0)\mathbf{r}_j]\right|^2,\qquad\frac{1}{\tau_1{}'}=\frac{1-\mu}{\tau_1},$$

$$\alpha_{\mathbf{q}s}{}^j=(4\pi/\hbar^2)|\varphi_{\mathbf{q}s}{}^j|^2\delta(\Omega_{js}-\Omega_{\mathbf{q}}),$$

$$\beta_{\mathbf{q}s}{}^j=(2\pi/\hbar^2)\left(|h_{\mathbf{q}s}{}^j|^2+\frac{1}{4}|\varphi_{\mathbf{q}s}{}^j|^2\right)\delta(\Omega_{js}-\Omega_{\mathbf{q}}),$$

$$\gamma_{\mathbf{q}s}{}^j=(\pi/\hbar^2)(\varphi_{\mathbf{q}s}{}^{j*}h_{\mathbf{q}s}{}^j+\mathrm{H.c.})\,\delta(\Omega_{js}-\Omega_{\mathbf{q}}),$$

$$\xi_{\mathbf{q}\mathbf{q}'}^{js}=(2\pi/\hbar^2)|\chi_{\mathbf{q}\mathbf{q}'}^{js}|^2\delta(\Omega_{js}-\Omega_{\mathbf{q}}+\Omega_{\mathbf{q}'});$$

$\varphi^j_{\mathbf{q}s}$ is the constant of the interaction of an unpaired electron of the jth impurity molecule with the phonons of mode s and with the wave vector $\mathbf{q}$; $h^j_{\mathbf{q}s}$ and $\chi^{js}_{\mathbf{q}\mathbf{q}}$ are the constants of the resonant and nonresonant interactions of the pseudolocalized and lattice delocalized phonons, respectively; Ω_{js} is the frequency of the pseudolocalized phonon of mode s coupled to the jth impurity site; $\Omega_{\mathbf{q}}$ is the frequency of the lattice phonons having a wave vector $\mathbf{q}$; $\mathbf{r}_j$ is the radius vector of the jth impurity molecules. We have introduced phenomenologically in these equations terms that describe the energy relaxation of the system of resonant lattice phonons to the heat bath (by heat bath is meant here the subsystem of nonresonant lattice phonons plus the helium bath) with a characteristic time τ_{pn} governed by the intrinsic anharmonicity in the system of lattice phonons and by the phonon scattering from the sample boundaries.

At liquid-helium temperatures ($T=1.5$–4.2 K), when resonant damping of pseudolocalized phonons to the "lattice" dominates, one can separate a certain central frequency of the librational (electron-vibrational) sublevel $|2'\rangle$. The form factor of the $|2\rangle\to|2'\rangle$ transition line is highly accurately Lorentzian:

$$f(\Omega_{js})=\Delta/[\Delta^2+\pi^2(\Omega_{js}-\Omega_0)^2],$$

where Δ is the line half width.

In the weak electron-phonon coupling approximation, when $|h_{\mathbf{q}s}{}^j|\gg|\varphi_{\mathbf{q}s}{}^j|$, as is the case for many mixed molecular crystals including the pyrene-doped diphenyl crystal, Eqs. (7) and (8) can be solved independently of Eqs. (5) and (6). This yields

$$\langle N(\Omega_0)\rangle=\bar{n}+a_1\exp(-t/\tau_l{}')+a_2\exp(-t/\tau_{\mathrm{pn}}),\quad(9)$$

where n is the equilibrium number of phonons,

$$a_1=\frac{(1+\sigma)\tau_{\mathrm{pn}}\delta N}{\sigma\tau_l+(1+\sigma)^2\tau_{\mathrm{pn}}},\qquad a_2=\frac{\sigma\{\tau_l+(1+\sigma)\tau_{\mathrm{pn}}\}\delta N}{\sigma\tau_l+(1+\sigma)^2\tau_{\mathrm{pn}}},$$

δN is the initial excess of vibrational photons in the pseudolocalized phonon mode $\sigma=(v/u)^3$, while v and u are the velocities of the lattice and pseudolocalized phonons. From the same system of kinetic equations (5)–(8) we obtain

$$\langle n(\Omega_0)\rangle=\bar{n}-a_1\exp(-t/\tau_l{}')+a_2\exp(-t/\tau_{\mathrm{pn}}).\quad(10)$$

We have introduced in Eqs. (9) and (10) the following new parameters: $\tau_l'=\tau_l/(1+\sigma)$ is the effective time in which equilibrium is established between the pseudolocalized-phonon subsystem and the resonant lattice phonon subsystem; $\tau_{\mathrm{pn}}=\tau_l+(1+\sigma)\tau_{\mathrm{pn}}$ is the effective relaxation time of the combined system of pseudolocalized and resonant lattice phonons in the heat bath. The role of the action of these parameters in the two-step dissipation process is illustrated in Fig. 2. From (9) and (10) follows an effective lengthening of the times of relaxation to the thermostat at $\sigma\gg1$, in analogy with the "phonon-bottleneck" effect in paramagnetic relaxation.[12] At low pseudolocalized-phonon velocities ($u\ll v$) this leads to local heating of the sample.

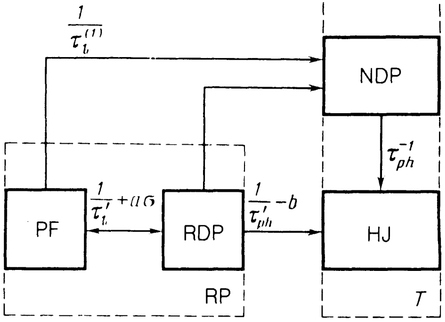

FIG. 2. Energy transfer from pseudolocalized phonons (PF) to a heat bath (T): RP—resonant phonons, EDP—resonant delocalized phonons, HT—helium heat bath.

Let us dwell on the contribution of the cubic anharmonicity to the interaction between the pseudolocalized and lattice phonons. In the low-temperature approximation one need take into account only the resonant phonons, with allowance for a frequency spread within the inhomogenous $|2\rangle \to |2'\rangle$ line width. Under this condition, solution of the system (5)–(8) yields

$$\langle N(\Omega_0)\rangle = C_1 + a_1' \exp\,(b - 1/\sigma\tau_{\rm pn}) + a_2' \exp\,[-\sigma(1/\tau_l + a)], \tag{11}$$

where C_1, a_1', and a_2' are constants,

$$b = \rho(\Omega_0)\frac{2\pi\Delta}{\hbar^2}\sum_{q'(\neq q_0)}(|\chi_{q_0q'}(\Omega_0)|^2 + |\chi_{q'q_0}(\Omega_0)|^2)\bar{n}_q,$$

$$a = \rho(\Omega_0)\frac{2\pi\Delta}{\hbar^2}\sum_{q'(\neq q_0)}|\chi_{q_0q'}(\Omega_0)|^2,$$

and $\rho(\Omega_0)$ is the density of the phonon states of the lattice; the summation is over the frequencies of the inhomogeneous width.

Analysis of the solution (11) shows that the cubic anharmonicity of the aforementioned phonons leads to an additional lengthening of the relaxation time $\tau_{\rm pn}'$ of the electronic excitation to the thermostat and to acceleration of the establishment of quasiequilibrium between the two phonon subsystem. The changes are due to Raman-type scattering by nonresonant phonons, at frequencies within the limits of the inhomogeneous width of the librational level. The situation is made clear in Fig. 3a. Similar processes accompany absorption (emission) of a resonant lattice phonon with emission (absorption) of a nonresonant phonon whose frequency is within the inhomogeneous width of the librational sublevel (since there is no broad spectrum of thermal phonons at helium temperatures). On the other hand, with increase of temperature (from 4.2 to 20 K and higher) one can neglect the contribution of the resonant phonons and the scattering processes (described by terms of type $B_{js}^{+}b_q b_q^{+}$) and take into account only scattering by nonresonant phonons. The character of this scattering is explained in Fig. 3b. We are dealing here with processes of the type of Raman scattering with absorption (emission) of a large photon from the lattice, with frequency $\Omega_q > \Omega_0$, and emission (absorption) of a difference-frequency phonon (see, e.g., Ref. 12). In this case solution of Eqs. (7) and (8) yields

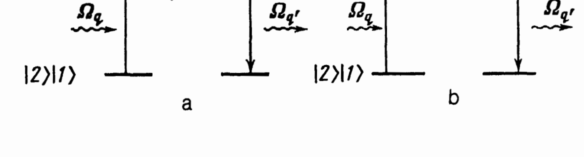

FIG. 3. Inelastic (Raman) scattering of lattice delocalized phonons by pseudolocalized phonons: a) with participation of real levels belonging to the inhomogeneously broadened profile of the pseudolocalized phonons; b) with participation of virtual levels γ. The energy conservation law $\hbar\Omega_{js} = |\hbar\Omega_q - \hbar\Omega_q'|$ holds in all the processes.

$$\langle N(\Omega_0)\rangle = C_1 + a_1 \exp\left[-\frac{1}{\sigma\tau_{\rm pn}} - \frac{\sigma-1}{\sigma\tau_l^{(1)}}\right] + a_2 \exp\left[-\frac{\sigma+1}{\tau_l} - \frac{\sigma+1}{\sigma\tau_{\rm pn}} - \frac{1}{\tau_l^{(1)}}\right], \tag{12}$$

where

$$\frac{1}{\tau_l^{(1)}} = \sum_{\mathbf{q}\neq\mathbf{q}'}\{\xi_{\mathbf{q}'\mathbf{q}}(\Omega_0) - \xi_{\mathbf{q}\mathbf{q}'}(\Omega_0)\}\bar{n}_{\mathbf{q}}. \tag{13}$$

Analysis of (12) shows that on satisfaction of the inequality

$$|(\sigma-1)/(\sigma\tau_l^{(1)})| > 1/(\sigma\tau_{\rm pn}),$$

which is valid for many mixed molecular crystals at the temperatures indicated, processes of the "phonon bottleneck" type are hindered. In this case the effective relaxation is via intermediate levels of the continuous phonon spectrum on account of the strong phonon anharmonicity.

Assuming that super-radiance develops over times $\tau_1' \ll t \ll \tau_{pn}'$, where the change of the phonon occupation numbers can be neglected compared with the change of the electron-level population difference, and using the law of energy conservation in an "impurity molecules plus field" system, we obtain the following expression for the intensity of the optical Dicke super-radiance:

$$I_{\rm SR} = \frac{\hbar\omega_0}{4\mu(\tau_R)^2}\tau_1\,{\rm sech}^2\frac{t-t_0}{2\tau_R}, \tag{14}$$

where $1/\tau_R = \mu N/\tau_1 - 1/T_2$ is the reciprocal SR pulse duration, t_0 is the delay of the SR pulse, T_2 is the phase-relaxation time defined in the present case as

$$\frac{1}{T_2} = \frac{1}{T_2(\infty)}\left(\frac{\langle N(\Omega_0)\rangle + \langle n(\Omega_0)\rangle}{2} + \langle N(\Omega_0)\rangle\langle n(\Omega_0)\rangle\right),$$

$$\frac{1}{T_2(\infty)} = \frac{4\pi}{N\hbar^2}\sum_j\sum_{qs}|\varphi_{qs}^{\,j}|^2\delta(\Omega_{js} - \Omega_q),$$

and $T_2(\infty)$ is the lifetime of the continuum phonons (in Ref. 4, $T_2(\infty)$ was designated $\tau_{\rm pn}$). In the low-temperature approximation we can obtain the expression

$$\frac{1}{T_2} = \frac{1}{T_2(0)} + \frac{1}{T_2(\infty)}\exp\left(-\frac{\hbar\Omega_0}{k_B T}\right). \tag{15}$$

Here

$$\frac{1}{T_2(0)} = \frac{\sigma}{1+\sigma}\frac{\delta N(\Omega_0)}{T_2(\infty)},$$

k_B is the Boltzmann constant, and $\delta N(\Omega_0)$ is the initial excess of the pseudolocalized phonons over their equilibrium value. Recognizing that the average number of resonant phonons is $\langle N\rangle = \delta N(\Omega_0) + \exp(\hbar\Omega_0/k_B T)$, we obtain for $\sigma \gg 1$

$$\frac{1}{T_2} = \frac{1}{T_2(\infty)}\langle N\rangle. \tag{16}$$

Let us dwell briefly on the influence of the inhomogeneous broadening on the formation of the SR. The theory of this influence was developed in a number of papers (e.g., in Refs. 13–15). Let us discuss the physics of the problem under conditions of broad-band excitation of the line. We consider for the sake of clarity a set of pendulums (approximately equal in length) fastened on one stretched string and all in the upper unstable equilibrium position. Each pendulum is taken to correspond to a definite molecule in an excited state, and the stretched string characterizes the radiation field common to all molecules. Assume that one pendulum left its upper unstable position and began its fall. This fall is "felt" through the common string by the remaining pendulums, so that all begin to fall after a certain time. In the upshot, an avalanche of falling pendulums sets in, the equivalent of the super-radiant avalanche. Since the pendulum lengths are unequal (as are the working frequencies ω_0^j of the molecules under inhomogeneous broadening conditions), the set of phases after one and the same time interval t will be different (with values $\varphi_j = \omega_0^j t$ for the different molecules). If the phase difference turns out to exceed π, the oscillations of such pendulums become subsequently mutually incoherent. The most important feature of superradiance is the correlation self-induction time τ_c (for the set of pendulums this is the time of propagation, along the string, of a signal reporting the fall of the first pendulum). If this time is very long, the fall of even the nearest pendulum can start after the first pendulum has acquired a phase difference larger than π, and the avalanche is incoherent from the very outset. For an actual impurity crystal characterized by a definite value of τ_c, the molecules that will participate in the super-radiance correspond to a spectral width τ_c^{-1}, i.e., a set of spectral "packets" within the limits of the indicated width will be "cut out" from the inhomogeneously broadened line. In the case of pyrene-doped dihenyl, the inhomogeneously broadened ${}^1A_g \leftrightarrow {}^1B_{2u}$ line is characterized by three broad peaks.[4] Since the number of molecules corresponding to each peak is much larger than for the remaining sections of the line, and the SR intensity is proportional to N^2, the probability of a super-radiant avalanche at the frequency of the peaks turns out to be considerably larger. The peaks of the super-radiance spectrum become sharply pronounced.[4] Thus, excitation of SR is possible even under conditions of broadband excitation, and the self-induction of correlations serves also as a "monochromator" of sorts.

A few words concerning some estimates for pyrene-doped diphenyl crystals (transition ${}^1B_{2u} \to {}^1A_g$). The correlation self-induction time τ_c is estimated at $(3N\lambda^2 L/8\pi\tau_1)^{-1}$, where $N = 10^{13} - 10^{14}$ cm^{-3}, $\lambda = 3{,}73\cdot 10^{-3}$ cm, $L = 0{,}2$ cm, and $\tau_1 = 10^{-7}$ s (corresponding to a transition electric dipole moment of several Debye). Estimates yield $\tau_c = 10^{-10}$–10^{-11} s, and a part of inhomogeneously broadened line with spectral width $\approx \tau_c$ will take part in the SR process.

3. EXPERIMENTAL INVESTIGATION OF OPTICAL DICKE SUPER-RADIANCE IN A PYRENE-DOPED DIPHENYL CRYSTAL UNDER LOCAL HEATING CONDITIONS

The SR experiment was perfomed on a pyrene-doped diphenyl crystal (measuring $0.1\times 0.1\times 0.4$ cm) in the temperature range 1.5–4.2 K. The concentration of the pyrene molecules was 0.25 mol.%. The SR was produced on the 0–0 (${}^1B_{2u} \to {}^1A_g$) transition of pyrene at a wavelength $\lambda = 373.9$ nm. The excitation ($|1\rangle \to |3\rangle$ pumping) sources were three laser types with different degrees of pulsed-radiation coherence: 1) nitrogen laser ($\lambda = 337$ nm, emission spectrum width $\Delta\nu \approx 10$ cm^{-1}, pulse duration $\Delta t_p = 8$ ns); 2) YAG:Nd^{3+} laser ($\lambda = 354.7$ nm, $\Delta\nu \approx 0.1$cm^{-1}, $\Delta t_p = 10$ ns); 3) neodymium-glass laser ($\lambda = 353.2$ nm, $\Delta\nu \approx 25$ cm^{-1}, $\Delta t_p = 10$ ns). Experiment has shown that irrespective of the degree of monochromaticity and coherence of the excitation, SR was always observed and its intensity was practically independent of the coherence of the source radiation. At the same time, the pump-spectrum width influenced the spectral characteristics of the SR pump, viz., the SR signal was emitted by the entire inhomogeneously broadened line in the case of broad-band pumping and by its spectral components for narrow-band pumping. The experimental apparatus is described in Ref. 3. The spectral properties of the SR as functions of the excitation method were studied with a Fabry–Perot interferometer and an STÉ − 1 spectrograph. The SR kinetics was recorded with an ÉLU-FT coaxial photomultiplier and an I2 − 7 high-speed oscilloscope. In contrast to Refs. 3 and 4, the SR signal was recorded for both sides of the z axis, as illustrated in Fig. 4. The polar diagram of the SR flashes turned out to be equal in both z directions.

To investigate the influence of the local heating of the crystal on the SR, we varied in the experiment the pump-pulse repetition frequency. A nitrogen-laser pump was used for the quantitative measurements. In that case it was technically most convenient to record the SR pulse parameters after the second pump pulse. The dependence of the relative intensity $I_{\rm SR}$ of this SR signal on the pump pulse repetition frequency is shown in Fig. 5a. The crystal temperature was 4.2 K. It was found that at a repetition frequency $f = 10$ Hz the SR signal intensity of each succeeding pulse decreased, vanished after the twelfth pulse, and ordinary luminescence was observed. No noticeable changes of the SR intensity was observed at a crystal temperature 1.5 K for pump pulse repetition frequencies in the range from 2 to 25 Hz. The SR behaved differently in the temperature range $T = 2.5$–3.5 K, where it changed into luminescence already after the fifth pump pulse (at $f = 10$ Hz). The decrease of the SR-pulse intensity after each succeeding pump pulse is demonstrated in Fig. 5b.

4. ANALYSIS OF EXPERIMENTAL DATA

The starting point for the estimates is Eq. (14), which is written out for two pump-pulse repetition frequencies (f and f'). Using Eq. (16), where the value of $\langle N\rangle$ after the l th pulse is

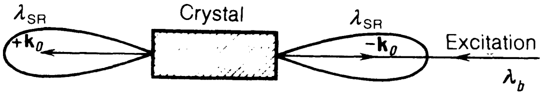

FIG. 4. Spatial pattern of superradiance of a pyrene-doped dyphenyl crystal.

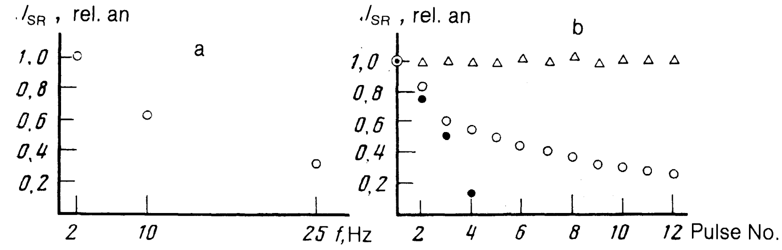

Fig. 5. Results of experimental investigation of superradiance (SR) in a pyrene-doped diphenyl crystal: a) SR intensity after the second pump pulse vs the pulse repetition frequency at 4.2 k; b) SR intensity after each succeeding pulse at $f = 10$ Hz and at sample temperatures 1.5 K (Δ), 3.03 K (●) and 4.2 K (○).

$$\langle N\rangle^{(l)}=\bar{n}+\delta N\frac{1-\exp(-l/f\tau_{\mathrm{pn}})}{1-\exp(-1/f\tau_{\mathrm{pn}})}, \tag{17}$$

we obtain for the ratio of the SR signal intensities after the second and first pump pulses

$$\frac{I_2(f)}{I_1}=\left[1-\frac{\tau_R}{T_2(0)}\left(1+\frac{\tau_R}{T_2(\infty)}\langle N\rangle^{(1)}\right)^{-1}\times\exp\left(-\frac{1}{f\tau_{\mathrm{pn}}}\right)\right]^2. \tag{18}$$

The relation for the repetition frequency f' is similar. Dividing one by the other, we obtain an equation for the effective relaxation time:

$$\tau_{\mathrm{pn}}=[(f')^{-1}-(f)^{-1}]\left\{\ln\frac{[1-(I_2(f)/I_1)]^{1/2}}{[1-(I_2(f')/I_1)]^{1/2}}\right\}^{-1}. \tag{19}$$

We turn to Fig. 5b, where we use for the estimates the values of $I_2(f)$ and $I_2(f')$ pertaining respectively to the repetition frequencies $f = 25$ Hz and $f' = 10$Hz, and assume for I_1 the value of I_2 at the repetitive frequency 2 Hz (inasmuch as $I_2 \approx I_1$ in this case). The estimates show that $\tau'_{\mathrm{pn}} = 9\cdot 10^{-2}$ s in a pyrene-doped diphenyl crystal. The time τ_{pn} of relaxation of the lattice phonons to the thermostat is determined mainly by their scattering from the sample boundaries. In this case we have[12]

$$\tau_{\mathrm{pn}}=(2Q+1)L/2v, \tag{20}$$

where Q is the sample quality factor, L the linear dimension of the sample, and v the lattice-phonon velocity. At $Q = 10$, $L = 0.4$ cm, and $v = 3\cdot 10^5$ cm/s we have $\tau_{\mathrm{pn}} \approx 10^{-5}$ s. Since $\tau_l \approx 10^{-10}$–10^{-11} s and $\tau'_{\mathrm{pn}}/\tau_{\mathrm{pn}} \gg 1$ we get $\sigma \approx \tau'_{\mathrm{pn}}/\tau_{\mathrm{pn}} \approx 10^4$. The allows us to find the velocity of the pseudolocalized phonons, viz., $u = v/\sigma^{-1/2} \approx 10^4$ cm/s. We estimate T_2 from Eq. (8) and from the $I_{SR}(f)$ plot of Fig. 5a. At the measured reciprocal SR duration $\tau_R^{-1} = 2\cdot 10^8$ s^{-1} we obtain $T_2(0) \approx 7\cdot 10^{-9}$ s. According to Ref. 4, $T_2(\infty) \approx 10^{-11}$ s, therefore $\delta N = T_2(\infty)/T_2(0) \approx 10^{-3}$. At $T = 4.2$ K, when $\bar{n} \approx 3\cdot 10^{-2}$, we have $\langle n\rangle^{(1)} \approx 4\cdot 10^{-3}$, which is the average number of phonons at the time of the first response. This enables us to find the sample's effective local heating temperature T_{eff} from the equation

$$T_{\mathrm{eff}}=\Omega_q/k_B\ln\{\langle n\rangle^{(1)}\}^{-1}. \tag{21}$$

It was found to be 4.45 K, and the sample temperature rise after the next pump pulse was $\Delta T_{\mathrm{eff}} = 0.25$ K.

The specific heat of the investigated pyrene-doped diphenyl crystal is given by

$$C=[\hbar(\omega_{13}-\omega_{21})N_M]/M\Delta T_{\mathrm{eff}};,$$

where M is the sample mass, ω_{13} and ω_{21} are the frequencies of the $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |1\rangle$ transitions, and N_M is the number of active impurity molecules. For 10^{16} cm^{-3} active molecules per unit volume we get

$$C=1.15\ \mathrm{kgf}^{-1}\cdot\mathrm{J/deg}.$$

5. CONCLUSION

We have demonstrated experimentally and theoretically that, depending on the pump-pulse repetition frequency, the formation of SR in doped crystals varies from pulse to pulse. The SR method was used to determine the effective local-heating temperature and the increase of this temperature from pulse to pulse in a pyrene-doped diphenyl crystal. Since the optical Dicke super-radiance method is developing into an effective coherent-spectroscopy method,[2,4,5] and offers promising technical applications,[2] our investigations are quite timely. To investigate the role of lattice and pseudolocalized phonons in the evolution of the SR, we have used for the first time the nonequilibrium-statistical-operator method, and determined from an analysis of the experimental data the velocity of pseudolocalized phonons. Finally, these investigations, with pyrene-doped diphenyl as an example, have shown that the SR method can be used to determine so important a parameter as the specific heat of a crystal at low temperatures.

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