Dicke superradiance in the field of a coherent phonon wave

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The paper examines superradiance in impurity crystals in the field of a coherent phonon wave excited by two ultrashort laser pulses via Raman scattering processes at the moment of preparation of the initial state of an ensemble of emitters. It is shown that by varying the power of the excitation pulses and their mutual direction of propagation, one can control the superradiance parameters and extract data on the electron–phonon coupling constant and its anisotropy. © *1996 American Institute of Physics.* © *1996 American Institute of Physics.* [S1063-7761(96)01507-7]

1. INTRODUCTION

The problem of spontaneous coherent emission (Dicke superradiance[1]) in impurity crystals taking into account the interaction of emitters, impurity particles, and thermal phonons, has been studied by a number of researchers.[2–7] Weak emitter–phonon coupling generally leads to a temperature-dependent decrease in the superradiance intensity.[5,6] But with strong adiabatic electron–phonon coupling, for which the energy spectrum of the system acquires a set of electron–vibrational levels consisting of a zero-phonon line and vibrational repetitions, there is a finite probability of emission of superradiance pulses not only in the zero-phonon transition but also in the vibronic transitions, with the intensity of these pulses, their delay times, and durations strongly depending on the electron–phonon coupling parameter and the temperature of the sample (whose dimensions are smaller than the radiation wavelength).[2,7] Similar results were obtained in Refs. 3 and 4 using the Thomson model,[8] which describes Brillouin scattering of light in a resonant medium with the participation of a single absorbed (or emitted) phonon.

Experimentally, superradiance on electron–vibrational transitions of O_2^- molecules was realized in the polar dielectrics KCl:O_2^- (see Refs. 9 and 10). Naboĭkin *et al.*[5] and Andrianov *et al.*[6] studied Dicke superradiance in mixed molecular crystals of diphenyl with pyrene on pyrene centers, which typically display weak electron–phonon coupling.

2. STATEMENT OF THE PROBLEM

Obviously, the parameters of superradiance can be effectively controlled by exciting a coherent phonon wave, say by a time-dependent Raman process, instead of exciting thermal phonons, which because of their insignificant population are ineffective at low temperatures. In this case the experimental setup changes somewhat: to prepare the initial superradiance state at time $t=0$ there must be, in addition to an ultrashort laser pulse of frequency ω_1, a second ultrashort pulse of frequency ω_2 propagating at an angle to the first. Then, in addition to optical excitation of impurity centers in the sample at frequencies ω_1 and ω_2, a coherent phonon wave is excited at the difference frequency $\omega_1-\omega_2=\Omega$, and this wave modulates the frequency of the quantum transitions of the emitters (as in the case in which only thermal phonons participate). Changing the direction of propagation of the excitation laser pulses and tuning the difference frequency $\omega_1-\omega_2$ to resonance with different phonon modes, one can not only control the superradiance dynamics but also use the behavior of the characteristic superradiance parameters to determine the features of the electron–phonon coupling: the magnitude of the coupling of localized electron states with the excited phonon modes and the anisotropy of this coupling. Note that the role of a coherent phonon wave in the light echo phenomenon was studied by Wilson *et al.*[11] (see also Ref. 12).

We study an ensemble of optical emitters—impurity atoms or molecules imbedded in a crystal lattice. A natural approach to describing the electron states of the impurity particles in resonant phenomena is to use the idealized scheme of a two-level quantum system.[1] We assume that each such two-level system interacts with the radiation field and the coherent phonon wave (excited in this case by ultrashort laser pump radiation). The interaction of the quantum levels and thermal phonons can be ignored because the phonon modes of the lattice are "frozen" if low temperatures are used in the experimental setup. It is legitimate to study a selectively excited coherent phonon wave by classical means, i.e., by assuming that the wave has an amplitude q, a frequency Ω, and a phase ϕ.

We write the Hamiltonian of the system of optical emitters interacting with the radiation field and the coherent phonon wave as

$$H=H_p+H_{ph}+H_{\text{int}}+V(t), \tag{1}$$

where

$$H_p=\omega_0\sum_j S_j^z, \quad H_{ph}=\sum_{\{k\}}\omega(k)a^+(k)a(k),$$

$$H_{\text{int}}=\sum_{j,\{k\}}[g_j(k)a(k)(S_j^++S_j^-)+\text{H.c.}],$$

$$V(t)=\lambda\sum_j S_j^z\cos(\Omega t+\phi_j).$$

Here $a(k)$ is the quantized amplitude of the radiation field for the mode $(\mathbf{k},s)$ with frequency ω_k and polarization

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$\mathbf{e}_s$, $S_j^{\pm,z}$ is the pseudospin variable, which obeys the commutation relations for angular momentum and describes the jth ($j=1,2,\ldots,N$) two-level atom with energy splitting ω_0, and $g_j(k)$ and λ are the electron–photon and electron–phonon coupling constants:

$$g_j(k)=g(k)\exp(i\mathbf{k}\cdot\mathbf{r}_j)=-i\sqrt{2\pi c|k|/V}(\mathbf{d}\cdot\mathbf{e}_s)\exp(i\mathbf{k}\cdot\mathbf{r}_j),$$

with $\mathbf{d}$ and $\mathbf{r}_j$ the dipole moment and radius vector of the jth particle and V the quantization volume of the radiation field, and ϕ_j the phase of the phonon wave at the point occupied by the jth particle. Throughout this paper we take $\hbar=1$.

3. DERIVATION OF THE MASTER EQUATION

The statistical operator of a system described by the Hamiltonian (1) satisfies the Liouville equation (the evolution of the system is studied over time intervals shorter than the time of irreversible dephasing of the polarization of the resonant medium)

$$i\frac{d\rho}{dt}=[H,\rho], \tag{2}$$

which the canonical transformation

$$\rho\rightarrow\rho'+U^+\rho U, \tag{3}$$

reduces to

$$i\frac{d\rho'}{dt}=[H_{\text{int}}(t),\rho'], \tag{4}$$

where

$$U=\exp\left\{i\int^t dt'[H_p+H_{ph}+V(t')]\right\}, \tag{5}$$

$$H_{\text{int}}(t)=\sum_{j,\{k\}} g_j(k)a(k)\left\{S_j^+\exp\left[i\left(\omega_0 t+\frac{\lambda}{\Omega}\sin(\Omega t+\Phi_j)\right)\right]\right.$$
$$\left.+\text{ H.c.}\right\}\exp[-i\omega(k)t]+\text{ H.c.} \tag{6}$$

To identify the contributions of the harmonics whose frequencies are integral multiples of Ω, it is convenient to expand the exponential function in (6) in a Bessel-function series based on the formula

$$e^{ia\sin x}=\sum_{n=-\infty}^{\infty} J_n(a)e^{inx}.$$

Then

$$H_{\text{int}}(t)=\sum_{j,\{k\},n} g_j(k)J_n\left(\frac{\lambda}{\Omega}\right)a(k)$$
$$\times[S_j^+\exp[i(\omega_0-\omega(k)+n\Omega)t]$$
$$\times\exp(in\phi_j)+\text{ H.c.}]+\text{ H.c.} \tag{7}$$

Equation (4) can be transformed in a standard manner into the integro-differential equation

$$\frac{d\rho'}{dt}+i[H_{\text{int}}(t),\rho'(0)]$$

$$=-\int_0^t[H_{\text{int}}(t),[H_{\text{int}}(t'),\rho'(t')]]dt'. \tag{8}$$

Finding the trace in the photon-field variables of both sides of Eq. (8), we arrive at equations for the reduced density matrix $\sigma=\text{Tr}_{ph}[\rho'(t)]$, which describes the evolution of the quantum states of the impurity subsystem only:

$$\frac{d\sigma}{dt}=-\int_0^t d\tau\,\text{Tr}_{ph}[H_{\text{int}}(t),[H_{\text{int}}(t-\tau),\rho'(t-\tau)]]. \tag{9}$$

Here we have used the fact that $\text{Tr}_{ph}([H_{\text{int}}(t),\rho'(0)])=0$, which follows from the factorization of ρ' at $t=0$: it is assumed that initially the system is prepared in a state with density matrix

$$\rho'(0)=\sigma(0)|0\rangle\langle 0|, \tag{10}$$

where $|0\rangle\langle 0|$ is the density matrix of the radiation field (we may assume that for frequencies in the optical range the radiation field is in the vacuum state $|0\rangle$ with zero temperature).

Since in the case of superradiance the field has the shortest correlation time (proportional to the reciprocal of the optical-frequency bandwidth), in a certain sense the field can be interpreted as a reservoir for the atomic system. The interaction of this system with such a wide-band reservoir rapidly wipes out any memory of the system about its past, with the result that the temporal behavior of the optical emitters is Markovian. This justifies replacing $\rho'(t-\tau)$ by $\rho'(t)$ in (9) and extending the upper limit of the τ-interval to ∞. Moreover, we assume that $\rho'(t)$ can be written as

$$\rho'(t)=\sigma(t)|0\rangle\langle 0|+\Delta\rho',$$

where $\Delta\rho'$ is at least of order H_{int}. Then to second order in H_{int} we obtain a closed equation for the reduced matrix density:

$$\frac{d\sigma}{dt}=-\int_0^\infty d\tau\,\text{Tr}_{ph}[H_{\text{int}}(t),[H_{\text{int}}(t-\tau),\sigma(t)|0\rangle\langle 0|]]. \tag{11}$$

After substituting the expression (7) for $H_{\text{int}}(t)$ into the right-hand side of Eq. (11), expanding commutators, taking the trace, and integrating with respect τ we arrive at

$$\frac{d\sigma}{dt}=\sum_{j,l,\{k\},n,m} J_n\left(\frac{\lambda}{\Omega}\right)J_m\left(\frac{\lambda}{\Omega}\right)$$
$$\times\exp[i(n-m)\Omega t]\exp(in\phi_j)\exp(-im\phi_l)$$
$$\times\Bigg\{g_j^*(k)g_l(k)[S_l^+,S_j^-\sigma]$$
$$\times\left[\frac{iP}{\omega_0-\omega(k)+m\Omega}+\pi\delta[\omega_0-\omega(k)+m\Omega]\right]$$
$$+g_j(k)g_l^*(k)[\sigma S_l^-,S_j^+]$$
$$\times\left[\frac{iP}{\omega_0+\omega(k)+m\Omega}+\pi\delta[\omega_0+\omega(k)+m\Omega]\right]\Bigg\}$$
$$+\text{ H.c.}\equiv-\Lambda(t)\sigma. \tag{12}$$

In the discussion that follows we ignore the principal part (denoted by P), which yields only a small imaginary term, i.e., causes a small shift in the resonance frequency, and the nonresonant terms containing the factor $\delta[\omega_0+\omega(k)+m\Omega]$.

The characteristic time it takes the optical emitters to go into a coherent state is proportional to $\|\Lambda\|^{-1}$ (here $\|\cdots\|$ stands for the operator's value in frequency units) and is much longer than the period $T=2\pi/\Omega$ of the rapid harmonic "jitter" superimposed on the less rapid evolution process of spontaneous coherent emission. Hence it is natural to decompose the real motion, described by the density matrix σ, into slow motion averaged over the period T and rapid "jitter." It is convenient to realize the decomposition procedure by employing the method of "temporal projection" operators:[13]

$$\sigma(t)\to\tilde{\sigma}(t)=\frac{1}{T}\int_0^T dt\sigma(t)\equiv P^t\sigma(t), \tag{13}$$

where the projection operator P^t performs the averaging of rapidly varying quantities over the period T. Next we introduce the operator $Q^t=1-P^t$. Then the decomposition of the density matrix into the slowly and rapidly varying parts can be written as

$$\sigma=P^t\sigma+Q^t\sigma. \tag{14}$$

Successively applying the operators P^t and Q^t to Eq. (12), we arrive at two differential equations instead of one:[13]

$$\frac{dP^t\sigma}{dt}=-P^t\Lambda(t)P^t\sigma-P^t\Lambda(t)Q^t\sigma, \tag{15}$$

$$\frac{dQ^t\sigma}{dt}=-Q^t\Lambda(t)P^t\sigma-Q^t\Lambda(t)Q^t\sigma. \tag{16}$$

On the basis of Eq. (16) we can formally represent the solution for the rapidly varying (oscillating) part of the density matrix in the form

$$Q^t\sigma=-\int^t dt'Q^{t'}\Lambda(t')\tilde{\sigma}(t')-\int^t dt'Q^{t'}\Lambda(t')\sigma(t'). \tag{17}$$

Using (17) in Eq. (15) for the slowly varying part of the density matrix, we see that the order of smallness in Λ of the term $P^t\Lambda Q^t\sigma$ is no less than two (or no less than four in H_{int}), so that the term can be ignored. As a result, averaging Eq. (12) over the "jitter" reduces, according to (15), to averaging the operator $\Lambda(t)$ over the period $2\pi/\Omega$:

$$\frac{d\tilde{\sigma}}{dt}=\pi\sum_{j,l,n}J_n^2\left(\frac{\lambda}{\Omega}\right)\exp[in(\phi_j-\phi_l)]$$
$$\times\sum_{\{k\}}|g(k)|^2\exp[i\mathbf{k}\cdot(\mathbf{r}_j-\mathbf{r}_l)]$$
$$\times\delta(\omega_0-\omega(k)+n\Omega)[S_j^-\tilde{\sigma},S_l^+]+\text{ H.c.} \tag{18}$$

This expression constitutes the most general form of the master equation for superradiance in the presence of a coherent phonon wave. However, to use this equation to obtain results that can be compared with experimental data, we introduce a "coarsening" procedure. The point here is that real samples containing emitters are much larger than the radiation wavelength: $k_0R\gg1$ (here $k_0=\omega/c$ and $R=\max|\mathbf{r}_j-\mathbf{r}_l|$). Hence $\exp[i\mathbf{k}\cdot(\mathbf{r}_j-\mathbf{r}_l)]$ must be averaged over the ensemble of particles. We apply a similar procedure to the factor $\exp[i(\phi_j-\phi_l)]$, assuming that the phase difference $\delta\phi_{jl}=\phi_j-\phi_l$ is uniformly distributed over the interval 0 to 2π with density $1/2\pi$. The result of ensemble averaging,

$$\overline{\exp[i\mathbf{k}(\mathbf{r}_j-\mathbf{r}_l)]}\equiv\Gamma(|\mathbf{k}|),$$

depends on the shape of the sample, while averaging over the phase spread $\delta\phi_{jl}$ results in the exponential function $\exp[in\delta\phi_{jl}]$ being replaced by the Kronecker symbol:

$$\frac{1}{2\pi}\int_0^{2\pi}d\phi_{jl}\exp(in\delta\phi_{jl})=\delta_{n,0}. \tag{19}$$

Allowing for (19) and passing in (18) from the sum over $\{k\}$ to an integral with respect to the wave vectors k and a sum over the polarizations s of the photon field,

$$\sum_{\{k\}}\cdots\to\sum_s\frac{V}{(2\pi)^3}\int d^3k\cdots,$$

we obtain

$$\frac{d\tilde{\sigma}}{dt}=-\gamma\Gamma(k_0)J_0^2\left(\frac{\lambda}{\Omega}\right)\sum_{j,l}(S_j^+S_l^-\tilde{\sigma}-2S_l^-\tilde{\sigma}S_j^+$$
$$+\tilde{\sigma}S_j^+S_l^-), \tag{20}$$

where $\gamma=2|\mathbf{d}|^2\omega^3/3c^3$.

Thus the dependence of n-phonon processes ($n=1,2,\ldots$) on the randomly distributed phase shifts $\delta\phi_{jl}$ (with constant density $1/2\pi$) must lead to averaging their contributions to zero when superradiance is initiated in large samples. The entire modulation effect of a coherent phonon wave on optical centers in this case resides in the factor $J_0^2(\lambda/\Omega)$, which renormalizes the electron–photon coupling constant. This factor is similar to the Debye–Waller factor, which characterizes the reduction in the unshifted component (the zero-phonon line) of an optical transition

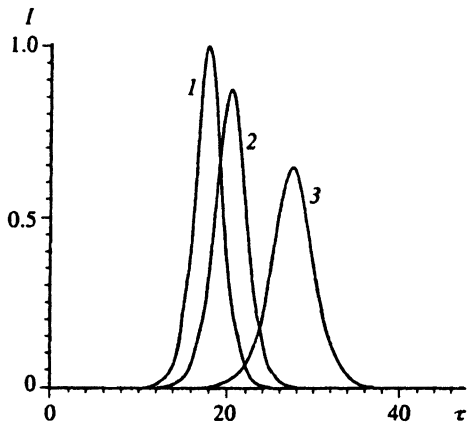

FIG. 1. The intensity I of Dicke superradiance (normalized to $N\omega_0/4\tau_R^{(0)}$) as a function of $\tau=t/\tau_R^{(0)}$ for three values of the energy W of the excitation laser pulses: *1*), $W=0$; *2*), $W=1.25\times10^{-3}$J; and *3*), $W=2.25\times10^{-3}$J ($N=10^{18}$).

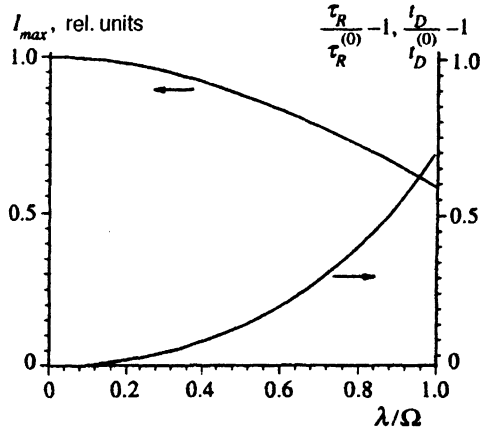

FIG. 2. The peak intensity $I_{\max}$, the delay time t_D, and the superradiance pulse duration τ_R as functions of the electron–phonon coupling constant λ/Ω.

when elastic scattering of thermal phonons is taken into account. However, in contrast to the case of thermal phonons, a coherent phonon wave does not lead to homogeneous dephasing of the polarization of the optical centers.

4. RESULTS AND DISCUSSION

The standard form[14] of the operator part of Eq. (20) makes it possible to immediately write an (approximate) expression for the intensity I of superradiance under conditions in which all emitters are initially completely inverted and the transverse macroscopic polarization is zero, i.e., $\mathrm{Tr}_p[S_j^z\tilde{\sigma}(0)]=\frac{1}{2}$ and $\mathrm{Tr}_p[S_j^+S_l^-\tilde{\sigma}(0)]=0$:

$$I(t)=-\omega_0\sum_j\frac{d}{dt}\mathrm{Tr}_p[S_j^z\tilde{\sigma}(t)]$$

$$=\frac{N\omega_0}{4\tau_R}\mathrm{sech}^2\left(\frac{t-t_D}{2\tau_R}\right), \qquad (21)$$

where the renormalized delay time t_D and duration τ_R of the superradiance pulse are defined as

$$\tau_R=\tau_R^{(0)}J_0^{-2}\left(\frac{\lambda}{\Omega}\right), \quad t_D=\tau_R\ln N=t_D^{(0)}J_0^{-2}\left(\frac{\lambda}{\Omega}\right) \qquad (22)$$

(here $\tau_R^{(0)}=[\gamma\Gamma(k_0)N]^{-1}$ and $t_D^{(0)}$ are the values of the parameters in the absence of a coherent phonon wave).

The renormalization factor $J_0^2(\lambda/\Omega)$, which enters into the expressions for t_D, τ_D, and I because of the modulation of the quantum states of the emitters by a coherent phonon wave, leads (since $J_0^2(\lambda/\Omega)<1$ for $\lambda/\Omega\neq0$) to an increase in the time of formation of a superradiance pulse, the spreading of the pulse, and a decrease in the peak intensity. The advantage of using a coherent phonon wave is that not only does it ensure effective control over the superradiance process but it also allows extracting information about the strength of the coupling of the electron states of the emitters and the various phonon modes thanks to the possibility of selectively exciting these modes (and simultaneous "freezing" of the thermal phonons caused by low temperatures). These facts are illustrated by Fig. 1, which depicts the time dependence of the intensity I for different values of the total energy W of the excitation pulses. Here we have allowed for the fact that the dimensionless parameter λ/Ω can be written as[11] $\lambda/\Omega=q_0\sqrt{2Sm\Omega/\hbar}$, where $q_0=AW$ is the phonon amplitude, S is the Huang–Rees factor, m is the phonon-mode reduced mass, A is a constant (for the a_{1g}-mode of naphthalene, $\Omega=1385\,\mathrm{cm}^{-1}$, $S=0.01$, $A=0.3\times10^{-13}\mathrm{dyn}^{-1}$, and $m\simeq10^3m_p$, with m_p the proton mass). Figure 2 illustrates the dependence of a superradiance pulse on the electron–phonon coupling constant λ/Ω (for a fixed value of W). Obviously, in the latter case it is possible to study the anisotropy of the electron–phonon coupling by varying the direction of propagation of the phonon wave (i.e., by changing the directions in which the excitation laser pulses are applied).

[1] É. A. Manykin and V. V. Samartsev, *Optical Echo Spectroscopy* [in Russian], Nauka, Moscow (1984).
[2] A. P. Saĭko, Dokl. Akad. Nauk Belarus SSR, **25**, 1077 (1981).
[3] E. K. Bashkirov, E. M. Sorokina, Fam Le Kien, and A. S. Shumovskiĭ, Kratk. Soobshchen. OIYaI (JINR), No 2, 8 (1984).
[4] N. N. Bogolyubov, Jr., E. K. Bashkirov, Fam Le Kien, and A S Shumovskiĭ, *Preprint P-17-85-938*, Dubna, JINR (1985).
[5] Yu. V. Naboĭkin, S. A. Andrianov, P. V. Zinov'eva *et al.*, Zh. Éksp. Teor. Fiz. **89**, 1146 (1985) [Sov. Phys. JETP **62**, 660 (1985)].
[6] S. A. Andrianov, P. V. Zinov'eva, Yu. V. Malyukin, Yu. V. Naboĭkin, V. V. Samartsev, N. B. Silaeva, and Yu. E. Sheĭbut, Zh. Éksp. Teor. Fiz. **91**, 1990 (1986) [Sov. Phys. JETP **64**, 1180 (1986)].
[7] A. P. Saĭko and V. S. Kuz'min, Dokl. Akad. Nauk Belarus SSR, **34**, 998 (1990).
[8] B. V. Thomson, J. Phys. C **3**, L147 (1970).
[9] R. Florian, L. O. Schwan, and D. Schmid, Solid State Commun. **42**, 55 (1982).
[10] R. Florian, L. O. Schwan, and D. Schmid, Phys. Rev. A **29**, 2709 (1984).
[11] W. L. Wilson, G. Wackerle, and M. D. Fayer, J. Chem. Phys. **88**, 3407 (1988).
[12] A. P. Saĭko and V. S. Kuz'min, Teoret. Mat. Fiz. **83**, 291 (1990).
[13] A. P. Saĭko, Fiz. Tverd. Tela (St. Petersburg) **35**, 38 (1993) [Phys. Solid State **35**, 20 (1993)].
[14] G. S. Agarwal, *Quantum Statistical Theories of Spontaneous Emission and Their Relation to Other Approaches*, Springer-Verlag, Berlin (1974).

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