Elementary excitations and Raman scattering in a system of nonequilibrium phonons

S. A. Bulgadaev and I. B. Levinson

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
(Submitted July 18, 1974)
Zh. Eksp. Teor. Fiz. 67, 2341–2351 (December 1974)

Renormalization of the long-wave optical phonon spectrum due to the strongly nonequilibrium quasimonochromatic distribution of acoustic phonons with a frequency equal to half that of the optical phonon is studied. It is shown that long-lived long-wave collective excitations are present in such systems of acoustic phonons, and their frequencies are close to that of the optical phonon. These elementary excitations are manifest in the optical Raman scattering spectrum and lead to a complex structure of the Stokes scattering line.

INTRODUCTION

With a high degree of probability, it can be assumed as experimentally proven that the creation of distributions of nonequilibrium short-wave acoustic phonons with a small frequency spread $\Delta\omega$ about some central frequency ω_0 is possible.[1,2] Such phonons originate in the decay of long-wave optical phonons,[3,4] which are generated by light (usually by beats of two laser beams with frequencies ν_1 and ν_2, such that $\nu_1 - \nu_2$ is close to the frequency Ω_0 of optical phonons with $k = 0$.[1, 5-7]).

It follows from the law of momentum conservation that an optical phonon with $\mathbf{k} = 0$ decays into two acoustic phonons with opposite momenta $\mathbf{q}$ and $-\mathbf{q}$; therefore, the energies of the resultant acoustic phonons are identical and are completely determined by the law of energy conservation: $\omega_q = \Omega_0/2$. Thus the spread $\Delta\omega$ about $\omega_0 = \Omega_0/2$ is determined only by the value of the wave vector of the optical photon $k \neq 0$ or by an indeterminacy in the law of energy conservation. For parallel light beams, the momentum of the excited optical phonon is $k \sim \Omega_0/c$, so that $\Delta\omega \sim \Omega_0 s/c$, where s is the sound velocity and c the light velocity in the medium. The indeterminacy in the law of energy conservation gives, as has been shown previously,[8] $\Delta\omega \sim \Gamma_0$ or $\Delta\omega \sim \Delta\nu$, where Γ_0 is the width of the spontaneous decay of the optical phonon with $k = 0$, and $\Delta\nu$ is the spectral width of the laser beams. Typical values are $\Omega_0 \sim 1000\ \mathrm{cm}^{-1}$, $\Gamma_0 \sim 1\ \mathrm{cm}^{-1}$, $s/c \sim 10^{-4}$, and $\Delta\nu \lesssim 1\ \mathrm{cm}^{-1}$, so that the distribution of acoustic phonons is highly monochromatic in any case: $\Delta\omega/\omega_0 \sim 10^{-3}$. For the powers used experimentally,[1] the occupation numbers of the acoustic phonons reached $N_q \sim 1$.

It is natural to raise the question as to how the presence of the distribution N_q renormalizes the spectrum of the long-wave optical phonons.[9] It is convenient here to have the following picture in mind. Let there be two branches of optical phonons with close or identical frequencies at $k = 0$. One branch is pumped by the light and acoustic phonons are created. The second (control) branch is not excited by the light directly, but interacts with the created acoustic phonons. It can be verified that in such a situation the distribution N_q does not depend on the presence of the control branch and therefore can be considered as given by the conditions of excitation of the pumped branch. On the other hand, the presence of the distribution N_q leads to a renormalization of the spectrum of the control branch, which can be studied, for example, through Raman scattering or absorption of light of low intensity by the phonons of this branch.

In this paper it will be shown that the renormalization of the spectrum of long-wave optical phonons, brought about by the quasi-monochromatic distribution of the acoustic phonons, is qualitatively different from the renormalization in the ordinary case of a broad distribution ($\Delta\omega \gg \Gamma$). There are two such qualitative differences. First, a new branch of elementary excitations of the collective type appears with frequency Ω_0 and lifetime of the order of $(\Delta\omega)^{-1}$. For sufficiently large N_q this branch mixes with the bare branch of the optical phonon. Second, it turns out that sufficiently narrow distributions of acoustic phonons ($\Delta\omega < \Gamma_0/2$) do not stimulate the decay of optical phonons, as follows from the usual representations based on the kinetic equations for the occupation numbers, but impede it. The qualitative renormalization of the spectrum involves a qualitative change in the character of the Raman scattering by the control branch. The basic effect here is the splitting of the Stokes lines into a triplet at sufficiently large N_q.

1. DISTRIBUTION OF ACOUSTIC PHONONS

The distribution of acoustic phonons that arises in the decay of the optical phonons has been calculated previously.[8] It was assumed there that there is a single optical branch O and a single acoustic branch A. By a single acoustic branch is meant the transverse TA branch whose short-wave phonons have a long lifetime $\tau \gg \Gamma_0^{-1}$ [10]. Actually, the optical phonons decay not only into transverse acoustic phonons, but also into longitudinal ones (LA). However, the number of LA phonons that appear is small in comparison with the number of TA phonons, because there is a much shorter lifetime for the LA phonons and the O $\to$ 2TA and O $\to$ 2LA decay probabilities are of the same order of magnitude. The decay into longitudinal phonons decreases the population of the transverse branch of interest to us; however, the corresponding "branching factor" is of the order of unity, since the decay probabilities are of the same order.

If there exists a decay O $\to$ TA + LA, then this leads to the generation of TA phonons with frequencies near $\omega_0 \neq \Omega_0/2$. Such a distribution of phonons renormalizes the spectrum of optical phonons into a nonresonant and much weaker one, and therefore does not have to be taken into account.

In order to use the results of the previous research,[8] it must be shown that the distribution of acoustic phonons does not depend on the presence of a control branch of optical phonons. This actually occurs for those pump-power restrictions assumed in[8].

We write down equations of the type of (1.14) from[8] for both optical branches; the pumping branch (index 1) and the control (index 2), neglecting, in accord with the assumption $\tau^{-1} \ll \Gamma_0$, the widths of the acoustic phonons:

$$\frac{1}{2}A_1\int d^3q\,\delta(\Omega-\omega_{\mathbf{q}}-\omega_{\mathbf{k-q}})\,[N_{\mathbf{q}}N_{\mathbf{k-q}}-n_1(k)\,(N_{\mathbf{q}}+N_{\mathbf{k-q}}+1)\,]=-G(k), \tag{1.1}$$

$$\frac{1}{2}A_2\int d^3q\,\delta(\Omega-\omega_{\mathbf{q}}-\omega_{\mathbf{k-q}})\,[N_{\mathbf{q}}N_{\mathbf{k-q}}-n_2(k)\,(N_{\mathbf{q}}+N_{\mathbf{k-q}}+1)\,]=0. \tag{1.2}$$

Here G(k) describes the pumping of branch 1 by light, $n(k) \equiv n(k\Omega)$ is the occupation number of the optical modes, and A_1 and A_2 are the corresponding constants of the decay $O \to 2A$. It was shown in[8] that the term with $N_q N_{k-q}$ is small in comparison with the term which contains G. If we assume that $A_1 \sim A_2$, it then follows directly that $n_2(k) \ll n_1(k)$. Thanks to this inequality we can neglect in the balance equation of acoustic phonons [(1.11) of[8]] the contribution from the control branch, and the distribution N_q thus turns out to be independent of the presence of the control branch.

It was similarly shown that if the pumping has a Lorentzian spectral shape, then the distribution N_q also has a Lorentzian shape for both strong and weak pumping. For simplicity, we assume that the pumping is exactly resonant, i.e., $\omega_0 = \Omega_0/2$, and that the shape of N_q is always Lorentzian:

$$N_{\mathbf{q}}=N_0\frac{(\Delta\omega/2)^2}{(\omega_{\mathbf{q}}-\omega_0)^2+(\Delta\omega/2)^2}. \tag{1.3}$$

We can hope that the latter assumption has no strong effect on the qualitative character of the subsequent results.

In what follows, we shall not consider the pumped branch, assuming that its role reduces to the regularization of the parameters N_0 and $\Delta\omega$. For weak pumping, when $N_0 \ll 1$, the quantity $\Delta\omega$ is determined by the smaller of the values of $\Delta\nu$ and Γ_0; for strong pumping, when $N_0 \gg 1$, we always have $\Delta\omega \sim \Delta\nu$.[8]

2. SPECTRUM OF THE ELEMENTARY EXCITATIONS

The spectrum of the long-wave elementary excitations is determined by the retarded Green's function G_r of the optical phonons with k = 0. The polarization operator $P_r(\Omega)$, which corresponds to this Green's function, can be computed with the help of Eqs. (2.1) and (2.2) from[8], where, we substitute for N_q the distribution (1.3). Near the bare poles $\pm\,\Omega_0$ the polarization operator can be computed in explicit form:

$$P_r(\Omega)=\frac{\Gamma_0}{\Omega_0}\left(\pm i+4N_0\frac{\Omega_0\Delta\omega}{\Omega^2-\Omega_0^2\mp 2i\Omega_0\Delta\omega}\right), \tag{2.1}$$

where the upper and lower signs correspond to the cases $\operatorname{Im}\Omega^2 < 0$ and $\operatorname{Im}\Omega^2 > 0$. The Green's function

$$G_r(\Omega)=\Omega_0^2[\Omega^2-\Omega_0^2-\Omega_0^2P_r(\Omega)\,]^{-1} \tag{2.2}$$

has poles when analytically continued through the cut $\Omega^2 > 0$ from each of its edges; the corresponding contributions are complex conjugates.

We now give the equation for the determination of the poles near $+\Omega_0$:

$$[\Omega-(\Omega_0+i\Gamma_0/2)\,]\,[\Omega-(\Omega_0+i\Delta\omega)\,]-N_0\Gamma_0\Delta\omega=0. \tag{2.3}$$

This equation recalls the equation for the mixing of two branches, and the coupling coefficient is proportional to N_0. Its solution gives rise to the critical value

$$N_0^*=(\alpha-1)^2/8\alpha, \quad \alpha=2\Delta\omega/\Gamma_0. \tag{2.4}$$

If we introduce $\xi = N_0/N_0^*$, we can write the roots of (2.3) in the form

$$\xi<1:\quad \operatorname{Re}\Omega_{1,2}=\Omega_0,\quad \operatorname{Im}\Omega_{1,2}=\frac{1}{4}\Gamma_0[\,(\alpha+1)\mp(\alpha-1)\sqrt{1-\xi}\,],$$

$$\xi>1:\quad \operatorname{Re}\Omega_{1,2}=\Omega_0\pm\frac{1}{4}\Gamma_0(\alpha-1)\sqrt{\xi-1},\quad \operatorname{Im}\Omega_{1,2}=\frac{1}{4}\Gamma_0(\alpha+1). \tag{2.5}$$

It is also instructive to write out the corresponding residues:

$$\begin{aligned} \xi<1:&\quad \operatorname{Res}G_r(\Omega_{1,2})=\Omega_0\frac{\xi-2\mp2\sqrt{1-\xi}}{2\xi-2\mp2\sqrt{1-\xi}},\\ \xi>1:&\quad \operatorname{Res}G_r(\Omega_{1,2})=\Omega_0\frac{\xi-2\pm2i\sqrt{\xi-1}}{2\xi-2\pm2i\sqrt{\xi-1}}. \end{aligned} \tag{2.6}$$

The motion of the poles with increase in ξ in the complex plane is shown in Fig. 1. The merging of the poles corresponds to $\xi = 1$. For small N_0, one of the poles (Ω_1, upper sign) describes an optical phonon with spontaneous decay, while the second pole (Ω_2, lower sign) describes a new long-wave elementary excitation in the system of phonons with frequency Ω_0 and lifetime $1/2\Delta\omega$. It is obvious that this elementary excitation is long-lived only for quasimonochromatic distributions of the acoustic phonons.

The new elementary excitation can be interpreted as a collective excitation in the system of acoustic phonons, since the corresponding pole is also present in the two-particle Green's function of the acoustic phonons. Here the optical phonons play the role of intermediate particles in the interaction between the acoustic phonons. The simplest vertex corresponding to this case is proportional to Γ_0. If $\Gamma_0 \to 0$, then the direct interaction between acoustic phonons is cut off. At the same time, the location of the new pole remains completely determined: $\Omega_2 \to \Omega_0 + i\Delta\omega$. In this case the situation very much recalls zero sound (if we disregard the fact that the law of dispersion does not have an acoustic character).

When N_0 increases, remaining less than the critical value N_0^*, only the widths of the elementary excitations change, and the frequencies are identical and equal to Ω_0. Here the widths come closer together, approaching the mean arithmetic value of the bare values of Ω^*, which is achieved at $N_0 = N_0^*$. In the subsequent increase in N_0, the widths of the excitations do not change; however, the frequencies of the excitations change—they lie symmetrically on opposite sides of Ω_0 and the splitting of the frequencies increases with increase of N_0. It is obvious that the origin of the elementary excitations is lost in the region $N_0 \geq N_0^*$.

3. RAMAN SCATTERING

The complicated structure of the spectrum of elementary excitations with k = 0 can be discovered with the help of Raman scattering of the weak light signal which does not create an additional non-equilibrium phonon distribution. For this purpose, it is necessary to study the shape of the lines of Raman scattering with frequency shift $\nu - \nu' \equiv \tilde{\nu} \approx \pm\,\Omega_0$. However, it must be kept in mind that usually the characteristic momentum k_0 at which the spectrum differs materially from the spectrum at

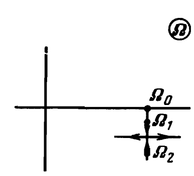

FIG. 1

$k = 0$ is the dimension of the Brillouin zone, i.e., $k_0 \approx \Omega_0/s$. This momentum is always greater than the momentum transfer $\bar{f} = f - f'$ in the scattering of light from f into f'. Therefore the scattering of light at any angle usually probes the spectrum at $k = 0$. If the same spectrum is renormalized because of the interaction with the narrow acoustic distribution, then $k_0 \sim \Delta\omega/s$, which is comparable with $f \sim \nu/c$ in the scattering of visible light through $\pi/2$. Therefore, forward scattering is preferable for probing the spectrum at $k = 0$, when $f \sim \Omega_0/c$ and can be made smaller than $k_0 \sim \Delta\omega/s$.

Assuming that one can set $k = 0$, we find the following expression for the probability of scattering into the complete solid angle and in the frequency range $d\nu' = d\bar{\nu}$ in the Stokes region $\bar{\nu} \approx +\Omega_0$:

$$w_s(\bar{\nu})\,d\bar{\nu} = \frac{w_0}{2\pi\Omega_0}\{[n(\Omega)+1]\operatorname{Im} G_r(\Omega)\}_{\Omega=\bar{\nu}}. \tag{3.1}$$

and in the anti-Stokes region $\bar{\nu} \approx -\Omega_0$:

$$w_A(\bar{\nu})\,d\bar{\nu} = -\frac{w_0}{2\pi\Omega_0}\{n(\Omega)\operatorname{Im} G_r(\Omega)\}_{\Omega=-\bar{\nu}}. \tag{3.2}$$

Here w_0 is the total probability of spontaneous Raman scattering; $n(\Omega)$ is the occupation number of the $n(k)$ state with $k = 0$. The presence of $n(\Omega)$ reflects the fact that the given stationary distribution N_q not only renormalizes the spectrum but also leads to a certain filling of the states due to the sticking of the acoustic phonons to the optical ones.

It follows from (1.2) at $k = 0$ that

$$n(\Omega) = \{N_q^2/(2N_q+1)\}_{\omega_q=\Omega_0/2}, \tag{3.3}$$

$$n(\Omega)+1 = \{(N_q+1)^2/(2N_q+1)\}_{\omega_q=\Omega_0/2}. \tag{3.4}$$

The characteristic width of these functions is $\Delta\omega$. For a wide distribution ($\Delta\omega \gg \Gamma_0$ and $N_0 \ll \Delta\omega/\Gamma_0$), it follows from the results of the previous discussion that $\operatorname{Im} G_r(\Omega)$ is a Lorentzian peak of width $\Gamma_0(2N_0 + 1)$, centered about Ω_0. Within the limits of its width, we can neglect the change in the functions (3.3) and (3.4) and set $\Omega = \Omega_0$. Here $n(\Omega_0)$ becomes the ordinary occupation number n_0 of the state $k = 0$. It is thus seen that for broad distributions, (3.1) and (3.2) transform into ordinary formulas for Raman scattering with account of line broadening. We note that there is no frequency shift because of the symmetry of the distribution N_q relative to $\Omega_0/2$.

For narrow distributions $\Delta\omega \lesssim \Gamma$ the situation changes radically—the shape of the line is determined not only by the Green's functions, but also by the filling factors (3.3) and (3.4). For convenience, we reckon $\tilde{\nu}$ from the "normal" center of the line $\bar{\nu} = \pm\Omega_0$: $\tilde{\nu} = \bar{\nu} - \Omega_0$ for the Stokes line and $-\tilde{\nu} = \bar{\nu} + \Omega_0$ for the anti-stokes line.

We consider the limiting case in which the effects associated with the narrowness of the distribution of acoustic phonons are more clearly expressed:

$$\Delta\omega \ll \Gamma_0, \quad N_0 \gg N_0^* = \Gamma_0/8\Delta\omega. \tag{3.5}$$

Here

$$\Omega_{1,2} = \Omega_0 \mp \Gamma_0\sqrt{\xi}/4 + i\Gamma_0/4, \tag{3.6}$$

i.e., the splitting of the frequencies of the elementary excitations is greater than their width. This means that $\operatorname{Im} G_r(\Omega)$ has the shape of two isolated peaks corresponding to $\tilde{\nu} = \pm\Gamma_0\sqrt{\xi}/4$.

We now consider the frequency dependence of the filling factors. It is easy to see that $n(\Omega)$ gives a peak of width $\Delta\omega$ about $\tilde{\nu} = 0$. Thus, we can generally expect the appearance of three peaks in $w(\tilde{\nu})$. In order to determine which of them appear, we must estimate their heights. In the region of the peaks $\operatorname{Im} G_r(\Omega)$, we have, for $\tilde{\nu} \sim \Gamma_0\sqrt{\xi}$,

$$\operatorname{Im} G_r \sim \Gamma_0^{-1}, \quad N_q \sim \alpha \ll 1, \quad n \sim \alpha^2, \quad n+1 \approx 1. \tag{3.7}$$

In the region of the peak $n(\Omega)$, for $\tilde{\nu} \lesssim \Delta\omega$, we have

$$\operatorname{Im} G_r \sim (\Gamma_0\xi)^{-1}, \quad N_q \sim N_0 \gg 1, \quad n \approx n+1 \sim N_0. \tag{3.8}$$

It follows from these estimates that there is a central peak in the Stokes region for $\tilde{\nu} = 0$ of width $\Delta\omega$ and height of the order of $(\Delta\omega)^{-1}$ and two lateral peaks at $\tilde{\nu} = \pm\Gamma_0\sqrt{\xi}/4$ of width $\Gamma_0/2$ and height of the order of $1/\Gamma_0$. The central peak is higher and narrower, the lateral ones are lower and wider, but the areas of all peaks are of the same order. In the anti-Stokes region, the location and widths of the peaks are the same; however, the height of the lateral peaks decreases by a factor of α^2 in comparison with the height of the central peak, which is the same as in the Stokes region.

4. THE GEOMETRY OF THE PUMPED AND CONTROL BRANCHES IN CRYSTALS OF THE DIAMOND TYPE

A situation with two branches of optical phonons (pumped and contro) can be realized by using, for example, the presence of different polarizations of the optical phonons. We consider a crystal with diamond symmetry, in which the interaction of the light with the optical phonons is described by the following Hamiltonian:

$$E_xE_yw_z + E_yE_zw_x + E_zE_xw_y, \tag{4.1}$$

where **E** is the field of the light wave and **w** is the relative shift of the sublattices. Let the pump fields $\mathbf{E}_1$ and $\mathbf{E}_2$ be polarized in the xy plane. Then only the component w_z is directly pumped by the light and the components w_x and w_y can be regarded as controls.

With such an approach, the question still arises as to the diagonality of the Green's function of the optical phonon relative to the polarizations. If there is no such diagonality, then it is not possible to consider the different polarizations independently. When pumping is absent, then diagonality follows from the cubic symmetry O_h, since **w** is transformed according to F_{2g}, and the symmetrized product $\{F_{2g} \times F_{2g}\}$ contains the trivial representation A_{1g} only once. In the presence of pumping, the symmetry is reduced and the appearance of nondiagonal components is possible in principle. We shall show that this does not occur if the pump fields E_1 and E_2 lie in the xy plane. In this case, the excitation of the phonons is determined by the quantity

$$E_xE_y = (E_{1x}+E_{2x})(E_{1y}+E_{2y}) = E_{1x}E_{2y} + E_{2x}E_{1y}. \tag{4.2}$$

Here we keep only the terms that are important for pumping, with the frequency difference $\nu_1 - \nu_2$. In fact (see [8]), if the beams consist of a large number of longitudinal modes with random phases, then the excitation is determined by the square of the quantity (4.2). It is easy to prove that this square is invariant to a transformation of the subgroup D_{4h} with the z axis. Therefore the system of phonons has the symmetry type D_{4h} in the considered excitation. Upon the reduction $O_h \to D_h$, we have

$$F_{2g} \to E_g + B_{2g}, \quad w_x,\ w_y \sim E_g, \quad w_z \sim B_{2g}. \tag{4.3}$$

Further, $E_g \times B_{2g}$ does not contain A_{1g}; therefore, there are no Green's functions connecting the pumped polarization w_z with the control polarizations w_x and w_y. The product $\{E_g \times E_g\}$ contains A_{1g} once; therefore there exists just one Green's function of the control phonons. Thus the considerations advanced show that in the chosen pump geometry we can consider two independent branches–pumping and control, which justifies the model used above.

In the process of probing the control branch, we must use a polarization that would not be scattered by the pumped phonons w_z, and observe a scattered-light polarization in which the pump beam is not scattered. This can be achieved, for example, in the case in which the pumps $\mathbf{E}_1$ and $\mathbf{E}_2$ are polarized along (110), the probing field $\mathbf{E}_0$ along (001), and the scattered field $\mathbf{E}'$ along $(1\bar{1}0)$.

5. DECAY OF OPTICAL PHONONS

In connection with experiments on the direct measurement of the lifetimes of the nonequilibrium optical phonons,[7, 11, 12] it is interesting to consider the nonequilibrium aspect of the problem, i.e., to ascertain how the presence of a narrow fixed distribution of acoustical phonons affects the decay of a given initial distribution of optical phonons. It should first be stipulated that such a statement of the problem differs materially from the experimental situation, where the distribution of the acoustic phonons is not fixed by the external conditions, but arises in the process of decay of the optical phonons. However, it can be hoped that some qualitative aspects are preserved in the assumed setup.

There exists an opinion that the diagram technique developed by Keldysh[13] is applicable only to the solution of problems in which the initial conditions are forgotten. In fact, as can be seen from analysis of this work, the conditions of applicability of the technique are much broader: it is sufficient that a moment of time exist at which the state of the system is such that Wick's theorem holds. If this moment is taken to be the initial moment, then the technique allows us to investigate the nonstationary development of the system from this moment. It is easy to see that for a phonon system, Wick's theorem holds if the different phonon modes are uncorrelated and each of them is in a state of "thermodynamic equilibrium" with an arbitrary temperature. The arbitrariness of the temperature means that the degree of excitation of the different modes is arbitrary and the state of the entire system is nonequilibrium. Thus there always exists a broad class of nonequilibrium states, the relaxation of which can be studied with the help of the Keldysh diagram technique.

On the basis of the foregoing, we solve the following problem: The state of the acoustic phonons is given and is determined by the distribution (1.3). At t = 0, we excite only the optical mode with k = 0 with the occupation number $n_0(0)$ and seek the occupation number n(t) of this mode at the subsequent instant. The set of equations for the Green's function of the optical mode with k = 0 can be written down with the aid of the Keldysh technique in the time representation. It is merely necessary to be integrated with respect to time not between $t = -\infty$ and $t = +\infty$, but between $t = 0$ and $t = +\infty$.

It is convenient to use the retarded function

$$G_r(x, x') = -i\theta(t-t')\langle[\psi(x), \psi(x')]\rangle \tag{5.1}$$

and the statistical function

$$G_s(x, x') = -i\langle\{\psi(x), \psi(x')\}\rangle, \tag{5.2}$$

where [. . .] and {. . . } denote the commutator and the anticommutator. Similar functions D_r and D_s for acoustic phonons are in fact given by Eqs. (1.1) and (1.2) of[8]. We then have

$$\hat{G}_0^{-1}(t) G_r(t, t') = \delta(t-t') + \int_0^\infty d\bar{t}\, P_r(t-\bar{t}) G_r(\bar{t}, t'), \tag{5.3}$$

$$\hat{G}_0^{-1}(t) G_s(t, t') = \int_0^\infty d\bar{t}\, P_r(t-\bar{t}) G_s(\bar{t}, t') + \int_0^\infty d\bar{t}\, P_s(t-\bar{t}) G_r(t', \bar{t}). \tag{5.4}$$

Here

$$\hat{G}_0^{-1}(t) = -(\Omega_0^{-2}\, \partial^2/\partial t^2 + 1). \tag{5.5}$$

Further, $P_r(t)$ is the time representation of the polarization operator $P_r(\Omega)$, which arises when we consider the spectrum

$$P_r(t) = ia^2 \int \frac{d^3q}{(2\pi)^3} D_r(\mathbf{q}, t) D_s(\mathbf{q}, t), \tag{5.6}$$

$P_s(t)$ is obtained from the same formula by the substitution $D_rD_s \to (D_rD_r + D_aD_a + D_sD_s)/2$. The coupling constant a^2 is proportional to Γ_0. From the equations we get

$$G_s(t, t) = -i\Omega_0[2n_0(t) + 1].$$

From (5.3) it is immediately seen that G_r depends only on the time difference and is the Fourier transform of (2.2). Substituting the thus-found G_r in (5.4), we obtain an inhomogeneous equation for G_s with the same operator as for G_r, and therefore G_s can be expressed in terms of G_r. Omitting the details of the solution, we write down the answer

$$n_0(t) = n_0(0) f(t) + s(t). \tag{5.7}$$

The first term describes the relaxation of the initial excitation of the mode k = 0 due to decay of the optical phonon into two acoustic phonons, and the second describes the excitation of this mode due to merging of two phonons from the given distribution into a single optical phonon. This term is not of interest to us and therefore we shall not write it down in explicit form.

The decay law takes the form

$$f(t) = \Omega_0^{-2}[G_r^2(t) + \Omega_0^{-2}(\partial_t G_r(t))^2]. \tag{5.8}$$

If we write

$$G_r(t) = -\Omega_0 A(t) \sin(\Omega_0 t + \varphi(t)), \quad t > 0, \tag{5.9}$$

then, in the absence of decay we have A(t) = 1 and $\varphi(t) = 0$, and after turning on the decay A(t) and $\varphi(t)$ are slowly changing functions of time. It is then seen that the decay law is

$$f(t) = A^2(t), \tag{5.10}$$

i.e., it is determined by the slowly-varying factor in $G_r(t)$. In the spectral representation, this means that the rate of change of f(t) is determined by the shift of the poles of $G_r(\Omega)$ from $\pm\Omega_0$.

We first consider the case of a broad distribution, $\Delta\omega \gg \Gamma_0$. Then, for $\xi \ll 1$, the decay law f(r) is determined by the pole Ω_1 for which we obtain $\mathrm{Im}\,\Omega_1 = \Gamma_0(2N_0 + 1)/2$. This corresponds to the rate of induced decay obtained from the kinetic equations for the occupation numbers. As is now seen, the criterion for the applicability of this result is not only $\Delta\omega \gg \Gamma_0$ but also $N_0 \ll \Delta\omega/\Gamma_0$.

The rate of induced decay of the phonons is larger than the rate of spontaneous decay—the broad distribution of acoustic phonons stimulates the decay. It is seen from the location of the poles that this property is preserved even for narrow distributions with $\Delta\omega > \Gamma_0/2$. However, for $\Delta\omega < \Gamma_0/2$, the situation changes; the increase in N_0 leads to the result that the imaginary parts of both poles turns out to be less than $\Gamma_0/2$ and therefore $f(t)$ will decay less rapidly than $\exp(-\Gamma_0 t)$. This means that such a distribution of the acoustic phonons delays the decay. For very narrow distributions, $\Delta\omega \ll \Gamma_0$, an increase of N_0 to the critical value $N_0^* = \Gamma_0/8\Delta\omega$ leads to a decrease in the rate of decay by a factor of two: $\Gamma^* = \Gamma_0/2$.

In order to connect these results with the experiments of[11, 12], we estimate N_0 in these experiments. By assuming the concentration of acoustic phonons to be of the order of the concentration $\tilde{n}$ of optical phonons and that $\Delta\omega \sim \Gamma_0$, we find

$$N_0 \sim \tilde{n}\Omega_0/M_0\Gamma_0, \tag{5.11}$$

where M_0 is the number of modes in 1 cm^3. In[12], $\tilde{n} \sim 10^{17}\ cm^{-3}$ $M_0 \sim 10^{23}\ cm^{-3}$, $\Omega_0/\Gamma_0 \sim 10^3$, so that $N_0 \sim 10^{-3}$. It is not surprising that in this case the rate of decay is identical with that of the spontaneous decay. In the experiments of Alfano and Shapiro,[11] the laser power was one-and-a-half orders of magnitude greater, which corresponds to an increase in $\tilde{n}$ in the regime of Raman scattering by three orders of magnitude; this gives $N_0 \sim 1$. In these experiments, a slowing down of the decay in comparison with the spontaneous by a factor of two was noted. Finally, this agreement with theory should not be given excessive importance, but it is possible that a qualitative comparison is meaningful.

6. CRITERIA

The polarization operator (2.1) corresponds to a diagram of lowest order (I in Fig. 2). Therefore, one should indicate the conditions under which diagrams of higher order (II and III in Fig. 2) can be discarded. It will be shown that this condition is the sufficient smallness of the number of acoustic phonons per unit cell

$$\tilde{N} = \bar{N} a_0^3 \ll \Delta\omega/\Gamma_0, \tag{6.1}$$

where $\bar{N}$ is the concentration of acoustic phonons.

In this connection, we note that the limitations on the pumping power set forth in[8] automatically lead to satisfaction of (6.1), because it is seen from Eqs. (3.19) and (4.2) of this paper that

$$\tilde{N} \lesssim (\Delta\nu/\Gamma_0\tau\Omega_0)^{1/2}. \tag{6.2}$$

If we take $\tau \sim 10^{-9}$ sec, as in[8], and typical values for the other parameters, then the limitation on the pumping means $\tilde{N} \lesssim 10^{-2}$, while the criterion (6.1) means $\tilde{N} \ll 1$. It should be kept in mind that the smallness of $\tilde{N}$ does not mean smallness of N_0, since

$$N_0 \sim \tilde{N}\Omega_0/\Delta\omega \sim 10^3\ \tilde{N}. \tag{6.3}$$

The limitation on the pumping means $N_0 \lesssim 10$; the critical value $N_0^* \sim 1$ can then be achieved and exceeded.

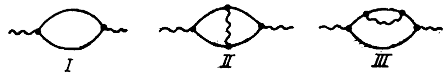

FIG. 2

For what follows, it is also useful to recall a result that follows from[8]:

$$\tilde{n} \ll \tilde{N} \lesssim 10^{-2}, \tag{6.4}$$

where $\tilde{n}$ is the number of optical phonons in a unit cell.

We now proceed to estimate of diagrams of different orders for the polarization operator. Diagram III of Fig. 2 corresponds to the renormalization of the Green's function of the acoustic phonon, a function assumed to be given; therefore, this diagram is unimportant. The required criterion will be obtained from a comparison of diagrams II and I. The expressions for the polarization operators of the first two orders in terms of the Green's function take the form

$$\begin{aligned} P_r(\Omega)^{\mathrm{I}} &= ia^2 \int \frac{d^3q}{(2\pi)^3} \int \frac{d\omega}{2\pi} D_s(\mathbf{q},\omega) D_r(\mathbf{q},\Omega-\omega), \\ P_r(\Omega)^{\mathrm{II}} &= -\frac{1}{2} a^4 \int \frac{d^3q}{(2\pi)^3} \int \frac{d\omega}{2\pi} \int \frac{d^3q'}{(2\pi)^3} \int \frac{d\omega'}{2\pi} \cdot \end{aligned} \tag{6.5}$$

$$\begin{aligned} \times \{ [D_r(\mathbf{q},\omega) D_r(\mathbf{q},\Omega-\omega) D_s(\mathbf{q}',\omega') D_a(\mathbf{q}',-\Omega-\omega') + rsar] \\ \cdot G_s(\mathbf{q}+\mathbf{q}',\omega+\omega') + [rssa+rsas+rrar]r \}, \end{aligned} \tag{6.6}$$

$$P_s(\Omega)^{\mathrm{I}} = i \cdot \frac{1}{2} a^2 (rr+aa+ss), \tag{6.7}$$

$$\begin{aligned} P_s(\Omega)^{\mathrm{II}} = -\frac{1}{2} a^4 \Big\{ \Big[rssr + \frac{1}{2}(rraa+aarr+rrss+ssrr) \Big] s \\ + [rsaa+rsss+srrr+rrsr+aars+ssrs] r \Big\} \end{aligned} \tag{6.8}$$

Here the Green's functions and all the arguments are not written out completely where these are clear, and only the indices of these functions are shown in the corresponding linear combinations.

The distinguishing feature of our problem is the existence of the narrow distributions $N_{\mathbf{q}}$ and $n_{\mathbf{k}}$, which are present only in D_s and G_s. Therefore, the terms which do not contain these functions are estimated in the usual fashion:

$$P^{\mathrm{II}}/P^{\mathrm{I}} \sim \Gamma_0/\Omega_0. \tag{6.9}$$

We first consider the estimate of P_r^{I}. Because of the factor $N_{\mathbf{q}}$ in D_s, the essential $\omega_{\mathbf{q}}$ lie in an interval of the order of $\Delta\omega$ near $\omega_0 = \Omega_0/2$. Because of $\delta(\omega \pm \omega_{\mathbf{q}})$ which enters into D_s, the essential ω are in an interval of the order of $\Delta\omega$ around $\pm\Omega_0/2$. The values of Ω of interest to us lie near $\pm\Omega_0$; therefore $\Omega - \omega$ is near $\pm\Omega_0/2$ and the denominators in D_r turn out to be small. They are estimated by the value of $\Delta\omega$ in place of the ordinary estimate Ω_0. Integration with respect to q in place of the ordinary q_0^3 gives $N_0 q_0^3\ \Delta\omega/\Omega_0$. As a result the estimate of P_r^{I} differs from the usual one by the factors

$$(\Delta\omega/\Omega_0)^{-1}(N_0\Delta\omega/\Omega_0) = N_0. \tag{6.10}$$

We now proceed to the estimate of P_r^{II}. We first estimate the term [rssa]r. It is easy to see that in this case both the functions D_r and D_a contain small denominators. The energy parameter $\omega + \omega'$ in G_r can be close to $\pm\Omega_0$; however, this fact has no value if the dispersion of the optical phonons is significant, because the directions of $\mathbf{q}$ and $\mathbf{q}'$ are never connected and therefore $|\mathbf{q} + \mathbf{q}'| \sim q_0$. Thus, the estimate of this term differs from the usual one by the factors

$$(\Delta\omega/\Omega_0)^{-2}(N_0\Delta\omega/\Omega_0)^2 = N_0^2. \tag{6.11}$$

It is obvious that the term [rsas]r is of the same order, and the term [rrar]r is unimportant, as was pointed out above.

In the estimate of the terms of P_r^{II} which contains G_s, it is necessary to keep in mind that G consists of two parts: one does not contain n_k, and the second is proportional to n_k. In the first case, the estimate of G_s does not differ from the estimate of G_r and then it is easy to prove that the terms [rrsa]s and [rsar]s are estimated by the additional N_0. In the second case, $q + q' = k \sim 0$. This has no effect on the estimate of the functions D_r and D_a, but decreases the phase volume from $q_0^3 q_0^3$ to $q_0^3 k^3$, where k^3 is the volume around $k = 0$, where the distribution of n_k is concentrated. After these observations, it is clear that the factor

$$n_k q_0^3 k_0^3 / q_0^3 q_0^2 \sim \tilde{n} a_0^3 \sim \tilde{n} \qquad (6.12)$$

arises in addition to N_0. Summing up all the estimates made, one can see that the ratio P_r^{II}/P_r^{I} can differ from the usual estimate (6.9) by the factors N_0 and $\tilde{n}$. If (6.1) is satisfied, then it follows from (6.3) and (6.4) that this ratio is small. The estimate of the ratio P_s^{II}/P_s^{I} is obtained similarly.

The authors express their gratitude for discussion of particular aspects of this research to L. P. Gor'kov and É. I. Rashba.

[1]M. J. Colles and J. A. Giordmain, Phys. Rev. Lett. **27**, 670 (1971).
[2]M. J. Colles, J. de Phys. **33**, suppl. au. No. 10. 41 (1972).
[3]R. Orbach, Phys. Rev. Lett. **16**, 15 (1966).
[4]R. Orbach, IEEE Trans. SU-14, 140 (1967).
[5]J. A. Giordmain and W. Kaiser, Phys. Rev. **144**, 676 (1966).
[6]J. P. Coffinet and F. de Martini, Phys. Rev. Lett. **22**, 60 (1969).
[7]A. Laubereau, D. von der Linde and W. Kaiser, Optics Comm. **7**, 173 (1973).
[8]I. B. Levinson, Zh. Eksp. Teor. Fiz. **65**, 331 (1973) [Sov. Phys.-JETP **38**, 162 (1974)].
[9]S. A. Bulgadaev and I. B. Levinson, ZhETF Pis. Red. **19**, 583 (1974) [JETP Lett. **19**, 304 (1974)].
[10]R. Orbach and L. A. Vredevoe, Physics 1, 91 (1964).
[11]R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. **26**, 1247 (1971).
[12]A. Laubereau, D. von der Linde and W. Kaiser, Phys. Rev. Lett. **27**, 802 (1971).
[13]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1515 (1964) [Sov. Phys.-JETP **20**, 1018 (1965)].

Translated by R. T. Beyer
248