Distribution of hot phonons generated by laser radiation

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(Submitted February 13, 1973)
Zh. Eksp. Teor. Fiz. **65**, 331–341 (July 1973)

Generation of long-wave optical phonons under "beats" of two laser beams with a frequency difference close to the phonon frequency is considered. The optical phonons produced decay into two short-wave acoustic phonons. The number of acoustic phonons thus produced depends strongly on the line shape of the exciting radiation. This number is finite for any excitation intensity, in contrast to the predictions of the Orbach theory.

INTRODUCTION

The possibility of generating a large number of non-equilibrium optical phonons with the aid of a laser has recently attracted attention. The generation is made possible by Raman scattering of the radiation from the lattice vibrations[1,2] or by beats of two laser beams whose frequency difference is close to the frequency of the lattice vibrations[3,4]. The total concentration of the produced optical phonons can be estimated from the formula $\bar{n} \sim Q/\Omega_0\Gamma_0$, where Q is the power absorbed per unit volume, Ω_0 is the frequency of an optical phonon with k = 0, and Γ_0^{-1} is the lifetime of this phonon. Typical values for experiments in diamond[4], viz., $\sim 10^7$ W/cm^3, $\Omega_0 = 2.5 \times 10^{14}$ sec^{-1}, and $\Gamma_0 = 3 \times 10^{11}$ sec^{-1} yield $\bar{n} \sim 10^{15}$ cm^{-3}. We note by way of comparison that the equilibrium concentration in diamond at T = 77°K is $\bar{n}_T \sim 10^{12}$ cm^{-3}.

The main mechanism that bounds the lifetime of the produced long-wave optical phonon in pure crystals is its decay into two short-wave acoustic phonons with opposite momenta. There is apparently no experimental information on the lifetime of such phonons. Theoretical estimates indicate that the transverse short-wave acoustic phonons (unlike the longitudinal ones) should have very large lifetimes[5]. We can therefore expect such phonons to accumulate in appreciable amounts when optical phonons are generated, as was indeed observed in luminescence experiments[4]. We assume henceforth for simplicity that there is only one mode of acoustic phonons, and that these phonons have a lifetime $\tau \sim 10^{-9}$ sec. The concentration $\bar{N}$ of the acoustic phonons can be estimated from the balance $\bar{N}/\tau\Gamma \sim \bar{n}\Gamma_0$, which yields $\bar{N} \sim 10^{17}$ cm^{-3}.

It is seen from the foregoing estimates that in the aforementioned experiments the phonon system is essentially in a non-equilibrium state. The imbalance is aggravated also by the fact that the produced phonons are distributed over a very small number of optical modes m and acoustic modes M. The number estimated of excited modes differs greatly in different papers. There is a correspondingly large discrepancy in the estimated occupation numbers $n \sim \bar{n}/m$ and $N \sim \bar{N}/M$.

Thus, Colles and Giordmaine[4] believe that the filled optical phonon modes **k** are those whose frequencies Ω_k differ from Ω_0 by an amount on the order of Γ_0; this yields $m \sim 10^{20}$ cm^{-3} and $n \sim 10^{-5}$ in diamond. On the other hand, Lauberau et al.[2] have assumed that the number of excited modes is determined by the uncertainty of the wave vectors of the light, an uncertainty connected with the finite region of effective interaction; this yields $m \sim 10^7$ cm^{-3} and $n \sim 10^{10}$. For comparison we indicate that the total number of optical modes in diamond is 3×10^{23} cm^{-3} and that at nitrogen temperature the equilibrium value is $n \sim 10^{-11}$. The number M of excited acoustic modes depends on the width $\Delta\omega_q$ of the distribution of the acoustic phonons near the frequency $\omega_{q_0} = \Omega_0/2$. Lauberau et al.[2] assume that $\Delta\omega_q \sim \Gamma_0$; this yields $M \sim 10^{20}$ cm^{-3} and $N \sim 10^{-2}$. Orbach[6,7] assumes in fact $\Delta\omega_q \sim \tau^{-1}$, meaning $M \sim 10^{17}$ cm^{-3} and $N \sim 10$.

It is clear from the foregoing that regardless of which of the estimates is correct, the situation in the discussed experiments is one with narrow phonon distributions, the widths of which can be determined by the lifetimes of the nonmonochromaticity of the excitation. In these situations it is therefore impossible to use the usual kinetic balance equations for the occupation numbers n_k and N_q, and it is necessary to use equations of more general character. The first attempt in this direction was made by Orbach[6,7], who in essence replaced the delta-function in the energy conservation law of the usual kinetic equation by a Lorentz curve of width τ^{-1}. While this idea is correct as far as the physical meaning of the problem goes, its realization was in our opinion incorrect. The point is that the energy conservation law pertains to the $\Omega_0 \to 2\omega_{q_0}$ decay and the uncertainty can be connected not only with the width τ^{-1} of the acoustic-phonon level, but also with the width Γ_0 of the optical-phonon level, which is larger by several orders of magnitude. An additional cause of the broadening can also be the nonmonochromaticity $\Delta\nu$ of the excitation. The beams used in the experiments have spectral widths $\Delta\nu \sim 0.1$–1 cm^{-1}, which are comparable with $\Gamma_0 = 1.5$ cm^{-1} and greatly exceed τ^{-1}.

At sufficiently high light intensities, when $N_q \gtrsim 1$, the $\Omega_0 \to 2\omega_{q_0}$ decay can become stimulated. The effective decay frequency then exceeds Γ_0 and should be obtained from the equations simultaneously with the occupation numbers. An important role can be assumed then also by the frequency renormalization $\Delta\Omega_0$, since it depends on the intensity and takes the optical phonon out of resonance when beats are produced between the two beams.

1. GENERAL EQUATIONS

The phonon generation theory constructed in the present paper pertains to the caee of stationary and spatially homogeneous excitation, when the correlation function $\langle E(r, t)E(\mathbf{r}', t')\rangle$ of the light field dependes only on the differences $r-r'$ and $t-t'$. This means that the duration of the laser pulse is larger than the lifetimes of all the phonons, and that effects of spatial amplification and attenuation of the light beams can be neglected.

It is assumed that the acoustic phonons relax on a thermostat with T = 0. Since only the small energy region of these phonons near ω_{q_0} is of importance, we can assume the corresponding relaxation time τ to be constant. The optical phonons are assumed to be active only in Raman scattering and inactive in infrared absorption. This simplifies the problem greatly, since it makes it possible to disregard polariton effects. Since we do not distinguish between longitudinal and transverse acoustic phonons, it is natural to neglect all the effects connected with the polarization of the light and of the optical phonons and with the anisotropy of the crystal. We therefore regard all the fields as scalar and the crystal as isotropic.

To derive the equations that replace the kinetic ones, we used the Keldysh diagram technique[8]. Only two of the four Green functions in this technique are independent, one retarded D_r and the other statistical D_s. For free phonons (say, acoustic), they are given by

$$D_r^0(q) = \omega_q^2[(\omega + i\eta)^2 - \omega_q^2]^{-1}, \quad \eta \to +0; \tag{1.1}$$

$$D_s^0(q) = -i\pi\omega_q(2N_q + 1)[\delta(\omega - \omega_q) + \delta(\omega + \omega_q)], \quad q = \{\omega, \mathbf{q}\}. \tag{1.2}$$

The exact function D_r is defined by a corresponding polarization operator Π_r, but it is more convenient to use in its place the quantities

$$\gamma(q) = -\operatorname{sign}\omega \cdot \omega_q \operatorname{Im}\Pi(q), \tag{1.3}$$

$$\Delta\omega(q) = {}^1\!/_2\omega_q \operatorname{Re}\Pi(q), \quad \omega_q + \Delta\omega(q) = \tilde{\omega}(q), \tag{1.4}$$

which represent the width and the shift of the level ω_q on the mass shell $\omega = \omega_q$.

Instead of the exact function D_s it is convenient to introduce, in analogy with (1.2), the function N(q), defined by the relation

$$D_s(q) = -i\pi\omega_q[2N(q) + 1]\{\delta(\gamma(q)\,|\,\omega - \tilde{\omega}(q)) + \delta(\gamma(q)\,|\,\omega + \tilde{\omega}(q))\}, \tag{1.5}$$

where we have introduced the smeared delta-function

$$\delta(\gamma\,|\,\omega - \omega_0) = \frac{\gamma}{2\pi}\frac{1}{(\omega - \omega_0)^2 + (\gamma/2)^2}. \tag{1.6}$$

Replacements for the kinetic equations are formulated for the functions N(q), γ(q), $\Delta\omega$(q) and the corresponding quantities n(k), Γ(k), and $\Delta\Omega$(k) for the optical phonons, where k = {Ω, **k**}. The system of equations consists of the balance equations for the "occupation numbers" N(q) and n(k) and the equations expressing the "widths" γ(q) and Γ(k) and the "shifts" $\Delta\omega$(q), $\Delta\Omega$(k) in terms of the occupation numbers.

The width of the acoustic phonon is

$$\gamma(q) = 1/\tau + \gamma^a(q), \tag{1.7}$$

where the first term is connected with the decay of the acoustic phonons on the thermostat, while the second is due to the interaction with the long-wave optical phonons

$$\gamma^a(q) = A\int d^4k\,\delta(\Gamma(k)\,|\,\Omega - \tilde{\Omega}(k)) \times \delta(\gamma(k-q)\,|\,\Omega - \omega - \tilde{\omega}(k-q))\{N(k-q) - n(k)\}. \tag{1.8}$$

Here A is the constant for the decay of an optical phonon into two acoustic phonons. The acoustic-phonon level shift due to scattering by the thermostat can be regarded as included in the nonrenormalized spectrum ω_q, and the shift due to the interaction with the long-wave optical phonons is

$$\Delta\omega^a(q) = \frac{1}{2\pi}A\int d^4k[P(\Gamma(k)\,|\,\Omega - \tilde{\Omega}(k))\delta(\gamma(k-q)\,|\,\Omega - \omega - \tilde{\omega}(k-q)) \times N(k-q) - \delta(\Gamma(k)\,|\,\Omega - \tilde{\Omega}(k))P(\gamma(k-q)\,|\,\Omega - \omega - \tilde{\omega}(k-q))n(k)], \tag{1.9}$$

where we have introduced the smeared principal-value function

$$P(\gamma\,|\,\omega - \omega_0) = \frac{\omega - \omega_0}{(\omega - \omega_0)^2 + (\gamma/2)^2}. \tag{1.10}$$

The acoustic-phonon balance equation takes the form

$$A\int d^4k\,\delta(\Gamma(k)\,|\,\Omega - \tilde{\Omega}(k))\delta(\gamma(k-q)\,|\,\Omega - \omega - \tilde{\omega}(k-q)) \times \{n(k)[N(q) + N(k-q) + 1] - N(q)N(k-q)\} = \tau^{-1}N(q). \tag{1.11}$$

The optical phonons interact with the electromagnetic field E, which is assumed to be classical, i.e., its components can be assumed to commute. The interaction with the field then makes no contribution to the width Γ and the shift $\Delta\omega$, which are expressed as follows:

$$\Gamma(k) = \frac{1}{2}A\int d^4q\,\delta(\gamma(q)\,|\,\omega - \tilde{\omega}(q)) \times \delta(\gamma(k-q)\,|\,\Omega - \omega - \tilde{\omega}(k-q))[2N(q) + 1], \tag{1.12}$$

$$\Delta\Omega(k) = \frac{1}{2\pi}\frac{1}{2}A\int d^4q\,\delta(\gamma(q)\,|\,\omega - \tilde{\omega}(q)) \times P(\gamma(k-q)\,|\,\Omega - \omega - \tilde{\omega}(k-q))[2N(q) + 1]. \tag{1.13}$$

The balance equation for the optical phonons is

$$\frac{1}{2}A\int d^4q\,\delta(\gamma(q)\,|\,\omega - \tilde{\omega}(q))\delta(\gamma(k-q)\,|\,\Omega - \omega - \tilde{\omega}(k-q)) \times \{N(q)N(k-q) - n(k)[N(q) + N(k-q) + 1]\} = -G(k). \tag{1.14}$$

The right-hand side describes here generation of optical phonons as a result of Raman scattering

$$G(k) = B\int d^4x\,e^{-ikx}\langle E(x)E(0)\rangle^2, \quad kx = \mathbf{kr} - \Omega t, \tag{1.15}$$

where B is the constant responsible for the interaction of the phonons with the light.

The constants A and B can be related to customarily measured quantities. The spontaneous-decay frequency of an optical phonon with k = 0 is

$$\Gamma_0 = \frac{\pi q_0^2}{s}A, \quad \omega_{q_0} = \frac{1}{2}\Omega_0, \quad s = \left|\frac{\partial\omega_q}{\partial\mathbf{q}}\right|_{q=q_0}. \tag{1.16}$$

The total probability of Raman scattering of a quantum ν with conversion into a quantum $\nu' = \nu - \Omega_0$ is

$$w = \frac{4\pi}{c^3}\nu\nu'^3 B. \tag{1.17}$$

In the derivation of the equations for the occupation numbers, shifts, and widths it was assumed that the phonon polarization operators can be calculated in the lowest order in the constants A and B. This is ensured by the smallness of the parameters Γ_0/Ω_0 and w/Ω_0.

The true occupation numbers can be obtained from the formulas

$$N_q = \int d\omega\,\delta(\gamma(q)\,|\,\omega - \tilde{\omega}(q))N(q), \tag{1.18}$$

$$n_k = \int d\Omega\,\delta(\Gamma(k)\,|\,\Omega - \tilde{\Omega}(k))n(k). \tag{1.19}$$

It is easily seen that if N(q) and n(k) change little when ω and Ω are changed by amounts on the order of γ and Γ, then all the smeared delta-functions can be replaced by ordinary ones and it is possible to integrate over the frequencies in (1.11) and (1.14). The equations are then closed on the mass shell and turn into the ordinary kinetic equations for N_q and n_k.

2. EQUATION FOR THE OCCUPATION NUMBERS OF LONG-LIVED ACOUSTIC PHONONS

We shall solve the system of equations for the occupation numbers, shifts, and widths under the following assumptions: 1) in Eqs. (1.11) and (1.14) we can neglect the term containing N(q)N(k − q) γ^a and $\Delta\omega^a$ do not exceed τ^{-1}, which in turn is smaller than all widths, i.e.,

Γ_0 and $\Delta\nu$. The corresponding criterion will be presented later.

Assumption (2) enables us to replace the smeared functions δ and P, with widths γ, by the usual functions, and assume that $\tilde{\omega}(q) = \omega_q$. We then obtain in place of (1.12) and (1.13)

$$\Gamma(k)=\frac{1}{2}A\int d^3\mathbf{q}\delta(\Omega-\omega_\mathbf{q}-\omega_{\mathbf{k}-\mathbf{q}})(2N_\mathbf{q}+1), \tag{2.1}$$

$$\Delta\Omega(k)=\frac{1}{2\pi}\frac{1}{2}A\int d^3\mathbf{q}\frac{P}{\Omega-\omega_\mathbf{q}-\omega_{\mathbf{k}-\mathbf{q}}}(2N_\mathbf{q}+1). \tag{2.2}$$

Owing to assumption 1), it follows from (1.14) that

$$n(k) = G(k)/\Gamma(k). \tag{2.3}$$

We now substitute (2.3) in (1.11), integrate with respect to Ω, and put $\omega = \omega_q$. Changing then from integration with respect to $\mathbf{k}$ to integration with respect to $q' = k - q$, we obtain

$$\frac{A}{2\pi}\int d^3q'\frac{G_{\mathbf{q},\mathbf{q}'}}{(\omega_\mathbf{q}+\omega_{\mathbf{q}'}-\Omega_{\mathbf{q}+\mathbf{q}'}-\Delta\Omega_{\mathbf{q},\mathbf{q}'})^2+(\Gamma_{\mathbf{q},\mathbf{q}'}/2)^2}\times(N_\mathbf{q}+N_{\mathbf{q}'}+1)=\frac{1}{\tau}N_\mathbf{q}; \tag{2.4}$$

$$G_{\mathbf{q},\mathbf{q}'}=G(\omega_\mathbf{q}+\omega_{\mathbf{q}'},\ \mathbf{q}+\mathbf{q}'), \tag{2.5}$$

$$\Gamma_{\mathbf{q},\mathbf{q}'}=\Gamma(\omega_\mathbf{q}+\omega_{\mathbf{q}'},\ \mathbf{q}+\mathbf{q}'),\quad \Delta\Omega_{\mathbf{q},\mathbf{q}'}=\Delta\Omega(\omega_\mathbf{q}+\omega_{\mathbf{q}'}\ \mathbf{q}+\mathbf{q}'). \tag{2.6}$$

Substituting (2.1) and (2.2) in (2.6), we obtain a closed system for the occupation numbers of the acoustic phonons. Once we obtain N_q, we can calculate Γ and $\Delta\Omega$ with the aid of (2.1) and (2.2), and then, using (2.4) and (1.9), find the distribution of the optical phonons

$$n_\mathbf{k}=\int\frac{d\Omega}{2\pi}\frac{G(k)}{[\Omega-\tilde{\Omega}(k)]^2+[\Gamma(k)/2]^2}. \tag{2.7}$$

The absorption power per cm^3 is expressed in terms of the concentration of the acoustic phonons $\overline{N}$ in the form

$$Q={}^1/_2\Omega_0\overline{N}/\tau. \tag{2.8}$$

Substituting (2.4), we obtain

$$Q=\Omega_0\int\frac{d^4k}{(2\pi)^4}\frac{G(k)}{[\Omega-\tilde{\Omega}(k)]^2+[\Gamma(k)/2]^2}\Gamma(k). \tag{2.9}$$

For comparison, we write down the total concentration of the optical phonons

$$\bar{n}=\int\frac{d^4k}{(2\pi)^4}\frac{G(k)}{[\Omega-\tilde{\Omega}(k)]^2+[\Gamma(k)/2]^2}. \tag{2.10}$$

We now obtain G(k) for a field consisting of two uncorrelated beams with wave vectors f_1 and f_2:

$$E(x)=E_1(x)+E_2(x); \tag{2.11}$$

$$E_1(x)=A_1(x)e^{if_1x}+\text{к.c.},\quad f=\{\nu,\mathbf{f}\}; \tag{2.12}$$

and similarly for E_2. Here A_1 and A_2 are slowly varying functions describing the nonmonochromaticity of the beams. It is assumed that $\nu_1-\nu_2 \approx \Omega_0$.

When calculating G(k) it is necessary to retain only those of $\langle E(x)E(0)\rangle^2$ which depend on the time with frequency $\nu_1-\nu_2$. We then obtain

$$G(k)=B\int d^4xe^{-ikx}\{\langle A_1(x)A_1(0)^*\rangle\langle A_2(x)^*A_2(0)\rangle\exp[i(f_1-f_2)x]+\text{c.c}\}. \tag{2.13}$$

We note also that

$$\int\frac{d^4k}{(2\pi)^4}G(k)=\frac{1}{2}B\left(\frac{4\pi}{c}\right)^2J^2,\quad J\equiv(J_1J_2)^{1/2}, \tag{2.14}$$

where J_1 and J_2 are the beam intensities.

It is seen from (2.13) that the function G(k) is localized near $\Omega = \overline{\Omega} \equiv \nu_1-\nu_2$ and $\mathbf{k} = \overline{\mathbf{k}} = \mathbf{f}_1-\mathbf{f}_2$. The degree of localization is determined by the temporal and spatial nonmonochromaticity of the beams, which can be described by the frequency and wave-vector spreads $\Delta\nu$ and $\Delta\mathbf{f}$.

It follows from (2.7) that the k-space region in which the optical modes are excited is determined by the spatial nonmonochromaticity, i.e.,

$$m\sim(\Delta\mathbf{f})^3/(2\pi)^3. \tag{2.15}$$

Only the degree of excitation of these modes depends on the temporal nonmonochromaticity $\Delta\nu$ and on the detuning $\overline{\Omega}-\Omega_0$. This can be easily verified, for example, in the case when G(k) factorizes into a product of functions of Ω and $\mathbf{k}$.

3. DISTRIBUTION OF ACOUSTIC PHONONS AT LOW SPATIAL NONMONOCHROMATICITY OF THE EXCITATION

To simplify (2.4), we assume that the $\mathbf{k}$-space region where the optical phonons are produced is smaller than the thickness of the spherical layer Δq near q_0 in q-space, where the acoustic phonons are produced. We can then put

$$G(k)=(2\pi)^3\delta(\overline{\mathbf{k}})\overline{G}(\Omega) \tag{3.1}$$

and obtain from (2.4) and (2.6) the following system of equations for N_q

$$\frac{4\pi\Gamma_0 s\tau}{q_0^2}\frac{\overline{G}(2\omega_\mathbf{q})}{[2\omega_\mathbf{q}-\Omega_0-\Delta\Omega(2\omega_\mathbf{q},0)]^2+[\Gamma(2\omega_\mathbf{q},0)/2]^2}(2N_\mathbf{q}+1)=N_\mathbf{q}, \tag{3.2}$$

$$\Gamma(2\omega_\mathbf{q},0)=\Gamma_0(2N_\mathbf{q}+1), \tag{3.3}$$

$$\Delta\Omega(2\omega_\mathbf{q},0)=\Gamma_0\frac{1}{2\pi}\int d\omega_{\mathbf{q}'}\frac{P}{\omega_\mathbf{q}-\omega_{\mathbf{q}'}}(2N_{\mathbf{q}'}+1). \tag{3.4}$$

The characteristic intensity in the present problem is the quantity

$$J^*=\frac{c}{4\pi}\left[\frac{q_0^2}{4\pi s\tau}\frac{\Delta\nu\Gamma_0}{2\pi B}\right]^{1/2}. \tag{3.5}$$

We now change over to the dimensionless quantities

$$x=\frac{2\omega_\mathbf{q}-\Omega_0}{\Gamma_0},\quad \Delta(x)=\frac{\Delta\Omega}{\Gamma_0},\quad L=\frac{J}{J^*},\quad a=\frac{\Delta\nu}{2\Gamma_0}. \tag{3.6}$$

We then obtain from (3.2)–(3.4)

$$2aL^2\frac{\varphi(x)}{[x-\Delta(x)]^2+[N(x)+{}^1/_2]^2}[N(x)+{}^1/_2]=N(x), \tag{3.7}$$

$$\Delta(x)=\frac{1}{\pi}\int dx'\frac{P}{x-x'}N(x'), \tag{3.8}$$

where $\varphi(x)$ is a dimensionless spectral form factor normalized to unity, so that $\varphi(x) \sim a^{-1}$.

At low intensities, when $L \ll 1$, the value of N(x) is small for all x, and we can put $\Delta(x) = 0$. It follows therefore from (3.7)

$$N(x)=2aL^2\frac{\varphi(x)}{x^2+{}^1/_4}. \tag{3.9}$$

We see thus that in the case of broad-band excitation, when $a \gg 1$, the distribution of the acoustic phonons duplicates the line contour of the optical phonon. In the case of narrow-band excitation, the distribution of the acoustic phonons duplicates the excitation spectral contour. This situation is analogous to resonance luminescence, where the role of the intermediate state is played by the state with an optical phonon[9].

At high intensities, when L >> 1, we can obtain a solution for two characteristic cases, a slowly decreasing excitation with a Lorentz contour

$$\varphi(x)=\frac{a}{\pi}\frac{1}{(x-\bar{x})^2+a^2}, \tag{3.10}$$

and a sharply decreasing excitation with a rectangular contour

$$\begin{aligned}\varphi(x)&=\frac{1}{2a}, \quad |x-\bar{x}|<a,\\ \varphi(x)&=0, \quad |x-\bar{x}|>a.\end{aligned} \tag{3.11}$$

Here $\bar{x}$ denotes the center of the excitation:

$$\bar{x}=\frac{\bar{\Omega}-\Omega_0}{\Gamma_0}.$$

We assume also that the detuning of the excitation relative to the optical phonon and the width of the excitation line are small, i.e.,

$$|\bar{x}|\leqslant 1, \quad a\leqslant 1. \tag{3.12}$$

In the region where N(x) >> 1, Eq. (3.7) takes the form

$$[x-\Delta(x)]^2+N(x)^2=2aL^2\varphi(x). \tag{3.13}$$

For a Lorentz excitation contour, the solution is

$$N(x)=\left(\frac{2}{\pi}\right)^{1/2}L\frac{a^2}{(x-\bar{x})^2+a^2}. \tag{3.14}$$

It is easy to show that in this case

$$\Delta(x)=\left(\frac{2}{\pi}\right)^{1/2}L\frac{a(x-\bar{x})}{(x-\bar{x})^2+a^2}. \tag{3.15}$$

In the region of the excitation contour we have $|x-x|$ ~ a, and therefore

$$N(x)\sim|\Delta(x)|\sim L\gg 1,$$

at the same time we have in this region

$$|x|\leqslant|\bar{x}|+|x-\bar{x}|\leqslant 1.$$

Thus, Eq. (3.13) is equivalent to the equation

$$\Delta(x)^2+N(x)^2=2aL^2\varphi(x), \tag{3.16}$$

which is obviously satisfied.

For a rectangular excitation contour, the solution is

$$\begin{aligned}N(x)&=La^{-1}[a^2-(x-\bar{x})^2]^{1/2}, \quad |x-\bar{x}|<a,\\ N(x)&=0, \quad |x-\bar{x}|>a.\end{aligned} \tag{3.17}$$

We can easily show that

$$\begin{aligned}\Delta(x)&=La^{-1}(x-\bar{x}), \quad |x-\bar{x}|<a,\\ \Delta(x)&=La^{-1}\{(x-\bar{x})-\operatorname{sign}(x-\bar{x})[(x-\bar{x})^2-a^2]\}, \quad |x-\bar{x}|>a,\end{aligned} \tag{3.18}$$

and verify that (3.13) is satisfied when L >> 1.

It is seen from the obtained solutions that both in the case of Lorentz excitation and in the case of rectangular excitation almost all the acoustic phonons are concentrated in a spherical layer of thickness $\Delta q\sim\Delta\nu/s$. However, whereas in the case of rectangular excitation there are no excited modes for which N_q >> 1 outside this layer, in the case of Lorentz excitation these modes exist also outside the layer; they are concentrated in a thicker layer with $\Delta q\sim(J/J^*)^{1/2}(\Delta\nu/s)$.

This circumstance has little effect on the total concentration of the acoustic phonons, which is equal to

$$\bar{N}=c\frac{1}{(2\pi)^3}\frac{4\pi q_0^2}{s}\Delta\nu\frac{J}{J^*}, \tag{3.19}$$

($c=\sqrt{\pi/8}$ for Lorentz excitation and $c=\pi/8$ for rectangular excitation) but affects strongly, as we shall show, the concentration of the optical phonons. This concentration can be obtained by substituting (3.1) in (2.10) and using (3.2). We then obtain

$$\bar{n}=\frac{1}{2\Gamma_0\tau}\int\frac{d^3q}{(2\pi)^3}\frac{N_q}{2N_q+1}. \tag{3.20}$$

At low intensities this yields the obvious balance equation

$$\bar{n}=\bar{N}/2\Gamma_0\tau. \tag{3.21}$$

We now introduce

$$\tilde{\Gamma}=\Gamma_0\langle(2N_q+1)^{-1}\rangle^{-1}, \tag{3.22}$$

where the angle brackets denote averaging with the aid of the distribution N_q. Then (3.20) can be rewritten in the form of a balance equation

$$\bar{n}=\bar{N}/2\tilde{\Gamma}\tau, \tag{3.23}$$

where $\tilde{\Gamma}$ has the meaning of the frequency of stimulated decay of the optical phonon. At high intensities we have $\tilde{\Gamma}\gg\Gamma_0$, and the spectral contour of the optical phonon broadens greatly. This is precisely why at high intensities, as seen from (3.19), $\bar{N}$ is independent of the detuning of the excitation relative to the optical phonon.

Substituting (3.14) and (3.17) in (4.20) we obtain explicit expressions for the total optical-phonon concentration at high intensities. For Lorentz excitation we get

$$\bar{n}=\frac{\pi^{3/4}}{2^{7/4}}\frac{1}{2\Gamma_0\tau}\frac{1}{(2\pi)^3}\frac{4\pi q_0^2}{s}\Delta\nu\left(\frac{J}{J^*}\right)^{1/2}; \tag{3.24}$$

and for rectangular excitation

$$\bar{n}=\frac{1}{4}\frac{1}{2\Gamma_0\tau}\frac{1}{(2\pi)^3}\frac{4\pi q_0^2}{s}\Delta\nu. \tag{3.25}$$

We see therefore that $\bar{n}$ increases with increasing J more slowly than $\bar{N}$. The reason is that when J is increased the frequency of the stimulated decay $\tilde{\Gamma}$ increases. In addition, it is seen from the last formulas that the excitation line shape affects strongly the concentration of the optical phonons. This can be deduced also from (3.20), if this formula is rewritten in the form

$$\bar{n}=M/2\Gamma_0\tau, \tag{3.26}$$

where M is the number of those acoustic modes for which N_q >> 1. It is clear from the foregoing remark that for rectangular excitation M does not depend on J, whereas for Lorentz excitation M increases like $J^{1/2}$.

According to (2.8), the absorbed power Q depends on J as $\bar{N}$ does, i.e., it increases like J^2 at low intensities and like J at high intensities. Substituting (3.1) in (2.9), we obtain

$$Q=\Omega_0\int\frac{\Gamma(\Omega,0)}{2\pi}\frac{d\Omega\bar{G}(\Omega)}{[\Omega-\bar{\Omega}_0-\Delta\Omega(\Omega,0)]^2+[\Gamma(\Omega,0)/2]^2}. \tag{3.27}$$

This demonstrates that the quantities $\Delta\Omega$ and Γ in (3.2) actually represent the shift and broadening of the spectral contour of the optical phonon.

4. CRITERIA AND DISCUSSION

Using the obtained solutions, we can indicate the criteria that must be satisfied to make the simplifications used in Secs. 2 and 3 valid. To justify assumptions 1) and 2) of Sec. 2 it is necessary to satisfy the conditions

$$\Gamma_0^2,\ (\Delta\nu)^2,\ \Gamma_0\Delta\nu\ll\frac{1}{\tau}\frac{q_0^2}{2\mu}, \tag{4.1}$$

$$J^2\leqslant(J^*)^2\frac{1}{\tau}\frac{q_0^2}{2\mu}\frac{1}{\Gamma_0\Delta\nu}. \tag{4.2}$$

Here μ is a parameter that determines the dispersion of the optical phonons,

$$\Omega_k = \Omega_0 - \frac{k^2}{2\mu}. \qquad (4.3)$$

The appearance of this parameter can be explained in the following manner. The term $N(q)N(k-q)$ describes the adhesion of acoustic phonons to form optical ones. The large dispersion of the optical phonons makes this adhesion possible only for acoustic phonons with almost oppositely directed momenta, thus greatly reducing the effectiveness of this process.

It suffices to assume that

$$\Delta\nu \lesssim \Gamma_0, \quad \frac{1}{\tau}\frac{q_0^2}{2\mu} \gg \Gamma_0^2. \qquad (4.4)$$

It is then obvious that the condition (4.1) is satisfied and there exists simultaneously a region of high intensities, $J \gg J^*$, where (4.2) is satisfied.

Assumption (3.1) is valid if

$$|\bar{k}|,\ |\Delta f| \ll \Delta\nu/s,\ \Gamma_0/s. \qquad (4.5)$$

For parallel beams we have

$$|\bar{k}| = |f_1 - f_2| \approx \Omega_0/c, \quad |\Delta f| \sim \Delta\nu/c.$$

It is therefore sufficient to assume that

$$\Delta\nu \lesssim \Gamma_0 \quad \Delta\nu/\Omega_0 \gg s/c. \qquad (4.6)$$

We can usually assume that $q_0^2/2\mu \sim \Omega_0$. Then the complete system of inequalities is

$$\Delta\nu \lesssim \Gamma_0, \quad \frac{\Gamma_0}{\Omega_0} \ll \frac{\tau^{-1}}{\Gamma_0}, \quad \frac{\Delta\nu}{\Omega_0} \gg \frac{s}{c}. \qquad (4.7)$$

These inequalities are satisfied at the aforementioned values of the parameters. It is of interest to estimate J^*. Using for diamond the value $w \sim 10^4$ sec^{-1} [10] and assuming $q_0 \sim 10^8$ cm^{-1}, $s \sim 10^6$ cm/sec, and $\nu \sim 10^{15}$ sec^{-1}, we obtain $J^* \sim 10^8$ W/cm^2, which lies in the intensity region where non-equilibrium phonons have been observed[4]. A more detailed comparison with experiment is impossible, since there are no data on the spectral character of the excitation. It is also not clear whether this experiment, in which the pulse duration is $t_p \sim 10^{-8}$ sec, corresponds to the stationary and homogeneous regimes.

The theory developed here does not confirm Orbach's conclusion[6,7] that instability exists, or more accurately, that there is no stationary solution when a certain critical intensity is reached. For rectangular excitation, the reason is that the concentration of the optical phonons does not exceed a certain value $\bar{n}_\infty$ lower than the critical value required for instability. For Lorentz excitation, the increase of $\bar{n}$ is offset by the broadening of the region where acoustic phonons are produced. We note that in accordance with (3.19) and (2.8) the characteristic absorbed power is

$$Q^* = \frac{1}{(2\pi)^3}\frac{4\pi q_0^2}{s}\Delta\nu\frac{\Omega_0}{\tau}. \qquad (4.8)$$

Yet Orbach's theory[6,7] corresponds to the characteristic power of[4], which is obtained from Q^* by replacing $\Delta\nu$ by τ^{-1}, i.e., a value lower by one or two orders of magnitude.

The author thanks S. I. Anisimov, Yu. M. Kagan, É. I. Rashba, V. B. Timofeev, and G. M. Éliashberg for a discussion of the work and for valuable remarks.

Note added in proof (23 May 1973). We have recently learned of a paper by M. J. Colles (J. de Phys. C4, suppl. au No. 10, 41, 1972), which contains a qualitative discussion of the essential times and widths.

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Translated by J. G. Adashko
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