magnetic structure. They are closer in character to the possible spectra of a rhombic antiferromagnet, but the presence of additional lines and the variation of the line intensities with increasing magnetic field indicate that the spectrum contains more than two AFMR branches, and the microwave polarization necessary to observe the AFMR seems to depend on the field. Thus, properties of AFMR may correspond to a helicoidal structure, but a more detailed analysis of the experimental data can be based only on a theoretical investigation of the spin-wave spectrum of the proposed magnetic structure.

CONCLUSION

Thus, the presented aggregate of investigations points to a complex magnetic structure of $FeCl_3$ and does not contradict the antiferromagnetic helicoid structure.[3] A definite part of the details of the experimental results lies outside the scope of the present article, but their discussion, as well as a more detailed analysis of the presented principal (from our point of view) results, call for a special theoretical consideration.

The authors are grateful to L. A. Kupriyanova, V. S. Frolova, and A. A. Motuz for help with the crystal growth.

[1]Crystal Structures 2, ed. R. W. G. Wyckoff, IS, New York, 1967; Gregory, J. Am. Chem. Soc. 73, 472 (1951).
[2]D. E. Earles, R. C. Axtmann, V. Hazony, and I. Lefkowitz, J. Phys. Chem. Solids 29, 1859 (1968).
[3]W. Cable, M. K. Wilkinson, E. O. Wollan, and W. G. Kocheler, Phys. Rev. 127, 714 (1962).
[4]Edwin R. Jones, Jr., O. B. Morton, L. Cathey, Theo. Auel, and E. L. Amma, J. Chem. Phys. 50, 4755 (1969).
[5]J. P. Stampel, W. T. Oesterhuis, and F. de S. Barros, J. Appl. Phys. 42, 1721 (1971).
[6]M. K. Wilkinson, J. W. Cable, and E. O. Wollan, Bull. Am. Phys. Soc. 5, 458 (1960).
[7]C. W. Kocher, Phys. Lett. A24, 93 (1967).
[8]A. F. Lozenko and S. M. Ryabchenko, Pis'ma Zh. Eksp. Teor. Fiz. 16, 332 (1972) [JETP Lett. 16, 234 (1972)].
[9]H. Mori and K. Kawasaki, Prog. Theor. Phys. 28, 971 (1962).
[10]H. Mori, Prog. Theor. Phys. 30, 576, 578 (1963).
[11]S. Maekawa, J. Phys. Soc. Jap. 33, 573 (1972).
[12]D. L. Huber, Phys. Rev. B6, 3180 (1972).
[13]A. Narath, J. Chem. Phys. 40, 1169 (1964).
[14]F. V. Bragin and S. M. Ryabchenko, Fiz. Tverd. Tela 15, 1050 (1973) [Sov. Phys. Solid State 15, 721 (1973)].

Translated by J. G. Adashko

Parametric generation of phonons

S. A. Bulgadaev, B. I. Kaplan, and I. B. Levinson

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences
(Submitted November 20, 1975)
Zh. Eksp. Teor. Fiz. 70, 1550–1565 (April 1976)

A theory of parametric generation of short-wave acoustic phonons during the two-phonon absorption of infrared radiation is constructed. The generation thresholds and the nature of the prethreshold nonequilibrium-phonon distribution arising in broad- and narrow-band pumping are found. As a mechanism limiting the growth of the number of phonons near the threshold, the merging of nonequilibrium phonons into phonons of a higher equilibrium branch is considered. Such a mechanism turns out to be effective only for broad-band pumping. The results of the paper can easily be extended to the case when the phonon generation is effected by the beats of two light beams in the optical range.

PACS numbers: 78.20.Hp

INTRODUCTION

Orbach[1] first drew attention to the fact the decay of a long-wave optical phonon into two short-wave acoustic phonons ($O \to 2A$) is a parametric process, and that, therefore, in generating optical phonons with the aid of light ($\nu \to O$), we can expect the appearance of an instability in the system of acoustic phonons. The instability threshold should be especially low for decay into transverse acoustic phonons (TA), whose lifetime is anomalously long.[2]

The corresponding experiment was performed by Colles and Giordmaine.[3] The indubitable result of the experiment is the generation of nonequilibrium acoustic phonons, but the existence of a sharply defined threshold does not follow from it.

The theory developed in[4] showed that there is no parametric instability in the process $\nu \to O \to 2A$. The point is that as the light intensity and, hence, the acoustic-phonon concentration increase, the rate of decay of the optical phonons also increases, since their decay becomes stimulated. As a result, the growth of the optical-phonon concentration slows down, and it turns out that this concentration reaches saturation, not having attained the threshold value. The same result was obtained by Sparks and Chow.[5]

As can be seen from these arguments, the absence of the acoustic-phonon instability is due to the presence of a real intermediate particle (the optical phonon). Such a situation obtains only in resonance pumping, when the light frequency ν_0 is close to the optical-phonon frequency Ω_0. In this case the process $\nu \to O \to 2A$ proceeds in two stages, and starts with the one-phonon absorption. If, on the other hand, the pumping is nonresonant, i.e., if $|\nu_0 - \Omega_0| \gg \Gamma_0$, where Γ_0 is the optical-phonon width, then the optical phonon in this process is virtual, and we have essentially the two-phonon absorption process to deal with. It can be expected that a parametric instability will occur in this case, although its threshold will be significantly higher (since the two-phonon absorption process is weaker than the one-phonon process). It is precisely this instability that is the subject of the present paper.

It is well known that there also exists another two-phonon absorption mechanism connected with second-order dipole moments. [6] In concrete crystals the contributions of these mechanisms may have different orders of magnitude, [7] but they are literally estimated in the same manner. Therefore, below we shall not distinguish between these mechanisms, but simply introduce an interaction leading to the decay $\nu \to 2A$.

In our opinion no satisfactory theory of parametric generation for processes of the type $\nu \to 2A$ exists. The existing theories, for example, [5] evade the question of the width of the distribution of the generated particles (in our case the acoustic phonons); if, however, this width is introduced, [8] then it is assumed to be independent of the pumping. Meanwhile, this width plays a decisive role, which can be seen from the following estimate for the threshold intensity.

The number of photons absorbable in 1 sec in 1 cm^3 is $JK/\hbar\nu_0$, where J is the light intensity and K is the two-phonon absorption coefficient. The number of phonons produced in 1 sec in 1 cm^3 is of the same order of magnitude. Therefore, their steady-state concentration $\bar{N} \approx JK\tau/\hbar\nu_0$, where τ is the phonon lifetime. However, the threshold is determined not by the concentration $\bar{N}$, but by the occupation numbers N: the threshold intensity is of the order of those intensities for which N attains values of the order of unity. Meanwhile, $N \approx \bar{N}/\Delta Z$, where ΔZ is the number of excited acoustic modes. If we assume that the light momentum $\mathbf{k} = 0$, then the phonons produced have opposite momenta $\mathbf{q}$ and $-\mathbf{q}$ and energy $\omega_1(\mathbf{q})$ and $\omega_2(\mathbf{q})$. If we further assume the light to be monochromatic, then $\omega_1(\mathbf{q}) + \omega_2(\mathbf{q}) = \nu_0$. It follows from this that $q = q_0$, i.e., that only acoustic modes on the surface of the sphere $q = q_0$ are excited, and, therefore, $\Delta Z = 0$. It is clear that in this approximation ΔZ is actually not determined and, therefore, it is impossible to determine the threshold; for this purpose, we should take into account the smearing of the sphere over some thickness $\Delta q \approx \Delta\omega/s$, where $\Delta\omega$ is the generated-phonon distribution width and s is the group velocity at $q = q_0$. Then $\Delta Z \approx \sigma\Delta\omega$, where σ is the phonon density of states, and from the condition that $N \approx 1$ we can "find" the threshold intensity

$$J^* \approx \hbar\nu_0\sigma\Delta\omega/K\tau \approx \hbar\Delta\omega/a_0^3K\tau,$$

where a_0^3 is the volume of the unit cell. Here we have used the fact that $\sigma \approx (\Omega_0 a_0^3)^{-1}$ and $\Omega_0 \approx \nu_0$.

Indeed, this formula does not provide an answer, but only poses the question: What determines $\Delta\omega$? It is clear that $\Delta\omega$ is determined by those factors that violate the energy and momentum conservation laws used above. This may be the finiteness of the light momentum $\mathbf{k}$, the width of the spectral distribution of the light, $\Delta\nu$, the width of the phonon states, γ. In the last case $\Delta\omega$ may turn out to be J dependent, since the actual γ does, in contrast to the spontaneous width τ^{-1}, depend on the nonequilibrium occupation numbers.

If we assume, for orientation, that $\Delta\omega \approx \Delta\nu \approx \tau^{-1} \approx \Gamma_0 \approx 1\ cm^{-1}$, which corresponds to $\tau \approx 10^{-11}$ sec, then for the typical values $K = 10\ cm^{-1}$ and $a_0 = 5 \times 10^{-8}$ cm, we find $J^* \approx 10^9\ W/cm^2$. The assumed τ corresponds to the lifetime of the longitudinal acoustic phonons (LA). The lifetime of the TA phonons is, in fact, unknown. Experiment[9] on silicon gives $\tau_{TA} \approx 10^{-6}$ sec; the results of the experiment[3] on diamond conforms with $\tau_{TA} \approx 10^{-9}$ sec. If, for the sake of caution, we take the latter figure, then we have for TA phonons $J^* \approx 10^7\ W/cm^2$. The estimates made above show that the threshold intensities for generation during two-phonon absorption lie in an entirely accessible region.

1. FORMULATION OF THE PROBLEM

The object of the present paper is to compute the distribution, $N_{\mathbf{q}}$, of nonequilibrium phonons generated by light during nonresonance pumping. The deviation from equilibrium will be greatest for the long-lived TA phonons; therefore, of greatest interest is the process $\nu \to 2TA$. However, this process may turn out to be forbidden (in crystals with a center of inversion), and it is therefore advisable to also consider the process $\nu \to TA + LA$.

We shall neglect the anisotropy of the crystal and the polarization of the light, and the generation processes of interest to us will be described by the Hamiltonian $E\varphi_{TA}^2$ or $E\varphi_{TA}\varphi_{LA}$, where E is the light field, while φ_A is the field of the acoustic phonons with $q \approx q_0$. The corresponding interaction constants are expressible in terms of K. All the phonon modes with q far from q_0 and not directly excitable by the light form some phonon field ψ. We shall assume that the field ψ does not experience excitation, i.e., that it is in thermodynamic equilibrium (with $T = 0$) and that its spectrum does not get renormalized. Therefore, we can assume that the field ψ forms for the field φ_A a thermostat. The lifetime of the phonons with $q \approx q_0$ is, because of the interaction (of the type $\varphi_A\psi^2$) with the thermostat, described by the relaxation times τ_{TA} and τ_{LA}.

As the mechanism limiting the growth near the threshold, we consider only an anharmonicity of the type $\varphi_A^2\psi$, which describes the flocculation of the generated phonons into phonons of higher equilibrium branches.

The light field in the sample is assumed to be given and to coincide with that field that would exist at the given point in space in the absence of the sample, i.e., to be a free field. This means that the sample should be so thin that the secondary fields inside it constitute a small fraction of the incident field.

It is assumed that the light field does not possess phase coherence. This means that there are in any frequency range of interest to us a large number of light modes with random phases (e.g., the various longitudinal modes in the pulse of a laser with Q switching), or that the duration of the experiment is longer than the floating time of the phase of each mode. Strictly speaking, in order to use the diagrammatic techniques, we must assume that each light mode obeys Gaussian statistics.[10] Under these assumptions the light field is completely determined by the occupation numbers n_k. We shall also assume that the light field is classical, i.e., we shall formally retain only the principal—in n_k—terms.

We assume, for convenience, that the light propagates inside the solid angle Δo, and has a Lorentzian frequency distribution of width $\Delta\nu$:

$$n_k = n_0\varphi(\nu_k) \quad \text{inside} \quad \Delta o,$$
$$n_k = 0 \quad \text{outside} \quad \Delta o,$$

$$\varphi(\nu) = \frac{(\frac{1}{2}\Delta\nu)^2}{(\nu-\nu_0)^2 + (\frac{1}{2}\Delta\nu)^2}. \tag{1.1}$$

The effects connected with the finiteness of k will not be considered, i.e., it is assumed that $k \ll \Delta q$, or $\Delta\omega/\nu_0 \gg s/c$. Assuming that $\nu_0 \approx \Omega_0 \approx 10^3$ cm^{-1} and $s/c \approx 10^{-5}$, we obtain $\Delta\omega \gg 10^{-2}$ cm^{-1}. This condition may turn out to be quite burdensome. Thus, it is almost violated when $\Delta\omega \approx \tau^{-1}$ and $\tau \approx 10^{-9}$ sec.

If the spectral width of the light is sufficiently large, i.e., if $\Delta\nu \gg \tau^{-1}$, then we can suppose that $\Delta\omega \approx \Delta\nu$, and this provides some basis for the use of the kinetic equation for the photon and phonon occupation numbers n_k and N_q to solve the formulated problem. However, it then turns out that $\Delta\omega \to 0$ near the threshold, and this calls in question the applicability of the kinetic equation. In the case of narrow-band pumping, i.e., in the case when $\Delta\omega \ll \tau^{-1}$, the classical theory of interaction between monochromatic waves is usually used (see the review[11]). However, there is no basis for the use of the classical theory in the infrared region at low temperatures, when the equilibrium occupation numbers are small. Furthermore, it may turn out that $\tau_{LA}^{-1} \gg \Delta\nu \gg \tau_{LA}^{-1}$ in the generation $\nu \to TA + LA$, so that none of the usually employed classical methods is applicable.

All the foregoing makes a unified, quantum-mechanical treatment based on the equations for the Green functions advisable. We use Keldysh's procedure[12] in the form of generalized kinetic equations.[4]

2. THE EQUATIONS

The generalized kinetic equations consist of balance equations and equations for the renormalization of the spectrum. The balance equations can be written in the form (departure = arrival):

$$N(p)\gamma(p) = B(p). \tag{2.1}$$

Here $N(p)$ is the generalized occupation number, which depends on the 4-momentum p (in contrast to the real occupation number N_p, which depends on the 3-momentum p). Further, $\gamma(p)$ is the "level width," while $B(p)$ is the generalized arrival term. The expressions for B and γ in terms of N resemble the standard expressions, with the only difference that the integration is performed over 4-momenta and not over 3-momenta and that the energy delta-functions are smeared out and shifted in accordance with the renormalization of the spectrum. Into the equations enter the "level shift" $\Delta\omega(p)$, which also depends on N.

If the particle p vanishes in a decay into the particles k and q, and is created when these particles merge, then

$$\gamma(p) = \iint \Delta(k)\Delta(q)[N(k)+N(q)+1], \tag{2.2a}$$

$$\Delta\omega(p) = \frac{1}{2\pi}\iint\left\{P(k)\Delta(q)\left[N(q)+\frac{1}{2}\right] + P(q)\Delta(k)\left[N(k)+\frac{1}{2}\right]\right\}, \tag{2.2b}$$

$$B(p) = \iint \Delta(k)\Delta(q)N(k)N(q). \tag{2.2c}$$

Here the double integral denotes

$$\int\frac{d^4k}{(2\pi)^4}\int\frac{d^4q}{(2\pi)^4}U_{kqp}\delta(p-k-q), \tag{2.3}$$

where U is a quantity proportional to the probability of the process $p \rightleftarrows k+q$; Δ and P are smeared-out and shifted delta and principal-value functions[4]:

$$\Delta(q) = \delta(\gamma(q)\,|\,\omega - \omega(q) - \Delta\omega(q)), \tag{2.4a}$$
$$P(q) = P(\gamma(q)\,|\,\omega - \omega(q) - \Delta\omega(q)). \tag{2.4b}$$

If the particle p vanishes by merging with the particle q and being converted into the particle k, and is produced in the decay of k into p and q, then

$$\gamma(p) = \iint \Delta(k)\Delta(q)[N(q)-N(k)], \tag{2.5a}$$

$$\Delta\omega(p) = \frac{1}{2\pi}\iint\left\{\Delta(k)P(q)\left[N(k)+\frac{1}{2}\right] + P(k)\Delta(q)\left[N(q)+\frac{1}{2}\right]\right\}, \tag{2.5b}$$

$$B(p) = \iint \Delta(k)\Delta(q)N(k)[N(q)+1]. \tag{2.5c}$$

The meaning of the double integral is the same as in (2.3), with the obvious replacement of $\delta(p-k-q)$ by $\delta(p+q-k)$.

Notice that if in the expressions for γ and B we replace the smeared-out and shifted functions Δ and P by the standard sharp functions and take the balance equation (2.1) on the mass shell, then it goes over into the standard kinetic equation. Therefore, the generalized kinetic equations can be interpreted as a basis for, and a refinement of, the formal procedure used earlier in the theory of magnon generation[13] and consisting in the replacement of the delta-function of the energy conservation law by some "line shape."

The usual occupation numbers are expressible in terms of the generalized occupation numbers as follows:

$$N(\mathbf{q}) = \int d\omega\,\Delta(q)N(q). \tag{2.6}$$

Let us now write out the generalized kinetic equations for the acoustic phonons, considering, for the sake of generality, the process $\nu \to TA + LA$, and henceforth setting, for brevity $TA \equiv 1$ and $LA \equiv 2$. The quantities

γ, $\Delta\omega$, and B consist of three parts corresponding to the contributions from the thermostat (index T), generation (index g), and the anharmonicity (index a).

The thermostat

The contributions from the thermostat can be found from the formulas (2.2), in which $N(k)$ and $N(q)$ are the occupation numbers for the field ψ, and are therefore equal to zero. Then it can be seen that $B_{1T}=0$, while $\Delta\omega_{1T}$ and γ_{1T} are smooth functions of ω and $\mathbf{q}$. Therefore, $\Delta\omega_{1T}$ can be included in the bare spectrum, while γ_{1T} can be assumed to be a constant τ_1^{-1}. The same conclusions hold for the phonon 2.

Generation

The contribution from the generation is computed from the formulas (2.5). Remembering that the photon field is assumed to be a free field, we find, using (1.1) and the smallness of k, that

$$\gamma_1(\omega,\mathbf{q})_g=-\lambda\int d\nu\,\varphi(\nu)\Delta_2(\nu-\omega,-\mathbf{q}), \tag{2.7a}$$

$$\Delta\omega_1(\omega,\mathbf{q})_g=\frac{1}{2\pi}\lambda\int d\nu\,\varphi(\nu)P_2(\nu-\omega,-\mathbf{q}), \tag{2.7b}$$

$$B_1(\omega,\mathbf{q})_g=\lambda\int d\nu\,\varphi(\nu)\Delta_2(\nu-\omega,-\mathbf{q})[N_2(\nu-\omega,-\mathbf{q})+1]. \tag{2.7c}$$

Here λ is the rate of change of the occupation numbers of the photons that is due to the generation:

$$\lambda=\frac{2}{\pi}\frac{JK}{\hbar\nu_0\sigma_{12}\Delta\nu}, \tag{2.8}$$

where $\sigma_{12}\equiv\sigma_{12}(\nu_0)$ is the two-phonon density of states:

$$\sigma_{12}(\nu)=\int\frac{d\mathbf{q}}{(2\pi)^3}\delta(\nu-\omega_{12}(\mathbf{q})),\quad \omega_{12}(\mathbf{q})=\omega_1(\mathbf{q})+\omega_2(\mathbf{q}). \tag{2.9}$$

The functions Δ_2 and P_2 figuring in the integrands are determined with the aid of (2.4), into which γ_2 and $\Delta\omega_2$ enter. It is natural that γ_2, $\Delta\omega_2$, and B_2 are determined by formulas similar to (2.7) with the indices 1 and 2 interchanged.

For the decay $\nu\to 2TA$ the correct formulas are obtained if we discard the indices 1 and 2 and define

$$\sigma(\nu)=\frac{1}{2}\int\frac{d\mathbf{q}}{(2\pi)^3}\delta(\nu-2\omega(\mathbf{q})). \tag{2.10}$$

Anharmonicity

It is *a priori* clear that the merging processes in which the LA phonons participate, i.e., the processes $LA+LA$ and $LA+TA$ are of little importance compared to the merging process $TA+TA$; for the number of nonequilibrium LA phonons is much less than the number of nonequilibrium TA phonons, while the rates of all the processes are of the same order of magnitude.

According to (2.5a),

$$\gamma_1(\omega,\mathbf{q})_a=\int\frac{d\mathbf{q}'}{(2\pi)^3}\int d\omega'\frac{U^a_{\mathbf{q}\mathbf{q}'}}{(2\pi)^3}\delta(\omega+\omega'-\Omega(\mathbf{q}+\mathbf{q}'))\Delta_1(\omega',\mathbf{q}')N_1(\omega',\mathbf{q}'), \tag{2.11}$$

where Ω is the energy of a phonon of the field ψ. On account of the isotropy of the problem, the product $\Delta_1 N_1$ does not depend on the direction of $\mathbf{q}'$, and therefore we have

$$\int\frac{do_{\mathbf{q}'}}{4\pi}\frac{U_{\mathbf{q}\mathbf{q}'}}{(2\pi)^3}\delta(\omega+\omega'-\Omega(\mathbf{q}+\mathbf{q}'))\equiv\alpha(q,q',\omega+\omega'). \tag{2.12}$$

This is a smooth function of q, q', and $\omega+\omega'$: therefore, it can be evaluated at $q=q'=q_0$ and $\omega+\omega'=2\omega_1(q_0)$. After this we can perform the integration over ω' with the aid of (2.6). Integrating again over q', we finally find

$$\gamma_1(\omega,\mathbf{q})_a=\alpha\bar{N}_1, \tag{2.13}$$

i.e., the rate at which the TA phonons merge turns out to be proportional to their concentration. In order of magnitude, $\alpha\approx\Gamma_0 a_0^3$, since Γ_0 is a measure of the anharmonicity of all the third-order processes in which phonons with energy of the order of the Debye energy and which do not have any special selection laws conforming to the conservation laws (like the TA phonons) participate.

It can be seen from (2.5b) that $\Delta\omega_{1a}$ has a smooth part which does not depend on the occupation numbers and which can be included in the bare spectrum. The remaining part can be represented as

$$\Delta\omega_1(\omega,\mathbf{q})_a=\frac{1}{2\pi}\beta\bar{N}_1, \tag{2.14}$$

where β is given by the same function that gives α, but with the delta function replaced by its principal value. It follows from (2.5c) that $B_{1a}=0$.

3. THE GENERATION $\nu\to 2TA$ WITHOUT ALLOWANCE FOR THE RESTRICTING ANHARMONICITY

Broad-band pumping: $\Delta\nu\gg\tau^{-1}$

In this case we can seek the solution for which γ and $\Delta\omega$ are smaller than $\Delta\nu$ and smaller than the characteristic interval of variation of $N(\omega,q)$ and $\gamma(\omega,q)$ with respect to ω. Under this assumption we can replace $\Delta(\nu-\omega,q)$ by $(\nu-\omega-\omega_q)$ and assume that $N(\omega_q,q)=N_q$. Then we obtain from (2.7a) and (2.7c)

$$\gamma(\omega,q)=\tau^{-1}-\lambda\varphi(\omega+\omega_q), \tag{3.1a}$$

$$B(\omega,q)=\lambda\varphi(\omega+\omega_q)(N_q+1). \tag{3.1b}$$

Substituting this into the balance equation (2.1) and solving it, we find

$$N(\omega,q)=\frac{\xi(N_q+1)}{2\varphi(\omega+\omega_q)^{-1}-\xi}, \tag{3.2}$$

where $\xi=2\lambda\tau$ is a dimensionless pumping parameter. Setting in this expression $\omega=\omega_q$, and solving it for N_q, we have

$$N=\frac{{}^1\!/_2\xi}{\varphi(2\omega_q)^{-1}-\xi}=N\frac{({}^1\!/_2\Delta\omega)^2}{(\omega_q-\omega_0)^2+({}^1\!/_2\Delta\omega)^2} \tag{3.3}$$

where

$$N_0 = N_{q_0} = {}^1/_2\xi(1-\xi)^{-1}, \quad \Delta\omega = {}^1/_2\Delta\nu(1-\xi)^{1/2}, \quad \omega_0 = {}^1/_2\nu_0. \tag{3.4}$$

Positive values of N_q are obtained for all q only when $\xi < 1$, i.e., $\xi = 1$ corresponds to the parametric-instability threshold. Below the threshold the phonon distribution has a Lorentzian form; as we approach the threshold, the distribution width $\Delta\omega \to 0$, while the height $N_0 \to \infty$. The total phonon concentration

$$\overline{N} = {}^1/_2\pi\Delta\nu\sigma(\nu_0)\xi(1-\xi)^{-1/2} \tag{3.5}$$

also tends to infinity. Notice that the result (3.3) can be obtained from the standard kinetic equation.

Above the threshold, finite N_q values are obtainable only for sufficiently detuned modes $|\omega_q - \omega_0| > 2^{-3/2}\Delta\nu(\xi-1)^{1/2}$. As we go higher above the threshold, the instability region broadens, and for each mode, q, there exists a threshold value equal to

$$\xi_q^* = 1 + 2\frac{(\omega_q-\omega_0)^2}{({}^1/_2\Delta\nu)^2}, \tag{3.6}$$

at which this mode becomes unstable.

Let us verify whether the assumption made is fulfilled. For $\xi < 1$, the characteristic interval of variation of $N(\omega, \mathbf{q})$ is, according to (3.2), $\Delta\nu$, and $\gamma \leqslant \tau^{-1}$. It is also easy to verify that the level shift $|\Delta\omega_q| \lesssim \tau^{-1}$. Therefore, it is clear that the solution is self-consistent.

In conclusion let us note the following circumstance. It can be seen from (3.1a) that, for $\omega = \omega_q$ and $q = q_0$, $\gamma(\omega, q)$ changes sign when $\xi = 2$, whereas N_{q0} becomes infinite as $\xi = 1$. In other words, at the considered threshold $\xi = 1$ the retarded Green function does not reveal an instability. A consequence of this is, as can be shown, using (3.2), the distinctive behavior of the noise, i.e., of the correlator

$$G_s(x, x') = -i\langle\varphi(x)\varphi(x') + \varphi(x')\varphi(x)\rangle, \quad x \equiv (\mathbf{r}, t). \tag{3.7}$$

Its Fourier transform, $G_s(\omega, \mathbf{q})$ is the frequency spectrum of the noise in the mode $\mathbf{q}$. As we approach the threshold of this mode, the total power of the noise in this mode increases like N_q, but the frequency form of the spectrum of the noise remains almost unchanged.

Narrow-band pumping: $\Delta\nu \ll \tau^{-1}$

For such pumping, we can seek the solution in which γ, $\Delta\omega$, and the characteristic interval of variation of $N(\omega, q)$ with respect to ω are greater than $\Delta\nu$. Then from (2.7a) and (2.7b) we can find the equations for the spectrum

$$\gamma(\omega, q) = \tau^{-1} - \mu\Delta(\nu_0-\omega, q), \tag{3.7a}$$

$$\Delta\omega(\omega, q) = \frac{1}{2\pi}\mu P(\nu_0-\omega, q), \tag{3.7b}$$

where $\mu = \frac{1}{2}\pi\lambda\Delta\nu$, and the following balance equation:

$$[\tau^{-1} - \mu\Delta(\nu_0-\omega, q)]N(\omega, q) = \mu\Delta(\nu_0-\omega, q)[N(\nu_0-\omega, q)+1]. \tag{3.8}$$

After writing Eq. (3.8) with the substitution $\omega \to \nu_0 - \omega$, we can solve the resulting pair of equations for $N(\omega, q)$ and find

$$f(\omega, q) \equiv \Delta(\omega, q)N(\omega, q) = \frac{\mu\Delta(\omega, q)\Delta(\nu_0-\omega, q)}{\tau^{-1} - \mu[\Delta(\omega, q) + \Delta(\nu_0-\omega, q)]}. \tag{3.9}$$

If we solve the equations for the spectrum, then we can find $\Delta(\omega, q)$, then $f(\omega, q)$ and

$$N_q = \int d\omega\, f(\omega, q). \tag{3.10}$$

Replacing ω by $\nu_0 - \omega$ in (3.7), we obtain a set of four algebraic equations for γ and $\Delta\omega$ in the arguments ω and $\nu_0 - \omega$. Let us go over to the dimensionless quantities

$$\begin{gathered} x = \tau\gamma(\omega, q), \quad u = \tau\gamma(\nu_0-\omega, q), \quad a = 2\tau(\omega-\omega_q), \\ y = 2\tau\Delta\omega(\omega, q), \quad v = 2\tau\Delta\omega(\nu_0-\omega, q), \quad b = 2\tau(\nu_0-\omega-\omega_q) \end{gathered} \tag{3.11}$$

and introduce

$$z = x + iy, \quad w = u - iv, \quad \eta = (2/\pi)\mu\tau^2. \tag{3.12}$$

Then the system assumes the form

$$z = 1 - \frac{\eta}{w+ib}, \quad w = 1 - \frac{\eta}{z-ia}. \tag{3.13}$$

It follows from (3.13) that

$$\frac{z-ia}{1-ia} = \frac{w+ib}{1+ib}. \tag{3.14}$$

Therefore, the solution can naturally be sought in the form

$$\begin{gathered} z - ia = \eta^{1/2}\left(\frac{1-ia}{1+ib}\right)^{1/2} s, \\ w + ib = \eta^{1/2}\left(\frac{1+ib}{1-ia}\right)^{1/2} s. \end{gathered} \tag{3.15}$$

Then for s we obtain the equation

$$s^2 - 2s\,\mathrm{ch}\,\theta + 1 = 0, \tag{3.16}$$

where

$$\mathrm{ch}\,\theta = [(1-ia)(1+ib)/4\eta]^{1/2}, \tag{3.17}$$

whence $s = e^{\pm\theta}$. Using (3.13), we can express the quantity f of interest to us in terms of s:

$$f(\omega, q) = \frac{1}{\mu\tau}\frac{(x-1)(u-1)}{x+u-1} = \frac{1}{\mu\tau}\frac{1}{|s|^4-1}.$$

Then

$$N_q = \frac{1}{\pi}\eta^{-1}\int da\,[\exp 4\vartheta(a, b) - 1]^{-1}, \tag{3.18}$$

where $\vartheta = \mathrm{Re}\,\theta$, and the integration is performed along the straight line $b + a = 2\tau(\nu_0 - 2\omega_q) \equiv 2c$ in the plane ab. The introduced quantity c is the dimensionless detuning of the mode q relative to the pump. It is clear that only the solution with $|s| > 1$ is preserved.

A finite solution for N_q exists when the integral (3.18) converges. A divergence can arise at each point where $\vartheta = 0$, i.e., $\cosh\theta = \cos \operatorname{Im}\theta$. It can be seen from (3.17) that this is possible only when $b = a$ and $a^2 + 1 < 4\eta$. Therefore, if $\eta < \frac{1}{4}$, then the integrand f is everywhere finite, and the integral converges (the convergence at infinity can easily be verified). If, on the other hand, $\eta > \frac{1}{4}$, then $f = \infty$ on the section of the straight line $a = b$ defined by the condition $|a| \leq (4\eta - 1)^{1/2}$.

Let us first consider the case $\eta = \frac{1}{4}$. In this case f has the singular point $a = b = 0$, in the vicinity of which, as can be computed,

$$\vartheta^2 = {}^1/{}_2 |a - b|. \tag{3.19}$$

For the noncentral modes ($c \neq 0$) the path of integration does not pass through the singular point. For the central mode ($c = 0$) the integration is performed along the straight line $b = -a$, where, according to (3.19),

$$\vartheta = |a|^{1/2}, \quad f \sim |a|^{-1/2}, \tag{3.20}$$

and therefore the integral also converges. Thus, for $\eta = \frac{1}{4}$ all the N_q are finite.

For $\eta > \frac{1}{4}$ the N_q are certainly finite for the considered modes, for which $|\omega_q - \omega_0| > \tau^{-1}(\eta - \frac{1}{4})^{1/2}$. In this case the path of integration does not cross the segment on which $f = \infty$. If, on the other hand, the opposite inequality is fulfilled, then such an intersection occurs at the point $a = b = c$. Expanding $\vartheta(a, b)$ about this point along the line of integration, we find

$$\vartheta = |a - c| \{(1 + c^2)[4\eta - (c^2 + 1)]\}^{-1/2}. \tag{3.21}$$

This means that the integral for N_q diverges logarithmically, i.e., that $\eta = \frac{1}{4}$ is a threshold for parametric instability.

For narrow-band pumping it is not possible to find the distribution in its explicit form. This can be done only for a weak pump, i.e., for $\eta \to 0$, when the distribution N_q turns out to be Lorentzian of width τ^{-1}. It is clear that the width cannot change in order of magnitude as we approach the threshold; for all the integrals converge and all the parameters are of the order of unity. For this reason, the values of N_q do not, in order of magnitude, exceed unity, and the concentration at the threshold is finite: $\overline{N} \approx a_0^{-3}(\tau^{-1}/\Omega_0)$. In this respect, the threshold for narrow-band pumping differs sharply from the threshold for broad-band pumping, in which the distribution narrows down and the concentration increases.

There exists above the threshold in phase space an unstable region that expands as we go higher above the threshold. For each mode the instability can be reached at

$$\eta_a^* = \frac{1}{4} + \frac{(\omega_q - \omega_0)^2}{\tau^{-2}} \tag{3.22}$$

This formula is quite similar to (3.6), and, in this respect, narrow-band pumping is similar to broad-band pumping.

Let us now compare the threshold intensities:

$$J^* = (\pi/4)(\hbar\nu_0\sigma/K)\Delta\nu\tau^{-1} \quad \text{(broad-band pumping)} \tag{3.23}$$

$$J^* = (\pi/8)(\hbar\nu_0\sigma/K)(\tau^{-1})^2 \quad \text{(narrow-band pumping)} \tag{3.24}$$

It can be seen that in the second case the threshold is determined by the total intensity J, while in the first case the threshold is virtually determined by the spectral density $J/\Delta\nu$. Physically, this is understandable; for the overlap of the contour of the pump line and the contour of the resonance curve of the phonon mode is considerable. The formulas (3.23) and (3.24) confirm the validity of the method used in the Introduction to estimate the threshold intensity, since it follows from the theory that at those intensities when $N \approx 1$, $\Delta\omega \approx \Delta\nu$ for broad-band pumping and $\Delta\omega \approx \tau^{-1}$ for narrow-band pumping.

Let us note in connection with the values of J^* that the threshold intensity obtained in[5] does not depend on $\Delta\nu$ and virtually coincides with (3.24). However, the given numerical value of 10^{19} W/cm^2 is incorrect (probably, this is the result of an arithmetical error); the correct value for nonresonance pumping for $\tau = 10^{-11}$ sec corresponds to the estimate 10^9 W/cm^2 made in the Introduction.

4. THE GENERATION $\nu \to TA - LA$ WITHOUT ALLOWANCE FOR THE LIMITING MECHANISM

Using (2.7a), we can write the balance equation in the following form:

$$N_1(\omega, q)\tau_1^{-1} = \lambda \int d\nu\, \varphi(\nu)\Delta_2(\nu - \omega, q)[N_1(\omega, q) + N_2(\nu - \omega, q) + 1]. \tag{4.1}$$

Multiplying by $\Delta_1(\omega, q)$ and integrating over ω, we find

$$N_1(q)\tau_1^{-1} = \lambda \int d\nu\, d\omega\, \varphi(\nu)\Delta_1(\omega, q)\Delta_2(\nu - \omega, q)[N_1(\omega, q) + N_2(\nu - \omega, q) + 1]. \tag{4.2}$$

Let us write down Eq. (4.2) for $N_2(q)$ and make the substitution $\omega \to \nu - \omega$ in it. Then the right-hand sides coincide, and we obtain

$$N_1(q)/\tau_1 = N_2(q)/\tau_2. \tag{4.3}$$

This means that irrespective of the nature of the pumping and the relationship between the lifetimes of the TA and LA phonons, their distributions over q have the same form, although the concentrations relate to each other as the lifetimes. It follows from this that there is a single threshold for the generation of both types of phonons.

Broad-band pumping: $\Delta\nu \gg \tau_2^{-1}, \tau_1^{-1}$

Proceeding in a manner similar to the case of identical phonons, we find

$$\gamma_1(\omega, q) = \tau_1^{-1} - \lambda\varphi(\omega + \omega_2(q)), \tag{4.4a}$$

$$B_1(\omega, q) = \lambda\varphi(\omega + \omega_2(q))[N_2(q) + 1] \tag{4.4b}$$

and then

$$N_1(\omega, q) = \frac{\xi_1[N_2(q) + 1]}{\varphi(\omega + \omega_2(q))^{-1} - \xi_1}, \tag{4.5}$$

$$N_1(q)=\frac{\xi_1[N_2(q)+1]}{\varphi(\omega_{12}(q))^{-1}-\xi_1} \quad (4.6)$$

where $\xi_1=\lambda\tau_1$. Writing down the equation, similar to (4.6), for $N_2(q)$, and solving the system, we find finally that

$$N_1(q)=\frac{\xi}{\varphi(\omega_{12}(q))^{-1}-\xi}\frac{\tau_1}{\tau_1+\tau_2}, \quad (4.7)$$

where $\xi=\xi_1+\xi_2=\lambda(\tau_1+\tau_2)$. It can be seen from this that the threshold is determined by the longer lifetime, while the width of the distribution over $\omega_{12}(q)$ is $\Delta\nu(1-\xi)^{1/2}$. If $\tau_1\gg\tau_2$, then for $\xi\approx 1$, i.e., in the case when the absorption nonlinearity is appreciable, we have $N_1\approx 1$ and $N_2\approx\tau_2/\tau_1\ll 1$. Like (3.3), the solution (4.7) can be obtained from the standard kinetic equation.

If τ_1 and τ_2 are of the same order of magnitude, then the conditions for the consistency of the found solution can be verified in the same way as in the case of identical phonons. However, if $\tau_1\gg\tau_2$, then some complications arise. The width of $N_1(\omega, q)$ with respect to ω is of the order of $\Delta\nu(1-\xi_1)^{1/2}$, as for N_2. If $\tau_1\approx\tau_2$, then $\xi_1\approx\xi_2$, and at the threshold $\xi_1+\xi_2=1$ both widths are of the order of $\Delta\nu$. If, on the other band, $\tau_1\gg\tau_2$, then $\xi_1\approx 1$ and $\xi_2\ll 1$ at the threshold. Therefore, the width of N_1 turns out to be smaller than $\Delta\nu$, and it is necessary to specially show that it remains greater than γ_1. The characteristic value (on the mass shell for the central mode) of $\gamma_1=\tau_1^{-1}-\lambda=\tau_1^{-1}(1-\xi_1)$. It can be seen from this that as $\xi_1\to 1$ the quantity γ_1 tends to zero more rapidly than the width N_1.

Narrow-band pumping: $\Delta\nu\ll\tau_1^{-1}$, τ_2^{-1}

The theory can be constructed in a manner similar to the case of the generation of identical phonons. On writing the system of equations in a dimensionless form, we find that the dimensionless pump parameter is $\eta=(2/\pi)\mu\tau_1\tau_2$. Further, we find that

$$N_1(q)=\tau_1\int d\omega\, F(a, b; \eta), \quad (4.8)$$

where F is some function of only the indicated parameters. In this case

$$a=2\tau_1(\omega-\omega_1(q)),\quad b=2\tau_2(\nu_0-\omega-\omega_2(q)), \quad (4.9)$$

so that the integration in the plane ab is performed along the straight line

$$a/\tau_1+b/\tau_2=2(\nu_0-\omega_{12}(q)). \quad (4.10)$$

If $\tau_1\gg\tau_2$, then we obtain

$$N_1(q)=\frac{1}{2}\int da\, F(a, 2\tau_2(\nu_0-\omega_{12}(q)); \eta). \quad (4.11)$$

It is clear from this that in the case of an appreciable absorption nonlinearity, when $\eta\approx 1$, and also at the very threshold the width of the distribution $N_1(q)$ is of the order of τ_2^{-1} and the occupation number of the central mode $N_1(q_0)\approx 1$. In this case the width of the distribution $N_2(q)$ is the same, but $N_2(q_0)\approx\tau_2/\tau_1\ll 1$. This means that LA phonons are virtually not generated, and the concentration of the TA phonons at the threshold is of the order of $a_0^{-3}\tau_2^{-1}/\Omega_0$.

Intermediate pumping: $\tau_1^{-1}\ll\Delta\nu\lesssim\tau_2^{-1}$

Let us seek the solution in which γ_1 is less than all the widths and the renormalization of the spectrum of the phonon 2 can be neglected. Then from (2.1) and (2.7) we obtain for the LA phonons the expressions

$$\gamma_2(\omega, q)=\tau_2^{-1}, \quad (4.12)$$

$$B_2(\omega, q)=\lambda\varphi(\omega+\omega_1(q))[N_1(q)+1], \quad (4.13)$$

$$N_2(\omega, q)=\tau_2 B_2(\omega, q). \quad (4.14)$$

The discarded correction to γ_2 is of the order of λ. Therefore, it is assumed that $\lambda\ll\tau_2^{-1}$. Further, from (2.7a) we find for the TA phonon that

$$\gamma_1(\omega, q)=\tau_1^{-1}-g(\omega, q), \quad (4.15)$$

where

$$g(\omega, q)=\lambda\int d\nu\,\varphi(\nu)\,\delta(\tau_2^{-1}|\nu-\omega-\omega_2(q))$$
$$=\lambda\frac{{}^1/_2\Delta\nu[{}^1/_2(\Delta\nu+\tau_2^{-1})]}{[\omega+\omega_2(q)-\nu_0]^2+[{}^1/_2(\Delta\nu+\tau_2^{-1})]^2}. \quad (4.16)$$

Substituting (4.13) and (4.14) into (2.7c), we find for the TA phonon that

$$B_1(\omega, q)=g(\omega, q)+[N_1(q)+1]b(\omega, q), \quad (4.17)$$

where

$$b(\omega, q)=\lambda^2\tau_2\int d\nu\,\varphi(\nu)\,\delta(\tau_2^{-1}|\nu-\omega-\omega_2(q))\,\varphi(\nu-\omega+\omega_1(q)). \quad (4.18)$$

By making estimates in the case when $\Delta\nu\lesssim\tau_2^{-1}$, we can show that $g\approx\lambda\tau_2\Delta\nu$ and $b\approx\lambda^2\tau_2^2\Delta\nu$. Therefore, under the assumption made above we can drop the second term in B_1 and find from (2.1) for the TA phonon the distribution

$$N_1(\omega, q)=g(\omega, q)/[\tau_1^{-1}-g(\omega, q)]. \quad (4.19)$$

The width of $N_1(\omega, q)$ with respect to ω near the threshold decreases, but, as in the corresponding case discussed above, we can show that γ_1 decreases faster. Therefore, evaluating (4.19) on the mass shell, we find

$$N_1(q)=[\tau_1^{-1}g(q)^{-1}-1]^{-1}, \quad (4.20)$$

where $g(q)$ is $g(\omega, q)$ with $\omega=\omega_1(q)$. The threshold is determined by the point where $N_1(q)$ for the central mode q_0 becomes infinite. From (4.16), we find

$$g(q_0)=\lambda\Delta\nu/(\Delta\nu+\tau_2^{-1}), \quad (4.21)$$

and the threshold value of λ is

$$\lambda^*=\tau_1^{-1}(\Delta\nu+\tau_2^{-1})/\Delta\nu. \quad (4.22)$$

It is obvious that $\lambda^*\ll\tau_2^{-1}$ and, therefore, the found threshold lies in the region where the assumptions made are valid. In dimensional quantities

$$I^* = \left(\frac{\pi}{2}\right)\left(\frac{\hbar\nu_0\sigma_{12}}{K}\right)\tau_1^{-1}(\Delta\nu+\tau_2^{-1}). \quad (4.23)$$

It can be seen that in the general case the threshold is not expressible in terms of the total intensity, or in terms of the spectral intensity. Only in the case when $\Delta\nu \ll \tau_2^{-1}$ is the threshold expressible in terms of the total intensity. From the "point of view" of the *TA* phonon, the wings of the pump are useless, but they are "used" by the *LA* phonon.

By substituting into (4.20) the explicit expression found from (4.16) for $g(q)$, we can verify that the distribution $N_1(q)$ has a Lorentzian character with respect to $\omega_{12}(q) - \nu_0$ and that its parameters are

$$\Delta\omega = (\Delta\nu+\tau_2^{-1})(1-\xi)^{1/2}, \quad (4.24)$$

$$N_1(q_0) = \xi(1-\xi)^{-1}, \quad (4.25)$$

where $\xi = \lambda/\lambda^*$. The total *TA*-phonon concentration

$$\bar{N}_1 = (\pi/2)\sigma_{12}(\Delta\nu+\tau_2^{-1})\xi(1-\xi)^{-1/2}. \quad (4.26)$$

The results pertaining to the parametric generation of the various particles show that in this case the threshold intensity lies in that region where the occupation numbers of the longer-lived particle become of the order of unity.

5. THE EFFECT OF THE LIMITING MECHANISM

In order to obtain a finite value for N_q at the threshold and obtain a solution above the threshold, we should include a limiting mechanism, and, as this mechanism, we shall consider the merging of *TA* phonons. Let us first consider the generation $\nu \to 2TA$ in broad-band pumping.

According to (2.13), allowance for the merging is equivalent to the replacement of τ^{-1} by $\tau^{-1} + \alpha\bar{N}$, i.e., the replacement of ξ by $\xi(1+\alpha\tau\bar{N})^{-1}$. Replacing ξ in (3.5) by the last quantity, we obtain an equation for $\bar{N}$. However, it is convenient to transform this equation into an equation for N_0, using the relations

$$\Delta\omega = {}^1/_2\Delta\nu(2N_0+1)^{1/2}, \quad \bar{N} = \pi\Delta\nu\sigma N_0(2N_0+1)^{-1/2}. \quad (5.1)$$

The equation for N_0 will be

$$\chi\frac{N_0}{(2N_0+1)^{1/2}} = \xi\frac{2N_0+1}{2N_0} - 1, \quad (5.2)$$

where $\chi = \pi\alpha\tau\Delta\nu\sigma$ is a dimensionless parameter responsible for the limiting mechanism. We shall assume this parameter to be small, so that the threshold will be sufficiently sharp. This means that

$$\chi \approx (\Gamma_0/\Omega_0)(\Delta\nu/\tau^{-1}) \ll 1. \quad (5.3)$$

For $\chi \ll 1$ we obtain at the threshold $\xi = 1$ the relations

$$N_0 \approx \chi^{-2/3} \gg 1, \quad \Delta\omega \approx \Delta\nu\chi^{1/3} \ll \Delta\nu. \quad (5.4)$$

Beyond the threshold in the region where $\xi - 1 \gtrsim 1$

$$N_0 = 2\chi^{-2}(\xi-1)^2, \quad \Delta\omega = {}^1/_4\Delta\nu\chi(\xi-1)^{-1}, \quad (5.5)$$

i.e., the distribution continues to narrow down and increase.

Let us now proceed to consider the generation $\nu \to 2TA$ in narrow-band pumping. It can be seen from (2.13) and (2.14) that allowance for the anharmonicity is equivalent to the renormalization of the relaxation time and the dispersion law:

$$\tau^{-1} \to \tilde{\tau}^{-1} \equiv \tau^{-1} + \alpha\bar{N}, \quad \omega_q \to \tilde{\omega}_q \equiv \omega_q - \frac{\beta}{2\pi}\bar{N}. \quad (5.6)$$

We can, in analogy to (3.11) and (3.12), introduce dimensionless variables, which will contain $\tilde{\tau}$ and $\tilde{\omega}_q$. Assuming that the solution for the spectrum is known, we can compute the concentration

$$\bar{N} = \frac{\rho}{4\pi\eta\tau}\iint da\,db\frac{1}{|s|^4-1} = \rho\tilde{\tau}^{-1}\Phi(\tilde{\eta}). \quad (5.7)$$

Here ρ is the one-phonon density of states at $\omega = \omega_0$ and Φ depends only on the indicated argument. It follows from the results of Sec. 3 that this function is defined only for $\tilde{\eta} < \frac{1}{4}$ and that at these values it is bounded by values of the order of unity; $\Phi(\tilde{\eta})$ is small for small $\tilde{\eta}$ and of the order of unity for $\tilde{\eta} \approx 1$.

The equality (5.7) can be understood as an equation for $\bar{N}$, since the dependence of $\tilde{\tau}$ and $\tilde{\eta}$ on $\bar{N}$ is known. As in the case of broad-band pumping, it is more convenient to go over to the dimensionless quantity N with the aid of the relation $\bar{N} = \rho\tau^{-1}N$. Then

$$\tilde{\tau}^{-1} = \tau^{-1}(1+\chi N), \quad \tilde{\eta} = \eta/(1+\chi N)^2, \quad \chi = \alpha\rho, \quad (5.8)$$

and the equation assumes the following form:

$$\frac{N}{1+\chi N} = \Phi\left(\frac{\eta}{(1+\chi N)^2}\right) \quad (5.9)$$

Using the estimate for α given in Sec. 2, we find $\chi \approx \Gamma_0/\omega_0 \ll 1$. It follows from this that (5.9) does not have solutions for $N \gg 1$. On the other hand, for $N \approx 1$ the left-hand side is of the order of unity, and therefore $\tilde{\eta} \approx \eta \approx 1$. Thus, Eq. (5.9) does not possess solutions with $\eta \gg 1$. If, on the other hand, $\eta \approx 1$, then the solution differs little from the solution with $\chi = 0$. Thus, the anharmonicity as a limiting mechanism in narrow-band pumping is ineffective. It does not enable us to go far beyond the threshold, but leads only to a small change in the solution and a small renormalization of the threshold.

The situation in the $\nu \to TA + LA$ generation by narrow-band and broad-band pumping does not differ from the situation in the $\nu \to 2TA$ generation. The intermediate pumping is analogous to the broad-band pumping, which fact can be seem from a comparison of (4.26) and (3.5). Therefore, the anharmonicity as a limiting mechanism is effective, although the meaning of the parameter χ will be different.

6. CRITERIA

In solving the problem of parametric generation, we assumed that those modes that are not directly excited by the field are in equilibrium and that the renormalization of the spectrum of these modes can be neglected. For these assumptions to be valid, it is necessary that the number of acoustic phonons excited by the light per unit cell be small: $\bar{N}a_0^3 \ll 1$, or, expressed in another form,

$$N_0\Delta\omega/\Omega_0 \ll 1. \qquad (6.1)$$

For this condition to be fulfilled, the rate, γ_a, at which the acoustic phonons merge into "equilibrium" modes should satisfy the relation $\gamma_a \approx \Gamma_0 \bar{N}a_0^3 \ll \Gamma_0$; this follows from (2.13) and the subsequent estimate for α. Thus, this rate is less than the rate of decay of the "equilibrium" modes and, therefore, the phonon concentration in the "equilibrium" modes is small compared to $\bar{N}$. As to the renormalization of the spectrum of the "equilibrium" modes, it can be estimated from formulas similar to (2.13) and (2.14), into which will enter the total concentration of all the phonons, i.e., virtually $\bar{N}$. It is clear that the fulfillment of (6.1) guarantees the smallness of the renormalization.

It can be shown that the fulfilment of the inequality (6.1) is sufficient for all the purely phonon polarization operators (i.e., without light lines) to be considered in the lowest order. This is precisely what is assumed in the derivation of the generalized kinetic equations.

For diagrams with light lines the retention of only the simplest diagrams requires the fulfilment of the criterion $N_0(\Gamma_0/\Omega_0) \ll 1$ (the smallness of the corrections to the light vertex). It is important to emphasize, however, that near the threshold, where cancellation of the dominant generation and phonon-relaxation terms occurs, the last criterion is replaced by the more rigid condition:

$$N_0(\Gamma_0/\Omega_0)^{1/2} \ll 1. \qquad (6.2)$$

Assuming, as above, for orientation that $\Delta\omega \approx \Gamma_0$, we obtain that in the framework of the theory we can attain $N_0 \lesssim 10$ and $\bar{N} \lesssim 10^{20}$ cm^{-3}, which, however, implies the existence of a state of strong nonequilibrium at low temperatures.

The conditions (6.1) and (6.2) lead to the conclusion that the solutions found in Sections 3 and 4 for broad-band pumping without allowance for the limiting mechanism become invalid fairly close to the threshold. If, however, we include the limiting mechanism, then in order for (6.1) and (6.2) to be fulfilled, it should be fairly strong. Thus, for example, as applied to the allowance for the merging of the TA phonons in the generation $\nu \to 2TA$ (Sec. 5), this means that $\chi \gg (\Gamma_0/\Omega_0)^{1/4}$. Together with (5.3) this gives

$$\Gamma_0/\Omega_0 \ll \tau^{-1}/\Delta\nu \ll (\Gamma_0/\Omega_0)^{1/4}. \qquad (6.3)$$

This is a very rigid criterion, and for the values $\Delta\nu = 1$ cm^{-1} and $\tau = 10^{-9}$ sec used it is almost violated. Therefore, at these values of the parameters we can count on only a qualitative description.

On the other hand, it should be noted that all the estimates given in the paper are purely tentative. It should be borne in mind that allowance for the numerical factors and the use of real crystal parameters can easily change any estimate by one, two and sometimes even three orders of magnitude in either direction.

In order that the light field can be assumed to be given, the thickness, d, of the sample should be less than $\tilde{K}^{-1}$, where $\tilde{K}$ is the coefficient of nonlinear absorption. It can be computed with the aid of the relation $\tilde{K} = Q/J$, where $Q = \hbar\omega_0\bar{N}/\bar{\tau}$ is the power delivered to the thermostat. Using, for example, (3.5), we can verify that $\tilde{K} \approx KN_0$, so that we should have $d \lesssim 10^{-2}$ cm.

7. GENERATION BY BEATS

A very promising method of exciting phonons is the method of beats of two laser beams whose frequencies ν_1 and ν_2 lie in the optical range, while the frequency difference $\nu = \nu_1 - \nu_2$ lies in the infrared region.[14] If $|\bar{\nu} - \Omega_0| \lesssim \Gamma_0$, we deal with resonance pumping, while if $|\bar{\nu} - \Omega_0| \gg \Gamma_0$, we deal with nonresonance pumping. This method requires high light intensities, since it uses a higher-order (in the photons) process. However, a great advantage of this method is the possibility of exciting phonons uniformly in the interior of the sample, since optical radiation with a quantum smaller than the width of the forbidden band in a dielectric is practically not absorbed.

The theory of excitation by beats can be constructed literally by the same method used in the case of excitation during absorption. In the equations only the meaning of the quantity $\lambda\varphi(\nu)$ changes. The quantity λ turns out to be proportional to the product, J_1J_2, of the beam intensities, while φ has the meaning of the convolution of the spectral contours of these beams:

$$\varphi(\nu) \sim \int d\nu' \int d\nu'' \varphi_1(\nu')\varphi_2(\nu'')\delta(\nu' - \nu'' - \nu). \qquad (7.1)$$

If, as before, we normalize φ such that $\max\varphi = 1$, then we can find for λ the following estimate (assuming that the beams do not differ greatly from each other):

$$\lambda \approx J^2 w^{(2)} c a_0^3/\nu^4\Delta\nu, \qquad (7.2)$$

where $w^{(2)}$ is the total probability of two-phonon Raman scattering. We do not know the experimental data for $w^{(2)}$. Literal estimates give $w^{(2)} \approx (s/c)^3(\Omega_0^3/\Gamma_0^2)$. Using this estimate and (3.23), (3.24), we can obtain an order-of-magnitude estimate for the ratio of the threshold intensities for generation by absorption and by beats (during nonresonance pumping). This ratio

$$\frac{J^*\,(\text{absorption})}{J^*\,(\text{beats})} \approx \begin{cases} \dfrac{(\Delta\nu\tau^{-1})^{1/2}}{\Gamma_0}\dfrac{\Gamma_0}{\Omega_0} & \text{(broad-band pumping)} \qquad (7.3) \\[2ex] \dfrac{\tau^{-1}}{\Gamma_0}\dfrac{\Gamma_0}{\Omega_0} & \text{(narrow-band pumping)} \qquad (7.4) \end{cases}$$

Here we have used the estimate $K \approx \Gamma_0/c$, which follows from the results of the paper[15]. It can be seen from the estimates obtained that for $\tau = 10^{-9}$ sec generation by beats requires intensities four-five orders of magnitude higher than the intensities required for generation by absorption, i.e., 10^{11}–10^{12} W/cm^2.

It is interesting to compare the characteristic intensities of resonance and nonresonance pumps. This can be done for broad-band pumping, using the result of the paper[4] for J^* in the case of resonance pumping by beats:

$$\frac{J^*\,(\text{resonance beats})}{J^*\,(\text{nonresonance beats})} \approx \frac{\Gamma_0}{\Omega_0}. \qquad (7.5)$$

The analogous ratio for absorption is, as follows from (3.23), simply the ratio of the coefficients of two-phonon and one-phonon resonance absorption:

$$\frac{J^*\,(\text{resonance absorption})}{J^*\,(\text{nonresonance absorption})} \approx \left(\frac{\Gamma_0}{\Omega_0}\right)^2, \qquad (7.6)$$

i.e., the resonance and nonresonance pump efficiencies differ less in generation by beats than in generation by absorption.

The authors express their gratitude to S. V. Iordanskiĭ and V. L. Pokrovskiĭ for a discussion of some aspects of the paper.

[1]R. Orbach, Phys. Rev. Lett. 16, 15 (1966); IEEE Trans. SU-14, 140, 1967.
[2]R. Orbach and L. A. Vredevoe, Physics 1, 91 (1964).
[3]M. J. Colles and J. A. Giordmaine, Phys. Rev. Lett. 27, 670 (1971).
[4]I. B. Levinson, Zh. Eksp. Teor. Fiz. 65, 331 (1973) [Sov. Phys. JETP 38, 162 (1974)].
[5]M. Sparks and H. C. Chow, Phys. Rev. B10, 1699 (1974).
[6]E. Burstein, J. J. Oberley, and E. K. Pyler, Proc. Ind. Acad. Sci. 28, 388 (1948); M. Lax and E. Burstein, Phys. Rev. 97, 39 (1955).
[7]D. A. Kleinman and W. G. Spitzer, Phys. Rev. 118, 110 (1960); E. Burstein, in: Phonons and Phonon Interactions (ed. Thor A. Bak), W. A. Benjamin, New York, Amsterdam, 1964, p. 276.
[8]A. F. Dite, V. B. Timofeev, V. M. Faĭn, and É. G. Yashchin, Zh. Eksp. Teor. Fiz. 58, 460 (1970) [Sov. Phys. JETP 31, 245 (1970)].
[9]J. W. Jack, H. W. T. Baron, and T. Smith, J. Phys. C7, 2961 (1974).
[10]S. A. Bulgadaev and I. B. Levinson, Zh. Eksp. Teor. Fiz. 67, 2341 (1974) [Sov. Phys. JETP 40, 1161 (1975)].
[11]V. E. Zakharov, V. S. L'vov, and S. S. Starobinets, Usp. Fiz. Nauk 114, 609 (1974) [Sov. Phys. Usp. 17, 896 (1975)].
[12]L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1515 (1964) [Sov. Phys. JETP 20, 1018 (1965)].
[13]R. M. White and M. Sparks, Phys. Rev. 130, 632 (1963).
[14]G. Chartier and S. Birand, Phys. Rev. Lett. 21, 1641 (1968).
[15]I. B. Levinson and É. I. Rashba, Pis'ma Zh. Eksp. Teor. Fiz. 20, 63 (1974) [JETP Lett. 20, 27 (1974)].

Translated by A. K. Agyei.

Polaritons in inhomogeneous crystals

V. I. Sugakov and V. N. Khotyaintsev

Kiev State University
(Submitted November 25, 1975)
Zh. Eksp. Teor. Fiz. 70, 1566–1574 (April 1976)

An exact solution of the problem of the motion of a polariton in an inhomogeneous crystal is found with allowance for spatial dispersion, if the energy of the bottom of the exciton band (optical phonons) linearly depends on the coordinates. Outside the region of the turning point for excitons, the polaritons represent either excitons or electromagnetic waves. In the region of the turning point, where the effects of the mixing of exciton and electromagnetic waves are large, transformation of some waves into others takes place, where the efficiency of such a conversion process depends on the degree of inhomogeneity. The properties of electromagnetic waves upon propagation in the directions of increasing and decreasing energy of the bottom of the band are found to be different, a reflected wave being present in the first case and absent in the second. The difference is also manifest in the dependence of the fraction of electromagnetic wave energy transformed into exciton energy on the direction of motion. The non-equivalence of opposite directions becomes very pronounced in the case of a gradual inhomogeneity; for one direction of propagation the electromagnetic wave is completely reflected, and for the other direction it is completely transformed into excitons (optical phonons). The latter process may be utilized for the generation of a coherent exciton beam. The physical nature of the phenomenon is explained and criteria are discussed for the applicability of the results to inhomogeneities of another type.

PACS numbers: 71.85.Ce

The problem of determining the spectrum of polaritons in an inhomogeneous crystal has a number of characteristic features distinguishing it from the corresponding problem in a homogeneous medium. From a macroscopic point of view, the distinctive feature consists in the fact that in the presence of spatial dispersion it is impossible to characterize an inhomogeneous medium by a dielectric constant which depends on the wave vector. However, if the effects of spatial dispersion are unimportant, one can introduce a dielectric constant that depends on the frequency and coordinates, but has in the polariton part of the spectrum singulari-

