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Spontaneous symmetry-breaking in a gas of nonequilibrium phonons

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Irradiation of a dielectric by nonresonance infrared light leads to excitation of short-wavelength phonon modes (two-phonon absorption). It is shown that there exists an intensity threshold above which spontaneous lowering of the symmetry occurs in the gas of nonequilibrium short-wavelength phonons—the stable state of the gas is one in which the phonon distribution function is of lower symmetry than the crystal.

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1. THE MODEL AND KINETIC EQUATION

An isotropic model of a crystal with a center of inversion is considered; in the crystal there are two acoustic branches (a transverse (TA) and a longitudinal (LA) branch) and several optical branches (O) (see Fig. 1). The crystal is at a low temperature $T \ll \omega_D$, where ω_D is the Debye frequency.

The frequency ν of the incident light does not coincide with any of the limiting ($q=0$) frequencies ω_0 of the optical phonons active in infrared absorption. In this case the absorption is associated with the creation of a pair of short-wavelength phonons (usually acoustic) and proceeds according to the scheme

$$\nu \to TA + LA; \qquad (1.1)$$

the frequencies of the phonons created are of the order of ω_D.

The LA phonons created are rapidly thermalized in spontaneous decay processes, and therefore their occupation numbers can be assumed equal to zero. Spontaneous decay of the TA phonons is impossible.[1] Therefore, they can be destroyed either by scattering by defects with the conversion TA → LA[2] or by interaction between nonequilibrium TA phonons. In lowest order in the anharmonicity the latter corresponds to the coalescence process

$$TA + TA \to O. \qquad (1.2)$$

O-branch phonons are also rapidly thermalized and if we assume their occupation numbers to be equal to zero the coalescence of two TA phonons is equivalent to their destruction.

The kinetic equation for the occupation numbers of the TA phonons can be written in the following form:

$$\dot{N}(\mathbf{q}) = D(\mathbf{q}) + G(\mathbf{q}). \qquad (1.3)$$

The term G describes the generation of TA phonons by the light and the term D describes the destruction of these phonons.

We shall consider first the generation term G, assuming, as in[3], that the spectral intensity of the exciting light is given by a Lorentzian form factor

$$\varphi(\nu) = \frac{(\Delta\nu/2)^2}{(\nu-\nu_0)^2 + (\Delta\nu/2)^2}, \quad \varphi(\nu_0) = 1 \qquad (1.4)$$

with central frequency ν_0 and width $\Delta\nu$. We then have[6]

$$G(\mathbf{q}) = \lambda\varphi(q)[N(\mathbf{q})+1], \qquad (1.5)$$

where

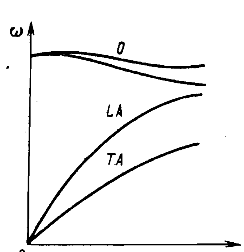

FIG. 1.

$$\lambda=2\pi^{-1}(JK/\sigma v_0\Delta v), \tag{1.6}$$

(where J is the intensity of the exciting light, K is the (linear) two-phonon absorption coefficient, and σ is the two-phonon density of states) and

$$\varphi(q)=\varphi(\omega_{TA}(\mathbf{q})+\omega_{LA}(\mathbf{q})). \tag{1.7}$$

Because of the form factor $\varphi(q)$, the TA phonons are excited near the surface of a sphere with radius q_0 determined from the equation $\varphi(q_0)=1$. The frequencies of these phonons are close to $\omega_0\equiv\omega_{TA}(q_0)$.

We turn now to the term D. Since there are two destruction processes for TA phonons,

$$D(\mathbf{q})=D_1(\mathbf{q})+D_2(\mathbf{q}), \tag{1.8}$$

where

$$D_1(\mathbf{q})=-\tau^{-1}N(\mathbf{q}) \tag{1.9}$$

and

$$D_2(\mathbf{q})=-\int d\mathbf{q}'W(\mathbf{s},\mathbf{s}')N(\mathbf{q})N(\mathbf{q}'). \tag{1.10}$$

Here τ is the time of "spontaneous" destruction of TA phonons, e.g., in the conversion TA→LA; it can be assumed to be independent of q in the small range of momenta near q_0 that is important for us. W is the probability of coalescence of two TA phonons with momenta $\mathbf{q}$ and $\mathbf{q}'$; it can be assumed to depend only on the directions of these momenta, specified by the unit vectors $\mathbf{s}$ and $\mathbf{s}'$.

2. THE STATIONARY SOLUTION AND THE CRITERION FOR ITS STABILITY

For convenience we shall introduce dimensionless quantities of various kinds. In place of q we introduce

$$x=(q-q_0)(s_{TA}+s_{LA})/(\Delta v/2), \tag{2.1}$$

where the s are the group velocities of the corresponding phonons. Then

$$\varphi(q)=\varphi(x)=(x^2+1)^{-1}. \tag{2.2}$$

In place of $W(\mathbf{s},\mathbf{s}')$ we introduce the dimensionless kernel $K(\mathbf{s},\mathbf{s}')$, such that

$$u(\mathbf{s})=\tau\int d\mathbf{q}'W(\mathbf{s},\mathbf{s}')N(\mathbf{q}')=\frac{\beta}{\pi}\int\frac{do'}{4\pi}K(\mathbf{s},\mathbf{s}')\int dx'N(x',\mathbf{s}'). \tag{2.3}$$

Here do is the element of solid angle and the dimensionless constant β is chosen in such a way that

$$\int\frac{do'}{4\pi}K(\mathbf{s},\mathbf{s}')=1. \tag{2.4}$$

In order of magnitude, $\beta\approx(\Gamma_0/\omega_D)(\Delta\nu/\tau^{-1})$, where Γ_0 is the typical width for third-order processes in which phonons with frequency of the order of the Debye frequency participate. In this notation the kinetic equation takes the form

$$\tau\dot{N}(x,\mathbf{s})=-[1-\xi\varphi(x)+u(\mathbf{s})]N(x,\mathbf{s})+\xi\varphi(x), \tag{2.5}$$

where $\xi=\lambda\tau$ is the dimensionless pumping parameter.

The stationary "solution" of Eq. (2.5)

$$N(x,\mathbf{s})=\frac{\xi\varphi(x)}{1-\xi\varphi(x)+u(\mathbf{s})}=\frac{\xi}{1+u(\mathbf{s})}\,\frac{1}{x^2+[(1+u(\mathbf{s})-\xi)/(1+u(\mathbf{s}))]} \tag{2.6}$$

is a Lorentzian distribution in $q-q_0$ with a width and height that depend on direction. Substituting (2.6) into (2.3) and integrating over x we find a nonlinear integral equation for u:

$$u(\mathbf{s})=\int\frac{do'}{4\pi}K(\mathbf{s},\mathbf{s}')\Phi(\xi;u(\mathbf{s}')), \tag{2.7}$$

where

$$\Phi(\xi;u)=\beta\xi[(1+u)(1+u-\xi)]^{-1/2}. \tag{2.8}$$

When $u(\mathbf{s})$ has been found from this equation we can find the stationary phonon distribution $N(x,\mathbf{s})$.

Next comes the question of the stability of this distribution. We represent the solution of the nonstationary equation in the form

$$N(x,\mathbf{s})+\delta N(x,\mathbf{s})e^{-pt/\tau} \tag{2.9}$$

and linearize (2.5) in δN about the stationary solution N. Then, substituting (2.9) into (2.3), we find

$$u(\mathbf{s},t)=u(\mathbf{s})+\delta u(\mathbf{s})e^{-pt/\tau}, \tag{2.10}$$

where u corresponds to the stationary solution N, and

$$\delta u(\mathbf{s})=\frac{\beta}{\pi}\int\frac{do'}{4\pi}K(\mathbf{s},\mathbf{s}')\int dx'\delta N(x',\mathbf{s}'). \tag{2.11}$$

On the other hand, after the linearization we obtain from Eq. (2.5)

$$\delta N(x,\mathbf{s})=-N(x,\mathbf{s})\frac{\delta u(\mathbf{s})}{1-p-\xi\varphi(x)+u(\mathbf{s})}. \tag{2.12}$$

Substituting (2.12) into (2.11) and integrating over x we obtain a linear integral equation for δu:

$$p\delta u(\mathbf{s})=\int\frac{do'}{4\pi}K(\mathbf{s},\mathbf{s}')[\Phi(\xi;u(\mathbf{s}'))-\Phi(\xi;u(\mathbf{s}')-p)]\delta u(\mathbf{s}'). \tag{2.13}$$

The conditions for solubility of this equation determine p. The distribution N is unstable if Eq. (2.13) is soluble for $p<0$. We remark that in this case the integral over x that arises certainly converges.

3. THE ISOTROPIC DISTRIBUTION AND ITS STABILITY

It is natural to seek first an isotropic stationary solution, when $u(\mathbf{s})=\text{const}$. Then, using (2.4), we obtain from (2.7) an algebraic equation for u:

$$u=\Phi(\xi;\, u). \tag{3.1}$$

For each ξ this equation has one real and positive solution $u(\xi)$, increasing monotonically with increase of ξ. Therefore, for each ξ there exists a single isotropic distribution. We shall consider its stability.

For the isotropic stationary solution the expression in square brackets in (2.13) can be taken outside the integral. Since the kernel K depends only on the angle between $\mathbf{s}$ and $\mathbf{s}'$, it can be represented in the following form:

$$K(\mathbf{s},\mathbf{s}')=4\pi\sum_{lm} k_l Y_{lm}(\mathbf{s})Y_{lm}^*(\mathbf{s}'), \tag{3.2}$$

where the Y_{lm} are spherical harmonics (with phase and normalization as in[4]). We note that (2.4) is then equivalent to $k_0=1$. It follows from (3.2) that on linearization about the isotropic stationary distribution the solutions of (2.13) are spherical harmonics and the solubility condition gives a series of independent algebraic equations

$$p=k_l[\Phi(\xi;\, u)-\Phi(\xi;\, u-p)], \tag{3.3}$$

in which we must substitute $u(\xi)$ for u. Then from (3.3) the solutions $p=p_l(\xi)$ are obtained. The isotropic distribution is unstable for a given ξ if $p_l<0$ holds for some l. This is possible only in the case when $k_l<0$. But if $k_l>0$, Eq. (3.2) cannot be satisfied by a negative p, since $\Phi(\xi; u)$ falls off with increase of u.

The threshold of instability is determined by the condition $p_l(\xi)=0$. When $p\to 0$ Eq. (3.3) is transformed into

$$\frac{1}{|k_l|}=-\frac{\partial}{\partial u}\Phi(\xi;\, u(\xi))\equiv\Psi(\xi), \quad k_l<0. \tag{3.4}$$

Solving this equation we find ξ_l^*—the threshold for loss of stability against distribution-function fluctuations that depend on the direction of $\mathbf{q}$, such as $Y_{lm}(\mathbf{s})$.

Thus, the question of the stability of the isotropic stationary solution turns out to be connected with the question of the existence of negative eigenvalues of the kernel K. Therefore, we shall consider the properties of this kernel in somewhat more detail. The coalescence (1.2) is possible only in the case when the momenta of the TA phonons form a definite angle χ, uniquely determined by the conservation laws:

$$2\omega_0=\omega_O(\mathbf{s}q_0+\mathbf{s}'q_0), \quad \cos\chi=\mathbf{s}\mathbf{s}', \tag{3.5}$$

where $\omega_O(q)$ is the O-phonon dispersion law. Therefore,

$$K(\mathbf{s},\,\mathbf{s}')=2\delta(\mathbf{s}\mathbf{s}'-\cos\chi), \tag{3.6}$$

$$k_l=P_l(\cos\chi). \tag{3.7}$$

The coefficient of the δ-function is chosen in accordance with the normalization (2.4).

It follows from the properties of Legendre polynomials that we can always find an l for which $k_l<0$, and that $|k_l|\leqslant 1$ for all l. If coalescence into several different optical branches is possible, there is an angle χ_i and a value β_i corresponding to each channel. If we choose

$$\beta=\sum_i \beta_i, \tag{3.8}$$

then

$$k_l=\langle P_l(\cos\chi_i)\rangle, \tag{3.9}$$

where $\langle\ldots\rangle$ denotes averaging over the coalescence channels, with weights β_i/β. It is obvious that $|k_l|\leqslant 1$ in this case too. The existence of negative k_l follows from the equality

$$K(\mathbf{s},\mathbf{s})=\sum_l (2l+1)k_l=0, \tag{3.10}$$

provided that, amongst the angles χ_i, there is no chance angle $\chi_i=0$. Now it is possible to show, starting from the known properties of the kernel K, that Eq. (3.4) necessarily has a solution, and only one at that. By direct calculation it is easily verified that

$$\Psi(\xi)>0, \quad \frac{d}{d\xi}\Psi(\xi)>0, \quad \lim_{\xi\to\infty}\frac{d}{d\xi}\Psi(\xi)=1 \tag{3.11}$$

and

$$\Psi(1)=\frac{1}{2}\left[1+\frac{u}{1+u}\right]<1, \quad u\equiv u(1). \tag{3.12}$$

Thus, starting from a certain value less than unity at $\xi=1$, $\Psi(\xi)$ increases monotonically and without limit with increase of ξ. Since $|k_l|\leqslant 1$, Eq. (3.4) necessarily has a solution $\xi_l^*>1$, and with increase of $|k_l|$ the threshold value ξ_l^* falls. This means that the lowest threshold corresponds to the negative value of k_l with the largest modulus. Differentiating (3.3) and letting ξ tend to ξ_l^*, we can show that

$$\frac{d}{d\xi}p_l(\xi_l^*)<0. \tag{3.13}$$

Thus, for $\xi<\xi_l^*$ we have $p_l(\xi)>0$ and the isotropic distribution is stable, while for $\xi>\xi_l^*$ we have $p_l(\xi)<0$ and the isotropic distribution is unstable.

Explicit expressions for the threshold can be found for the limiting values of the coalescence parameter:

$$\begin{aligned} &\xi_l^*=1+\beta^{2/3}(x^{1/3}-x^{-2/3}), \quad \beta\ll 1,\\ &\xi_l^*=\beta(x-2)^2(x-1)^{-3/2}, \quad \beta\gg 1, \end{aligned} \tag{3.14}$$

where

$$x=2/|k_l|>2.$$

4. THE BRANCHING EQUATIONS

It does not appear to be possible to find an anisotropic solution of the integral equation (2.7). Therefore, we shall confine ourselves to studying the solutions near the point of loss of stability (the point of bifurcation of the isotropic solution).[5] Let l_0 be that value of l for which ξ_l^* is a minimum. Denoting

$$u(s)=u(\xi_{l_0}^*)+w(s), \tag{4.1}$$

$$\mu=\xi-\xi_{l_0}^*, \tag{4.2}$$

we expand (2.8) in μ and w. The following equation for w is obtained:

$$w(s)=\int\frac{do'}{4\pi}K(s,s')\{a_{10}\mu+a_{01}w(s')+F(\mu;w(s'))\}. \tag{4.3}$$

Here and below, the constants

$$a_{mn}=\left(\frac{\partial}{\partial\xi}\right)^m\left(\frac{\partial}{\partial u}\right)^n\Phi(\xi_{l_0}^*;u_0),\quad u_0\equiv u(\xi_{l_0}^*), \tag{4.4}$$

and F denotes the higher terms of the expansion.

Equation (4.3) cannot be iterated in the nonlinearity, since its linear part cannot be solved for w. The insolubility is connected with the fact that the eigenvalues of the kernel $(4\pi)^{-1}a_{10}K$ that correspond to the eigenfunctions Y_{l_0m} are equal to $a_{10}k_{l_0}=1$; the latter follows from (3.4). Therefore, it is necessary to separate out from the kernel K the part responsible for the instability:

$$\frac{1}{4\pi}a_{10}K(s,s')=\bar{K}(s,s')+\sum_m Y_{l_0m}(s)Y_{l_0m}^*(s'). \tag{4.5}$$

After substitution of this representation into the linear part of Eq. (4.3) the coefficients

$$c_m=\int do\,w(s)Y_{l_0m}^*(s). \tag{4.6}$$

of the expansion in the "unstable" harmonics appear, and the equation itself takes the following form:

$$w(s)-\int do'\bar{K}(s,s')w(s')=\mu a_{10}+\sum_m c_mY_{l_0m}(s)+\int\frac{do'}{4\pi}K(s,s')F(\mu;w(s)). \tag{4.7}$$

We introduce the resolvent R of the kernel $\bar{K}$:

$$R(s,s')={\sum_{lm}}'r_lY_{lm}(s)Y_{lm}^*(s'),\quad r_l=a_{01}k_l(1-a_{01}k_l)^{-1}, \tag{4.8}$$

where the prime on the summation sign signifies that $l\neq l_0$ in the summation. Transforming now Eq. (4.7), we have

$$w(s)=\sum_m c_mY_{l_0m}(s)+\mu a_{10}(1-a_{01})^{-1}+\int do'Q(s,s')F(\mu;w(s')), \tag{4.9}$$

where

$$Q(s,s')=\frac{1}{4\pi}K(s,s')+\int\frac{do''}{4\pi}R(s,s'')K(s'',s')=\sum_{lm}q_lY_{lm}(s)Y_{lm}^*(s'),$$
$$q_l=k_{l_0},\quad l=l_0;\quad q_l=k_l(1-a_{01}k_l)^{-1},\quad l\neq l_0. \tag{4.10}$$

In the form (4.9), the equation for w can be iterated in the non-linearity. It is more convenient to seek not the function w itself, but the expansion coefficients c_m of the unstable harmonics and the expansion coefficients

$$b_{lm}=\int do\,w(s)Y_{lm}^*(s),\quad l\neq l_0 \tag{4.11}$$

of the "stable" harmonics. Multiplying (4.9) by $Y_{l_0m}^*$ and integrating, we find

$$\int do'Y_{l_0m}^*(s')F(\mu;w(s'))=0. \tag{4.12}$$

Analogously, multiplying by Y_{lm}^* with $l\neq l_0$, we obtain

$$b_{lm}=(4\pi)^{-1/2}\delta_{l,0}\mu a_{10}(1-a_{01})^{-1}+q_l\int doY_{lm}^*(s)F(\mu;w(s)). \tag{4.13}$$

As in the theory of phase transitions, the subsequent procedure is found to depend on the symmetry property of the "order parameter," i.e., in the present case, on the parity of l_0.

Even l_0. As will be seen from the following, in this case we obtain a solution in which c_m, $b_0\sim\mu$ and $b_{lm}(l\neq l_0)\sim\mu^2$. Therefore, in F it is necessary to keep only the next terms, of order μ^2:

$$F(\mu;w)={}^1/_2a_{20}\mu^2+a_{11}\mu w+{}^1/_2a_{02}w^2. \tag{4.14}$$

From (4.13) we find that, in lowest order,

$$b_0=(4\pi)^{-1/2}\delta_{l,0}\mu a_{10}(1-a_{01})^{-1}. \tag{4.15}$$

In substituting F into (4.12) it is necessary to keep only the leading terms, of order μ, in w, i.e., c_m and b_0. Then, using (4.15), we find an equation for c_m:

$$\mu A^{(1)}c_m+\sum_{m_1m_2}A^{(3)}_{m,m_1m_2}c_{m_1}c_{m_2}=0, \tag{4.16}$$

where

$$A^{(1)}=a_{11}+a_{10}a_{02}(1-a_{01})^{-1},$$
$$A^{(3)}_{m,m_1m_2}={}^1/_2a_{02}J_{l_0m;l_0m_1,l_0m_2}. \tag{4.17}$$

Here we have introduced notation for the integral of several spherical harmonics:

$$J_{lm;l_1m_1,l_2m_2\ldots}=\int do\,Y_{lm}^*(s)Y_{l_1m_1}(s)Y_{l_2m_2}(s)\ldots. \tag{4.18}$$

For even l_0 the coefficient $A^{(3)}\neq0$, and it can be seen that all the solutions of Eq. (4.16) give $c_m\sim\mu$, as was assumed. It follows from (4.13) with $l\neq l_0$ that $b_l\sim\mu^2$. We shall not need explicit expressions for these coefficients.

Odd l_0. In this case it is found that $c_m\sim\mu^{1/2}$, and all the $b_{lm}\sim\mu$. Now it is necessary to keep terms of order μ and $\mu^{3/2}$ in F:

$$F(\mu;w)={}^1/_2a_{02}w^2+{}^1/_6a_{03}w^3+a_{11}w\mu. \tag{4.19}$$

In substituting this expansion into (4.13) it is sufficient to keep only its leading term w^2, and also keep only the leading term, of order $\mu^{1/2}$ (i.e., c_m), in w. We then find

$$b_{lm}=(4\pi)^{-1/2}\delta_{l,0}\mu a_{10}(1-a_{01})^{-1}+\frac{1}{2}a_{02}q_l\sum_{m_1m_2}c_{m_1}c_{m_2}J_{lm;l_0m_1,l_0m_2}. \tag{4.20}$$

When the expansion (4.19) is substituted into (4.12) it is necessary to keep all three of the terms written out, and in substituting w into the leading term w^2 it is necessary also to keep the terms of b_{lm} in w. Using these terms (4.20), we find from (4.12) the following equation for c_m:

$$\mu A^{(1)} c_m + \sum_{m_1 m_2 m_3} A^{(4)}_{m,m_1m_2m_3} c_{m_1} c_{m_2} c_{m_3} = 0, \tag{4.21}$$

where the coefficient $A^{(1)}$ is the same as in (4.16), and

$$A^{(4)}_{m,m_1m_2m_3} = \frac{1}{2}(a_{02})^2 \sum_{l'm'} q_{l'} J_{l'm';l_0m_1,l_0m_2} J_{l_0m;l_0m_3,l'm'} + \frac{1}{6} a_{03} J_{l_0m;l_0m_1,l_0m_2,l_0m_3}. \tag{4.22}$$

It can be seen that all the solutions of (4.21) give $c_m \sim \mu^{1/2}$, as was assumed. We note that, with the chosen phase of the spherical harmonics, the coefficients $A^{(3)}$ and $A^{(4)}$ are real, and from the reality of w it follows that

$$c_m^* = (-1)^{l_0-m} c_{-m}. \tag{4.23}$$

5. CRITERION FOR THE STABILITY OF THE SOLUTIONS NEAR THE BIFURCATION POINT

To investigate the stability of the solutions near the bifurcation point we can expand the expression in square brackets in Eq. (2.13) in the small quantities μ, w and p. This equation then takes the form

$$\delta u(\mathbf{s}) = \int \frac{do'}{4\pi} K(\mathbf{s},\mathbf{s}')\,[a_{01} + G(p,\mu; w(\mathbf{s}'))]\,\delta u(\mathbf{s}'), \tag{5.1}$$

where G contains higher terms of the expansion, of orders not lower than μ, p and w. Multiplying (5.1) by a spherical harmonic and integrating, we find the coefficients of the expansion of $\delta u(\mathbf{s})$ in the Y_{lm}:

$$\delta u_{lm} = k_l \left\{ a_{01}\delta u_{lm} + \int do' Y_{lm}^*(\mathbf{s}') G(p,\mu; w(\mathbf{s}'))\,\delta u(\mathbf{s}') \right\}. \tag{5.2}$$

If $l \equiv l_0$, this equation can be solved for δu_{lm}, whence it can be seen that $\delta u_{lm} \sim G\delta u(\mathbf{s})$. Since G is small, this means that the major part of $\delta u(\mathbf{s})$ is made up of the unstable harmonics $Y_{l_0m}(\mathbf{s})$. The subsequent analysis of the stability is different for even and odd l_0, as was the case in the derivation of the branching equations.

Even l_0. Assuming that $p \sim \mu$, we find the leading terms of the expansion:

$$G(p,\mu; w) = \mu a_{11} - {}^1\!/_2 a_{02} p + a_{02} w. \tag{5.3}$$

With the assumption made, $G \sim \mu$, and therefore $\delta u_{l\neq l_0} \sim \mu \delta u_{l_0}$. In Eq. (5.2) with $l = l_0$, in the function $\delta u(\mathbf{s})$ in the integrand it is sufficient to keep only the unstable harmonics. The following system of equations is then obtained:

$$\sum_{m'} \{A_{mm'} - {}^1\!/_2 a_{02} p \delta_{mm'}\} \delta u_{l_0m'} = 0, \tag{5.4}$$

where

$$A_{mm'} = \mu A^{(1)} \delta_{mm'} + 2 \sum_{m_1} A^{(3)}_{m,m'm_1} c_{m_1}. \tag{5.5}$$

Equation (5.4) implies that $\frac{1}{2}a_{02}p$ is the eigenvalue of the matrix A. Since $c \sim \mu$, p also turns out to be of order μ, as was assumed.

Odd l_0. Assuming again that $p \sim \mu$, we find the leading terms of the expansion

$$G(p,\mu; w) = \mu a_{11} - {}^1\!/_2 a_{02} p + a_{02} w + {}^1\!/_2 a_{03} w^2. \tag{5.6}$$

In this case, $G \sim \mu^{1/2}$ and $\delta u_{l\neq l_0} \sim \mu^{1/2} \delta u_{l_0}$. If we take Eq. (5.2) for $l = l_0$, we cannot now confine ourselves just to the unstable harmonics in $\delta u(\mathbf{s})$, since, e.g., the term $w\delta u_{l\neq l_0}$ is of the same order as $w^2 \delta u_{l_0}$. The situation here is entirely analogous to that which obtained in the derivation of (4.21). Therefore, it is necessary first to express $\delta u_{l\neq l_0}$ in terms of δu_{l_0} using (5.2) for $l \neq l_0$, and then substitute them into (5.2) for $l = l_0$. Then the system (5.4) is obtained again, but now

$$A_{mm'} = \mu A^{(1)} \delta_{mm'} + 3 \sum_{m_1m_2} A^{(4)}_{m,m'm_1m_2} c_{m_1} c_{m_2}. \tag{5.7}$$

6. AXIALLY SYMMETRIC SOLUTIONS AND THEIR STABILITY

Any solution of Eq. (2.7) possesses the symmetry of one of the subgroups of the rotation group. Moreover, if this solution is rotated arbitrarily in space, we again obtain a solution. Both these remarks also apply, of course, to the solutions of the branching equations (4.16) and (4.21).

It is easiest of all to find axially symmetric solutions of the branching equations. The symmetry axis of such a solution can have arbitrary direction; by choosing the "quantization axis" z of the spherical harmonics to lie along the symmetry axis, we shall have $c_m = 0$ for $m \neq 0$. Then, from (4.16) we obtain

$$c_0 = -\mu A^{(1)}/A^{(3)}_{0.00}, \qquad l_0 \text{ even}, \tag{6.1}$$

and from (4.21),

$$c_0 = [-\mu A^{(1)}/A^{(4)}_{0.000}]^{1/2}, \qquad l_0 \text{ odd}. \tag{6.2}$$

We shall show that for $l_0 = 1, 2$ these exhaust all the possible solutions.

If $l_0 = 1$, the coefficients c_m transform like components of a vector $\mathbf{c}$, and

$$\sum_m c_m Y_m(\mathbf{s}) \sim \mathbf{cs}. \tag{6.3}$$

It is obvious that this function is axially symmetric about $\mathbf{c}$, along which we can point the z axis.

If $l_0 = 2$, the coefficients c_m transform like the components of a symmetric second-rank tensor, since

$$\begin{gathered} xz,\ yz \sim Y_{\pm 1}, \quad xy \sim Y_2 - Y_{-2}, \\ x^2 - y^2 \sim Y_2 + Y_{-2}, \quad x^2 + y^2 \sim Y_0. \end{gathered} \tag{6.4}$$

If the x, y and z axes are directed along the axes of the tensor, its xy, xz and yz components vanish. This means that, in solving (4.16) for $l_0=2$, we can assume without loss of generality that

$$c_{\pm 1}=0, \quad c_2=c_{-2}. \tag{6.5}$$

It then follows from (4.23) and (6.5) that

$$c_0^*=c_0, \quad c_2^*=c_2. \tag{6.6}$$

The system (4.16) is reduced to a system of two equations:

$$\begin{aligned} &\mu A^{(1)}c_0+2c_2^2A^{(3)}_{2,02}+A^{(3)}_{0,00}c_0^2=0,\\ &\mu A^{(1)}c_2+2c_0c_2A^{(3)}_{2,02}=0. \end{aligned} \tag{6.7}$$

Here we have used the symmetry property

$$A^{(3)}_{2,20}=A^{(3)}_{2,02}=A^{(3)}_{0,2\bar{2}}=A^{(3)}_{0,\bar{2}2}, \qquad \bar{2}=-2,$$

which follows from the explicit expression for the integral of three spherical harmonics in terms of Wigner coefficients. [4] The system (6.7) has a solution with $c_2=0$ and c_0 determined from (6.1); this solution is axially symmetric about the z axis. But if $c_2\neq 0$, then, calculating the coefficients $A^{(3)}$ explicitly, we can find that $c_2/c_0=\pm(3/2)^{1/2}$. Then,

$$\sum_m c_m Y_m(\mathbf{s})\sim(1-3\cos^2\theta)\pm 3\sin^2\theta\cos 2\varphi, \tag{6.8}$$

where θ and φ are the polar angles of the vector $\mathbf{s}$. Using the addition theorem, it is easy to verify that the latter expression coincides with $P_2(\mathbf{e}\cdot\mathbf{s})$, where, for the upper sign, the unit vector $\mathbf{e}\parallel x$, and for the lower sign, $\mathbf{e}\parallel y$. Thus, the solutions with $c_2\neq 0$ are also axially symmetric, but now about other axes of the tensor—x and y.

The matrices A determining the stability of the axially symmetric solutions are diagonal:

$$\begin{aligned} &A_{mm'}=\delta_{mm'}[\mu A^{(1)}+2c_0A^{(3)}_{m,m0}], \qquad l_0 \text{ even},\\ &A_{mm'}=\delta_{mm'}[\mu A^{(1)}+3c_0^2A^{(4)}_{m,m00}], \qquad l_0 \text{ odd}. \end{aligned} \tag{6.9}$$

The diagonal elements determine the damping constants p_m of fluctuations of the type Y_{l_0m}. Substituting the expression for c_0 from (6.1) and (6.2) into (6.9), we obtain

$$\begin{aligned} &p_m=-\mu a[1-2A^{(3)}_{m,m0}/A^{(3)}_{0,00}], \qquad l_0 \text{ even},\\ &p_m=-\mu a[1-3A^{(4)}_{m,m00}/A^{(4)}_{0,000}], \qquad l_0 \text{ odd}.\\ &a=-2A^{(1)}/a_{02}. \end{aligned} \tag{6.10}$$

The solutions of the branching equations and the corresponding damping constants are arranged differently for even and odd l_0. For even l_0 a solution $w(\mathbf{s})$ exists on both sides of the threshold; both the solution and the damping constants are completely independent of the properties of the kernel K that are associated with the stable harmonics, i.e., independent of the k_l with $l\neq l_0$. The ratio of the coefficients $A^{(3)}$ in (6.10) is simply a ratio of Wigner coefficients, and the stability is completely determined by the symmetry of the problem. On the other hand, for odd l_0 a real solution $w(\mathbf{s})$ exists only on one side of the threshold, where

$$\mu A^{(1)}/A^{(4)}_{0,000}>0, \tag{6.11}$$

since, according to (4.23), c_0 is pure-imaginary. The condition for the existence of the solution and the stability of the solution depend not only on k_{l_0} but also on the k_l with $l=0,2,\ldots,2l_0$, i.e., on the properties of the kernel K that are associated with the stable harmonics.

It is instructive to ascertain at which values of μ the anisotropy of the distribution (2.6) becomes of order unity, i.e., in effect, up to which values of μ the branching equations obtained by expanding in μ are valid. For this we can consider, e.g., the occupation numbers in the center of the distribution:

$$N(0,\mathbf{s})=\xi/(1+u(\mathbf{s})-\xi). \tag{6.12}$$

The anisotropy becomes of order unity when $w(\mathbf{s})$ is comparable with $1+u(\mathbf{s})-\xi$ for $\xi=\xi^*_{l_0}$. Estimates show that, irrespective of whether l_0 is even or odd, this happens when $\mu\approx\beta^{2/3}$, if $\beta\ll 1$, and when $\mu\approx\beta$, if $\beta\gg 1$; i.e., as we should expect, the characteristic range of variation of μ is the spacing between neighboring thresholds ξ^*_l, an estimate for which is obtained from (3.14).

We shall consider in more detail the simplest instabilities, with $l_0=1,2$. From (6.10) we find

$$\begin{aligned} &l_0=2{:}\ p_0=\mu a,\ p_{\pm 1}=0,\ p_{\pm 2}=-3\mu a,\\ &l_0=1{:}\ p_0=\mu a,\ p_{\pm 1}=0. \end{aligned} \tag{6.13}$$

The vanishing of the damping constants $p_{\pm 1}$ is a general property of the axially symmetric solutions; it reflects the neutrality of the equilibrium with respect to rotations of the distribution. In fact, under an infinitesimal rotation of the distribution $N(\mathbf{q})$, it changes by

$$\delta N(\mathbf{q})=i\delta\boldsymbol{\varphi}\mathbf{L}N(\mathbf{q}), \tag{6.14}$$

where $\delta\boldsymbol{\varphi}$ is the rotation vector, and $\mathbf{L}$ is the "angular-momentum" operator (in $\mathbf{q}$-space). If $N(\mathbf{q})$ is axially symmetric ($N(\mathbf{q})\sim Y_{l_00}$) then $\delta N(\mathbf{q})$ contains

$$(L_x\pm iL_y)Y_{l_00}\sim Y_{l_0\pm 1}. \tag{6.15}$$

It is easily verified that

$$A^{(1)}=-\frac{d}{d\xi}\Psi(\xi^*_{l_0})<0, \qquad a_{02}>0, \quad a>0.$$

Therefore, the axially symmetric distribution with $l_0=2$ is unstable below the threshold against fluctuations with $m=0$, which do not lower its symmetry, and unstable above the threshold against fluctuations with $m=\pm 2$, which do lower the symmetry. The instability below the threshold implies a transformation of the axially symmetric distribution into an isotropic distribution (with raising of the symmetry). The instability above the threshold implies the absence of nearly-isotropic solutions.

An axially symmetric distribution with $l_0=1$ is stable above the threshold, if it exists there, i.e., if $A^{(4)}_{0,000}<0$. Calculating the coefficient $A^{(4)}$, we can show that the condition for the existence of a solution above the threshold is

$$\frac{k_1}{k_1-1}+\frac{4}{5}\frac{k_1k_2}{k_1-k_2}>-\frac{3}{5}\frac{a_{03}}{(a_{02})^2}. \qquad (6.16)$$

For $\beta\ll 1$, from this inequality we obtain the condition

$$k_2>\frac{5|k_1|^2}{4-|k_1|}>0. \qquad (6.17)$$

It is interesting that if only one coalescence channel is operating and k_1 and k_2 are determined by formula (3.7), then (6.17) is never fulfilled. However, as soon as there are two coalescence channels it is possible to select β_1, β_2, χ_1, and χ_2 in such a way that (6.17) is fulfilled. This example is extremely instructive—the stability conditions and the character of the change of the distribution near the threshold can depend on quite detailed properties of the nonlinear mechanism.

7. SOLUTIONS WITH POINT-GROUP SYMMETRY

If $l_0>2$, there exist solutions of the branching equations with point-group symmetry. Thus, for $l_0=4$ there exists a solution with cubic symmetry, i.e., $\sum_m c_m Y_{4m}$ is a cubic harmonic with A_{1g} symmetry. It can be shown, however, that for even l_0 all solutions of the branching equations are unstable. For this it is sufficient to show that the matrix (5.5) has eigenvalues of opposite sign.

Multiplying Eq. (4.16) by two and comparing the result with (5.4), we can see that the matrix $A+\mu A^{(1)}I$ has eigenvalue zero, i.e., A has eigenvalue $-\mu A^{(1)}$. At the same time, $\mathrm{Tr}\,A=(2l_0+1)\mu A^{(1)}$, whence follows the existence of eigenvalues with sign opposite to the sign of $-\mu A^{(1)}$. In the calculation of the trace the second term in (5.5) makes no contribution, since

$$\sum_m 2\sum_{m_1} A^{(3)}_{m,m_1m}c_{m_1}=c_0a_{02}\sum_m J_{l_0m;l_00,l_0m}=0. \qquad (7.1)$$

The latter equality follows from the following property of the Wigner coefficients:

$$\sum_m (-1)^{l_0-m}\begin{pmatrix} l_0 & l_0 & l_0\\ -m & 0 & m\end{pmatrix}=\delta_{l_0,0}, \qquad (7.2)$$

which can be obtained easily from the orthogonality relations for these coefficients.

Odd l_0 have not been investigated, since the question of the stability in this case is connected with a very large number of parameters: even for $l_0=3$, besides k_3 three parameters (k_2, k_4 and k_6) appear.

8. RESULTS AND DISCUSSION

The principal result of this work can be formulated as follows: in two-phonon absorption of light with creation of nondecaying TA phonons there exists a threshold light-intensity J_s^* above which the nonequilibrium gas of TA phonons goes over into a state with lower symmetry than that of the crystal. The threshold J_s^* lies above the threshold J^* for parametric generation; the amount by which J_s^* exceeds J^* is determined by that nonlinear mechanism of TA-phonon destruction (coalescence) which limits the generation of TA phonons near the parametric threshold J^*. If the nonlinear destruction mechanism is weak compared with the linear mechanism ($\beta\ll 1$), then J_s^* is close to J^*—more precisely, $J_s^*-J^*\sim\beta^{2/3}$. If the nonlinear mechanism is strong ($\beta\gg 1$), J_s^* is substantially greater than J^*—more precisely, $J_s^*\sim\beta^{J^*}$.

Below the threshold J_s^* the phonon distribution function $N(\mathbf{q})$ is isotropic. The distribution is concentrated about a sphere $q=q_0$, in a certain spherical layer of thickness $\Delta q\ll q_0$. Above the threshold J_s^* such a distribution is unstable against certain fluctuations $\delta N(\mathbf{q})$, concentrated in the same spherical layer but destroying the isotropy of the distribution. The instability is connected with the following circumstance. By the conservation laws, the coalescence of two TA phonons from the spherical layer is possible only when the angle χ between the momenta of these phonons coincides with one of the several permissible χ_i; usually, none of the angles χ_i is equal to zero, i.e., identical TA phonons do not coalesce. We now picture a fluctuation $\delta N(\mathbf{q})$ concentrated predominantly in those directions of $\mathbf{q}$ between which the angles are far from all the χ_i. Since the gas is above the parametric-generation threshold, this fluctuation begins to grow, while preserving its angular distribution; at the same time the operation of the nonlinear suppression mechanism—the coalescence—will be made more difficult.

As a result of the development of such a fluctuation the phonon gas goes over into a state with an anisotropic distribution $N(\mathbf{q})$; as before, the distribution is concentrated about a sphere $q=q_0$, but the thickness Δq of the spherical layer and the maximum N_0 of the distribution (at $q=q_0$) now depend on direction.

Depending on the detailed properties of the coalescence probability, the transition from the isotropic to the anisotropic state at $J=J_s^*$ can resemble a first-order or a second-order phase transition. In the first variant, the anisotropy of the distribution $N(\mathbf{q})$ increases discontinuously to values of order unity when the threshold is exceeded by an arbitrarily small amount; solutions with small anisotropy are absent or unstable. In the second variant, stable distributions with small anisotropy arise when the threshold is slightly exceeded. In this case the anisotropy is determined by a spherical harmonic of a certain order l_0. For example, for $l_0=1$ there appears an axially symmetric distribution in which

$$\Delta q\sim a+b\cos\theta,\ b\sim(J-J_s^*)^{1/2}.$$

Here θ is the angle between $\mathbf{q}$ and a certain direction $\mathbf{e}$—the symmetry axis of the distribution, which can be arbitrarily oriented with respect to the crystal. We note that in the transition to such an anisotropic state, which does not possess a center of inversion, a heat flux arises spontaneously. Its magnitude can be estimated, assuming that the anisotropy of the distribution is of order unity:

$$Q \approx \frac{\hbar \Delta\nu}{a_0^3} s_{TA} \beta^{-1/2}, \qquad \beta \lesssim 1. \tag{8.1}$$

Here a_0^3 is the volume of the unit cell. Assuming that $\Delta\nu \approx 0.01$ cm^{-1}, which is typical for gas molecular lasers, we find $Q \approx 100$ W/cm^2.

Another manifestation of an axially symmetric distribution in the isotropic model is the existence of Goldstone modes corresponding to the group of rotations of the axis **e**. However, when the real crystal anisotropy is taken into account these modes disappear.

The threshold J_s^* can be detected from the singularity of the coefficient of nonlinear absorption of infrared radiation, which should have a kink or a discontinuity near J_s^*, depending on the type of transition.

In the model considered, two circumstances have not been taken into account: the presence of elastic scattering of TA phonons and the presence of crystal anisotropy. It is easy to show that in the framework of the isotropic model elastic scattering does not qualitatively alter the phenomenon, although quantitative factors (the thresholds and stability conditions) are changed.

To take the crystal anisotropy into account is rather complicated. For weak coalescence ($\beta \ll 1$) we are interested in the region near the parametric threshold J^*. However, in the anisotropic case, this threshold depends on the direction of the momentum on the constant-energy surface near which the phonons are created. It may happen that phonons created in the directions with the lowest threshold J^* cannot coalesce, and then the situation differs sharply from the isotropic case. However, this effect is blunted by effective elastic scattering of phonons by impurities, and the situation approximates to the isotropic situation. For strong coalescence ($\beta \gg 1$) the anisotropy of the thresholds J_s^* is unimportant, since in this case we are interested in the region of substantially higher intensities, at which the whole constant-energy surface is already excited. In the isotropic model considered, the loss of stability arose from the impossibility of coalescence of identical phonons. Since this property is not destroyed by anisotropy, we may suppose that the phenomenon as a whole is also preserved.

In crystals without a center of inversion, besides (1.1) the process $\nu \to 2$TA is possible. In this case the theory is entirely analogous, with the sole difference that the generation term (1.5) contains the sum $N(\mathbf{q}) + N(-\mathbf{q})$ in place of $N(\mathbf{q})$. Therefore, fluctuations with odd l, for which $\delta N(\mathbf{q}) + \delta N(-\mathbf{q}) = 0$, are not amplified by pumping and are always stable. Considering the case of even l, we can show that all anisotropic solutions near the bifurcation point are unstable, and therefore, the transition to the anisotropic state (if such a state exists) should occur discontinuously.

The last remark concerns the question of phase correlations in the phonon system. In the paper it has been assumed that there is no correlation between the phases of the individual phonon modes, although correlation could arise in the process of nonlinear interaction between modes (the so-called synchronization of modes). The frequency interval $(\Delta\omega)_s$ in which synchronization can arise is determined by the "nonlinear time," i.e., by the time for transfer of energy from one mode to another. Therefore, in our case,

$$(\Delta\omega)_s \approx \int d\mathbf{q}' W(\mathbf{s}, \mathbf{s}') N(\mathbf{q}'). \tag{8.2}$$

Using the estimates given in[6], it is easy to verify that $(\Delta\omega)_s \ll \Delta\omega$; this means that synchronization of modes is unimportant.

In conclusion we note that instabilities with respect to fluctuations that lower the symmetry of the system have also been noted in the classical problem of the parametric excitation of spin waves in a ferromagnet by monochromatic pumping.[7]

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