Theory of Stimulated Raman Scattering by Polaritons in Cubic and Uniaxial Crystals

V. L. Strizhevskiĭ
Kiev State University
Submitted September 17, 1971
Zh. Eksp. Teor. Fiz. 62, 1446–1460 (April, 1972)

Expressions are obtained for the gain g and the relative amplitudes of polariton radiation in stimulated Raman scattering by polaritons in cubic and uniaxial crystals. Two lines may appear in the scattering spectrum for cubic crystals, a polariton line and one due to scattering by a longitudinal phonon. With increase of scattering angle θ, the polariton line moves along the excited polariton dispersion curve, which is determined without taking absorption into account. When the phonon frequency is approached, the amplitude of the transverse part of the polariton wave decreases strongly as a result of growth of absorption and particularly of wave mismatch. A consequence of this is that in the region of sufficiently large θ the polariton wave excited at the mechanical (transverse Coulomb) phonon frequency becomes practically longitudinal. This anomalous longitudinal wave, which is maintained by the pumping field, can exist only in a pumped medium. In this case scattering is due to superposition of parametric polariton processes related to excitation of the above-mentioned wave and described by a quadratic polarization nonlinearity, and also of purely phonon type processes which can be described by a cubic nonlinearity and are of the same nature as in the case of nonpolar phonons. Because of the contribution of polariton processes, the expressions which determine the magnitude of g and also of the cross section σ for spontaneous Raman scattering in the region of large θ, differ from those for the case of nonpolar phonons. An example of the differences which arise is the "front-back asymmetry" of the g and σ angular dependence. It is also shown that owing to deformation of the dielectric constant by the pumping field, a cubic crystal becomes anisotropic: proper polarizations and the respective eigenvalues of g appear. In uniaxial crystals there exist scattering lines for ordinary, extraordinary and longitudinal polaritons centered on the corresponding dispersion curves without allowance for absorption. Expressions are obtained and analyzed which define the spectrum and angular dependence of g for these lines.

INTRODUCTION

STIMULATED Raman scattering (SRS) by polaritons, in crystals having no inversion center, has been attracting increasing attention of late. This phenomenon was investigated theoretically, in particular, in[1-6]. No complete theory has been developed for it, however, even for the case of excitation by plane monochromatic waves, and therefore a number of general physical laws remained unexplained.

The main shortcoming of the preceding studies were as follows: 1) No account was taken of the fact that the pump field, by deforming the permittivity of the medium, changes its symmetry. As a result, for example, a pumped cubic crystal becomes anisotropic. Corresponding proper polarizations set in together with corresponding eigenvalues of the gain. 2) The scattering by longitudinal phonons was not considered. 3) It was assumed that only waves with fixed transverse polarizations interact.

The latter assumption may not hold, for example, for a polariton wave in the vicinity of the phonon frequency. When the polariton line approaches (with increasing scattering angle θ) to the phonon frequency, the amplitude of the transverse part of the polariton wave (at the center of the line) decreases rapidly. This is due initially to the increased absorption, and with further increase of θ it is caused by the growth of the wave mismatch, which is due to the large difference between the lengths of the wave vectors of the visible and infrared bands (if the pump frequency lies in the visible region). As a result, the amplitude of the transverse polariton wave turns out to be commensurate with, and later on much lower than the amplitude of the longitudinal polariton wave, which is less sensitive to the value of the absorption and does not depend on the wave mismatch. Therefore, at sufficiently large θ, practically only longitudinal polariton waves are excited. On the other hand, if this is forbidden by the selection rules, the parametric polariton processes, which are described by the quadratic nonlinearity of the polarization, become completely unrealizable here, and the scattering is due to pure phonon processes, which are determined by the cubic nonlinearity, i.e., by processes of the same type as in the case of dipole-inactive oscillations.

It must be emphasized that we deal in this case with longitudinal waves occurring at the frequencies of the mechanical[7] phonons, which determine the poles (and not the zeroes) of the dielectric constant of the medium[1)]. Such waves are maintained by the pump field and exist only in a pumped medium. We shall therefore call them anomalous longitudinal waves, to distinguish them from the ordinary (normal) longitudinal waves that can exist also in an unpumped medium. The possibility of exciting anomalous longitudinal waves at the frequency of the mechanical phonons is the fundamental difference between scattering by dipole-active (polar) phonons and scattering by dipole-inactive (nonpolar) ones. In addition, of course, normal longitudinal waves can also be excited.

We shall show that these shortcomings narrow down considerably the region of applicability of the results of the earlier studies. We have therefore developed a

[1)]We recall that the frequencies of the mechanical phonons used by us in this paper coincide with the frequencies of the transverse Coulomb phonons, which can be introduced as a particular case of Coulomb excitons[7].

theory that is free, to a considerable degree, of these shortcomings. In view of the customary large absorption of crystals in the vibrational region of the spectrum even away from the phonon resonances, primary interest attaches to the cases when the absorption coefficient at the polariton frequency greatly exceeds the gain. We have performed such an investigation. Principal attention was paid to a calculation and analysis of the gain, which determines the intensities of both stimulated (SRS) and spontaneous (SpRS) Raman scattering[4,8].

We carry out our analysis in the approximation of a given stationary pump field, which is approximated by a linearly polarized plane monochromatic wave. It is assumed that the scattering medium takes the form of a layer bounded by the planes z = 0 and z = l. The pump wave

$$\mathbf{E}_l(\mathbf{r}, t) = \mathbf{e}_l A_l \exp(i(k_l z - \omega_l t)) + \text{c.c.}$$

propagates along the z axis. The subscripts l, s, and p will henceforth denote the pump (usually laser), Stokes, and polariton wave fields; ω are the frequencies, n and **k** are the refractive indices and the wave vectors in the unpumped medium, and **e** are real unit vectors of polarization. The medium is assumed to be nonmagnetic and transparent at the frequencies $\omega_{l,s}$.

1. CASE OF CUBIC CRYSTALS

We seek the Stokes and polariton fields in the form

$$\mathbf{E}_s(\mathbf{r}, t) = \sum_{\mu=1,2} \mathbf{e}_s^{(\mu)} A_s^{(\mu)} \exp\{i(\mathbf{k}_s\mathbf{r} - \omega_s t)\} + \text{к.с.},$$

$$\mathbf{e}_s^{(\mu)} \perp \mathbf{k}_s, \quad \mathbf{e}_s^{(1)} \perp \mathbf{e}_s^{(2)}, \quad k_s = q_s n_s, \quad q_s = \omega_s / c;$$

$$\mathbf{E}_p(\mathbf{r}, t) = \sum_{\sigma=1,2,3} \mathbf{e}_p^{(\sigma)} A_p^{(\sigma)} \exp\{i(\mathbf{w}\mathbf{r} - \omega_p t)\} + \text{к.с.},$$

$$\mathbf{w} = \mathbf{k}_l - \mathbf{k}_s, \quad \mathbf{e}_p^{(1,2)} \perp \mathbf{w}, \quad \mathbf{e}_p^{(1)} \perp \mathbf{e}_p^{(2)}, \quad \mathbf{e}_p^{(3)} = \mathbf{w}/w, \quad \omega_p = \omega_l - \omega_s, \tag{1}$$

The longitudinal component of the Stokes wave can obviously be neglected[2)], but this cannot be done for a polariton wave in the phonon region. As will be seen from the formulas that follow, with further advance into this region all three amplitudes $A_p^{(\sigma)}$ first become comparable, after which $A_p^{(3)}$ predominates, provided the excitation of the longitudinal waves is allowed by the selection rules. The phase shift of the polariton wave is determined by the vector **w** and not by $\mathbf{k}_p$ ($k_p = q_p\sqrt{\epsilon_p}$, $q_p = \omega_p/c$, $\epsilon_p = \epsilon_p' + i\epsilon_p''$ is the dielectric constant at the frequency ω_p). When the phases are chosen as in (1), the amplitudes $A_p^{(\sigma)}$ are smooth functions of z, so that the method of shortened equations is applicable[9] (see also[8]).

The fields $\mathbf{E}_{s,p}$ are related via the nonlinear part of the polarization of the medium $\mathbf{P}^{NL}(\mathbf{r}, t)$. The latter has at the frequency ω_s the following form[3)]:

$$P_{si}^{NL} = (\chi_{ijk} e_l^j e_p^{(\sigma)k} A_p^{(\sigma)*} A_l + \gamma_{ijkm} e_s^{(\mu)j} e_l^k e_l^m A_s^{(\mu)} |A_l|^2) \exp[i(\mathbf{k}_s\mathbf{r} - \omega_s t)] + \text{к. с.}$$

Here $\chi_{ijk} = \chi_{ijk}(\omega_l, -\omega_p)$ and $\gamma_{ijkm} = \gamma_{ijkm}(\omega_s, \omega_l, -\omega_l)$ are the corresponding nonlinear polarizabilities of the medium[9]. The terms quadratic in the field determine the contributions of the parametric processes, while the cubic terms determine the phonon contributions. A similar expression, with an obvious interchange of indices, holds also for the nonlinear polarization at the frequency ω_p. We note that in view of the strong absorption in the frequency region ω_p, the terms of P_{pi}^{NL} containing $\gamma_{ijkm}(\omega_p, \omega_l, -\omega_l)$ are immaterial and can be omitted[4)].

The shortened equations for the amplitudes $A_{s,p}^{(\mu,\sigma)}$ are obtained from Maxwell's equations by the standard procedure[9] and take the form

$$ik_s^z \frac{\partial A_s^{(\mu)}}{\partial z} + 2\pi q_s^2 (\chi^{\mu\sigma} A_p^{(\sigma)*} A_l + \gamma^{\mu\mu'} A_s^{(\mu')} |A_l|^2) = 0, \quad \mu = 1.2, \tag{2}$$

$$2iw^z \frac{\partial A_p^{(\sigma)*}}{\partial z} - iwe_p^{(\sigma)z} \frac{\partial A_p^{(3)*}}{\partial z} + (w^2 - k_p^{2*}) A_p^{(\sigma)*} - 4\pi q_p^2 \chi^{\mu\sigma} A_s^{(\mu)} A_l = 0, \quad \sigma = 1.2, \tag{3}$$

$$k_p^{2*} A_p^{(3)*} + iw\left(e_p^{(1)z} \frac{\partial A_p^{(1)*}}{\partial z} + e_p^{(2)z} \frac{\partial A_p^{(2)*}}{\partial z}\right) + 4\pi q_p^2 \chi^{\mu 3} A_s^{(\mu)} A_l^* = 0. \tag{4}$$

Here

$$\chi^{\mu\sigma} = e_s^{(\mu)i} e_l^j e_p^{(\sigma)k} \chi_{ijk}, \quad \gamma^{\mu\mu'} = e_s^{(\mu)i} e_s^{(\mu')j} e_l^k e_l^m \gamma_{ijkm}.$$

In deriving the system (2–4), we took into account the fact that $\chi_{ijk}(\omega_l, -\omega_s) = \chi_{kji}(\omega_l, -\omega_p)$[10,11].

In view of the strong absorption we have $|wA_p^{(\sigma')-1} \partial A_p^{(\sigma)} / \partial z| \ll |w^2 - k_p^{2*}|$, and we can therefore neglect in (3) and (4) the terms with the derivatives[5)], after which these equations yield

$$A_p^{(\sigma)*} = \frac{4\pi\chi^{\mu\sigma} A_s^{(\mu)} A_l^*}{\mu^2 - \varepsilon_p^*}, \quad \sigma = 1.2;$$

$$A_p^{(3)*} = -\frac{4\pi\chi^{\mu 3} A_s^{(\mu)} A_l^*}{\varepsilon_p^*}, \quad \mu = \frac{w}{q_p}. \tag{5}$$

Substituting the obtained expressions in (2), we arrive at a system of two differential equations with respect to $A_s^{(\mu)}$. We seek its solution in the form $A_s^{(\mu)} = B_\mu \exp \kappa z$, assuming B_μ and κ to be independent of z. We then obtain a system of algebraic equations with respect to B_μ. Choosing in a plane perpendicular to $\mathbf{k}_s$ a two-dimensional coordinate system $\tilde{\sigma}$ with axes along the unit vec-

2) It is smaller by a factor g/q_s than the transverse ones (g is the gain).

3) Summation over the repeated indices i, j, k, and m designating the tensor components is implied throughout. It will also be implied for the repeated indices $\mu = 1$ and 2 and $\sigma = 1$, 2, and 3. In some specially indicated cases the summation over σ is carried only over the values $\sigma = 1$ and 2. The unit vector $e_p^{(3)}$ will also be designated $e_p^{\parallel}$.

4) The corresponding criterion is of the form $\epsilon_p'' \gg 4\pi |\text{Im}\gamma| |A_l|^2$, where $\gamma = e_s^i e_s^j e_l^k e_l^m \gamma_{ijkm}$.

5) In addition to the solution that increases with z, which is of interest to us, the system (2-4) has also a solution that attenuates with increasing z, and the rate of attenuation is determined by the quantity $k'' = \text{Im}\, k_p$. Assuming that $k_p'' l \gg 1$, we take into account only the solution that grows in space. The criterion that allows us to neglect the terms with the derivatives in (3) and (4) can be represented in the form $g \ll \text{Im}\, k_p^2/w$.

tors $e_s^{(\mu)}$, we represent the equations for B_μ in the form of a tensor relation

$$\Delta_{\mu\mu'}B_{\mu'} = -2ik_s^z \varkappa B_\mu, \quad \mu = 1, 2, \tag{6}$$

where

$$\Delta_{\mu\mu'} = 4\pi q_s^2 |A_l|^2 (\gamma^{\mu\mu'} + 4\pi\chi^{\mu\sigma}\chi^{\mu'\sigma}u_\sigma), \; u_{1,2} = (\mu^2 - \varepsilon_p^*)^{-1}, \; u_3 = -(\varepsilon_p^*)^{-1}. \tag{7}$$

The quantities $\gamma^{\mu\mu'}$ and $\chi^{\mu\sigma}\chi^{\mu'\sigma}u_\sigma$ are the components, in the $\tilde{\sigma}$ system, of the second-rank tensors $\gamma_{ijkm}e_l^k e_l^m$ and $\chi_{ikm}\chi_{jnq}e_l^k e_l^n e_p^{(\sigma)r n} e_p^{(\sigma)q} u_\sigma$. Equating the determinant of the system (6) to zero, we obtain the quantity of interest to us

$$\varkappa = i\{\Delta_{11} + \Delta_{22} \pm [(\Delta_{11} - \Delta_{22})^2 + 4\Delta_{12}\Delta_{21}]^{1/2}\}/4k_s^z. \tag{8}$$

We shall need subsequently explicit expressions for the tensors χ and γ. They can be found[12,13] within the framework of the microscopic theory in the dipole approximation using as the basis the perturbation-theory states, say the mechanical excitons[7]. We present the following expressions:

$$\chi_{ijk}(\omega_l, -\omega_p) = \hbar^{-1}\sqrt{N}\sum_{f\nu} \alpha_{ij}^{(f\nu)} P_{f\nu}^k F_f(\omega_p) + \chi_{ijk}^0(\omega_l, -\omega_p), \tag{9}$$

$$\gamma_{ijkm} = (\hbar v_0)^{-1}\sum_{f\nu} \alpha_{ik}^{(f\nu)} \alpha_{jm}^{(f\nu)} F_f(\omega_p) + \gamma_{ijkm}^0, \tag{10}$$

$$F_f(\omega_p) = (\omega_f - \omega_p + i\tilde{\gamma}_f/2)^{-1} + (\omega_f + \omega_p - i\tilde{\gamma}_f/2)^{-1} \approx 2\omega_f(\omega_f^2 - \omega_p^2 + i\tilde{\gamma}_f\omega_p)^{-1}. \tag{11}$$

The summation in (9) and (10) is over all the dipole-active oscillations, the frequencies of which are set equal to $\omega_f - i\tilde{\gamma}_f/2$, where $\tilde{\gamma}_f$ are the attenuation constants. In a cubic crystal, the dipole active mechanical vibrations are triply degenerate[14]; to number the mutually degenerate vibrations, we introduce the index ν; e_ν (ν = 1, 2, 3) is a triad of real unit vectors of the polarization of the vibrations along the edges of the unit cube. Further, $P_{f\nu} = P_f e_\nu$ is the dipole moment of the transition $0 \to f\nu$ for the unit cell, referred to its volume v_0; $\alpha_{ij}^{(f\nu)}$ is the corresponding tensor of the phonon spontaneous Raman scattering per cell[15] and can be regarded as symmetrical; $N = V/v_0$ is the number of cells in the crystal. The tensor χ_{ijk}^0 gives the contribution made to χ_{ijk} by the remote electronic states; in the vibrational region of frequencies ω_p, the tensor χ_{ijk}^0 is practically real and is independent of ω_p. The tensor γ_{ijkm}^0 determines the contribution made to γ_{ijkm} by the electronic states and the nonpolar vibrations. Since allowance for γ_{ijkm} is essential only in the resonant region, where the resonance term predominates, the term γ_{ijkm}^0 can be omitted.

It is convenient to represent the tensor χ_{ijk} (9) in the form

$$\chi_{ijk} = \sum_f \left(\frac{s_f}{2\pi v_0\hbar\omega_f}\right)^{1/2} \frac{a_{ijk}^f}{\beta_f x_f}\frac{\tilde{\varphi}_f - i}{1 + \varphi_f^2}. \tag{12}$$

Here

$$\varphi_f = \frac{1 - x_f^2}{\beta_f x_f}, \; x_f = \frac{\omega_p}{\omega_f}, \; \beta_f = \frac{\tilde{\gamma}_f}{\omega_f}, \; s_f = \frac{8\pi V P_f^2}{\hbar\omega_f}, \tag{13}$$
$$a_{ijk}^f = \sum_\nu \alpha_{ij}^{(f\nu)} e_\nu^k, \; \tilde{\varphi}_f = \varphi_f + \beta_f x_f A_f (1 + \varphi_f^2),$$

$$A_f = \frac{\chi^0}{d_f}\left(\frac{2\pi v_0\hbar\omega_f}{s_f}\right)^{1/2},$$

s_f is the oscillator strength of the $0 \to f$ transition. Account is taken of the fact that in cubic crystals without an inversion center, which admit of polar vibrations that are active in the scattering, each of the tensors χ_{ijk}^0 and a_{ijk}^f has equal nonzero components provided all three indices are different[16]. The common value of these components is designated χ^0 for the tensor χ_{ijk}^0 and d_f for a_{ijk}^f. The quantity d_f coincides with the common value of the nonzero components of the tensor $\alpha_{ij}^{(f\nu)}$[14].

Using formulas (12) and (13), and also the relations

$$\sum_{\sigma=1,2} e_p^{(\sigma)k} e_p^{(\sigma)k'} = \delta_{kk'} - e_p^{\parallel k} e_p^{\parallel k'}, \; e_\nu^k e_{\nu'}^k = \delta_{\nu\nu'}, \tag{14}$$

we reduce the tensor $\Delta_{\mu\mu'}$ (7) to the form

$$\Delta_{\mu\nu} = \frac{8\pi q_s^2|A_l|^2}{\hbar v_0}\Big[\sum_f (\beta_f x_f \omega_f)^{-1}\frac{\tilde{\varphi}_f - i}{1 + \varphi_f^2}\eta_{\mu\nu}^{ff}$$
$$+ \sum_{ff'}\left(\frac{s_f s_{f'}}{\omega_f\omega_{f'}}\right)^{1/2}\frac{(\tilde{\varphi}_f - i)(\tilde{\varphi}_{f'} - i)}{\beta_f\beta_{f'}x_f x_{f'}(1 + \varphi_f^2)(1 + \varphi_{f'}^2)}$$
$$\times\left(\frac{\psi_{\mu\nu}^{ff'}}{\mu^2 - \varepsilon_p^*} - \frac{\zeta_{\mu\nu}^{ff'}}{\varepsilon_p^*}\right)\Big]. \tag{15}$$

We have introduced here the notation:

$$\psi_{\mu\nu}^{ff'} = \eta_{\mu\nu}^{ff'} - \zeta_{\mu\nu}^{ff'}, \; \eta_{\mu\nu}^{ff'} = \sum_\nu \alpha_\mu^{(f\nu)}\alpha_\nu^{(f'\nu)}, \; \zeta_{\mu\nu}^{ff'} = \zeta_\mu^f \zeta_\nu^{f'}, \tag{16}$$

$$\zeta_\mu^f = \sum_\nu \alpha_\mu^{(f\nu)}(e_p^\parallel, e_\nu), \; \alpha_\mu^{(f\nu)} = e_s^{(\mu)i} e_l^j \alpha_{ij}^{(f\nu)}.$$

It is seen from (15) and (16) that the tensor $\Delta_{\mu\mu'}$ is symmetrical, so that its real ($\Delta'_{\mu\mu'}$) and imaginary ($\Delta''_{\mu\mu'}$) parts can be referred to the principal axes. These include the most interesting singled-out scattering geometries[6)]. It is possible here to introduce the principal axes of the tensor $\Delta_{\mu\mu'}$ as a whole. We denote its principal values by Δ_μ. Using (8), we obtain accordingly $\varkappa_\mu = i\Delta_\mu/2k_s^z$.

We introduce further the gain $g_\mu = 2\mathrm{Re}\,\varkappa_\mu = -\mathrm{Im}(\Delta_\mu/k_s^z)$. We express it in terms of the nonlinear polarizabilities:

$$g_\mu = \frac{8\pi^2\omega_s I_l}{c^2 n_l n_s \cos\theta}\Big[4\pi\sum_{v=\perp,\parallel}\frac{\mathrm{Re}\,\chi_{\mu v}^2 - \tau_v\,\mathrm{Im}\,\chi_{\mu v}^2}{\varepsilon_p''(1 + \tau_v^2)} - \mathrm{Im}\,\Gamma_\mu\Big], \; \mu = 1, 2. \tag{17}$$

Here $I_l = cn_l|A_l|^2/2\pi$ is the pump intensity, θ is the scattering angle, i.e., the angle between $\mathbf{k}_l$ and $\mathbf{k}_s$, and Γ_μ are the principal values of the tensor $\gamma^{\mu\mu'}$. Furthermore,

$$\chi_{\mu\perp}^2 = (\chi^{\mu 1})^2 + (\chi^{\mu 2})^2, \; \chi_{\mu\parallel} = \chi^{\mu 3}; \; \tau_\perp = \tau = \frac{\mu^2 - \varepsilon_p'}{\varepsilon_p''}, \; \tau_\parallel = \tau_L = -\frac{\varepsilon_p'}{\varepsilon_p''}. \tag{18}$$

Formulas (17) and (18) determine two proper gain coefficients for Stokes waves with polarizabilities $e_s^{(\mu)}$, which correspond to the principal axes of the tensor $\Delta_{\mu\mu'}$.[7)] Thus, the pumped cubic crystal becomes aniso-

[6)]This limitation does not hold in the nonresonant region, when $\Delta''_{\mu\mu'} = 0$

[7)]It follows from (13), (17), and (18) that g does not depend on the choice of the unit vectors $e_p^{(\sigma)}(\sigma = 1,2)$ in a plane perpendicular to **w**.

tropic. The anisotropy is due to the deformation of the dielectric constant by the linearly polarized pump wave.

We shall discuss henceforth in detail the case of an isolated oscillation ω_f. We represent ϵ_p' and ϵ_p'' in the form

$$\varepsilon_p' = \varepsilon_\infty + \frac{s}{\beta x}\frac{\varphi}{1+\varphi^2}, \quad \varepsilon_p'' = \frac{s}{\beta x}\frac{1}{1+\varphi^2},$$

where ϵ_∞ is the high-frequency limit of ϵ_p' relative to ω_f. For this case, using the formulas presented above and omitting the indices f and μ, we have

$$g = \frac{g_0}{1+\varphi^2}\left[M\frac{(\tilde\varphi+\tau)^2}{1+\tau^2} + \Lambda\frac{(\tilde\varphi+\tau_L)^2}{1+\tau_L^2}\right], \quad g_0 = G\eta, \quad \eta = \sum_\nu [\alpha^{(\nu)}]^2,$$

$$G = \frac{16\pi^2\omega_s I_l}{c^2 n_l n_s \cos\theta\hbar\tilde\gamma x v_0}, \quad M = 1-\Lambda, \quad \Lambda = \frac{1}{\eta}\left[\sum_\nu \alpha^{(\nu)}(\mathbf{e}_p^{\parallel}, \mathbf{e}_\nu)\right]^2. \quad (19)$$

The structure of the tensor $\alpha_{ij}^{(\nu)}$ is known[14]. Using this, it is easy to verify that

$$\Lambda = \frac{[e_p^{\parallel x}(e_s^y e_l^z + e_s^z e_l^y) + e_p^{\parallel y}(e_s^z e_l^x + e_s^x e_l^z) + e_p^{\parallel z}(e_s^x e_l^y + e_s^y e_l^x)]^2}{(e_s^y e_l^z + e_s^z e_l^y)^2 + (e_s^z e_l^x + e_s^x e_l^z)^2 + (e_s^x e_l^y + e_s^y e_l^x)^2}. \quad (20)$$

Let us discuss certain properties of the gain g (19). Assuming that g_0, M, and Λ vary little within the limits of the scattering lines, and analyzing in analogy with[4,5] each of the two terms in (19), we conclude that g has two maxima, corresponding to $\tau = \varphi^{-1}$ and $\tau_L = \varphi^{-1}$. The former determines the center of the polariton scattering line (cf.[4,5]), and the latter the lines of scattering by a longitudinal phonon, inasmuch as the condition $\tau_L = \varphi^{-1}$ is equivalent to $\epsilon_\infty + s(1-x^2)^{-1} = 0$. We denote these lines by p and p_L, respectively. The line p is centered on the dispersion curve Π of the polaritons that would be produced in the course of scattering in the absence of absorption and wave mismatch. The line p_L remains unchanged in position relative to θ.

It is useful to note that since M and Λ are generally speaking of the same order, the values of g for the lines p and p_L (which we shall denote by g_p and g_L) are also of the same order. In particular, at the centers of the lines ω_p and ω_L

$$\frac{g_p^{max}}{g_L^{max}} = \frac{\omega_L}{\omega_p}\left[\frac{1+A(1-x^2)}{1+A(1-x_L^2)}\right]^2\frac{M}{\Lambda}, \quad x_L = \frac{\omega_L}{\omega_f}.$$

Particular interest attaches to the case of large θ, when the line p goes over into the phonon scattering line p_f, centered at the frequency ω_f. Here $\tau \gg 1$ and $\tau \gg \tilde\varphi$ (with the exception of the rare cases of very large $|\beta_A|$)[8]. We therefore have for the line p_f

$$g_f = G\eta M/(1+\varphi^2). \quad (21)$$

Let, for example, the scattering occur in the plane σ' of the face of the principal cube, and let $\mathbf{k}_l$ make an angle β_0 with one of its edges. We consider the cases when $\mathbf{e}_l$ lies in the σ' plane and is perpendicular to it. In both cases one of the eigenvectors $\mathbf{e}_s^{(\mu)}$ lies in the σ' plane ($\mathbf{e}_s^{(1)}$), and the other ($\mathbf{e}_s^{(2)}$) is perpendicular to it. This is easiest to demonstrate by verifying that $\psi_{12} = \zeta_{12} = 0$. In the former case ($\mathbf{e}_l \parallel \sigma'$)

$$\eta^{(1)} = d^2\sin^2(\theta+2\beta_0), \quad M^{(1)} = 1, \quad g_{f1} = \frac{Gd^2\sin^2(\theta+2\beta_0)}{1+\varphi^2};$$

$$\eta^{(2)} = d^2, \quad M^{(2)} = \sin^2(\theta_p - 2\beta_0), \quad g_{f2} = \frac{Gd^2\sin^2(\theta_p-2\beta_0)}{1+\varphi^2} \approx \frac{Gd^2\cos^2(\tfrac{1}{2}\theta+2\beta_0)}{1+\varphi^2}. \quad (22)$$

Here θ_p is the angle between **w** and $\mathbf{k}_l$. We have taken into account the fact that at large θ we can put $\theta_p \approx (\pi-\theta)/2$.

In the second case ($\mathbf{e}_l \perp \sigma'$)

$$\eta^{(1)} = d^2, \quad M^{(1)} = \sin^2(\theta_p - \theta - 2\beta_0), \quad g_{f1} = \frac{Gd^2\sin^2(\theta_p-\theta-2\beta_0)}{1+\varphi^2} \approx \frac{Gd^2\cos^2(\tfrac{3}{2}\theta+2\beta_0)}{1+\varphi^2}; \quad \eta^{(2)} = 0, \quad M^{(2)} = 1, \quad g_{f2} = 0. \quad (23)$$

Let now $\mathbf{k}_l$ be parallel to the large diagonal of the cube and let the scattering be observed in a plane σ'' passing through $\mathbf{k}_l$ and one of the edges, with either $\mathbf{e}_l \parallel \sigma''$ or $\mathbf{e}_l \perp \sigma''$. One of the eigenvectors $\mathbf{e}_s^{(\mu)}$ also lies in the scattering plane σ'' ($\mathbf{e}_s^{(1)}$), and the other ($\mathbf{e}_s^{(2)}$) is perpendicular to it. At $\mathbf{e}_l \parallel \sigma''$ we obtain

$$\eta^{(1)} = d^2\left(\sin^2\beta_1 + \frac{1}{3}\cos^2\beta_2\right), \quad M^{(1)} = \frac{(\sin\beta_1\cos\beta_3 + 3^{-1/2}\cos\beta_2\sin\beta_3)^2}{\sin^2\beta_1 + \tfrac{1}{3}\cos^2\beta_2},$$

$$g_{f1} = \frac{G\eta^{(1)}M^{(1)}}{1+\varphi^2}; \quad \eta^{(2)} = \frac{2}{3}d^2, \quad M^{(2)} = 1, \quad g_{f2} = \frac{2}{3}\frac{Gd^2}{1+\varphi^2};$$

$$\beta_1 = 2\alpha_0 - \theta, \quad \beta_2 = \alpha_0 - \theta, \quad \beta_3 = \alpha_0 + \theta_p, \quad \cos\alpha_0 = 3^{-1/2}.$$

On the other hand, if $\mathbf{e}_l \perp \sigma''$, then

$$\eta^{(1)} = d^2\sin^2\beta_2, \quad M^{(1)} = 1, \quad g_{f1} = \frac{Gd^2\sin^2\beta_2}{1+\varphi^2};$$

$$\eta^{(2)} = d^2, \quad M^{(2)} = \sin^2\beta_3, \quad g_{f2} = \frac{Gd^2\sin^2\beta_3}{1+\varphi^2} \approx \frac{Gd^2\cos^2(\tfrac{1}{2}\theta-\alpha_0)}{1+\varphi^2}.$$

The gain g determines fully the SRS intensity. According to[8], the spectral density of the surface brightness at the exit face is, in the Stokes frequency region,

$$B_s = B_s^0(e^{gl}-1), \quad B_s^0 = \hbar\omega_s^3 n_s^2/8\pi^3c^2. \quad (24)$$

Changing over at $gl \ll 1$ to the case of spontaneous Raman scattering (SpRS) we obtain the SpRS cross section per unit solid angle, per unit spectral interval, and per unit volume[9]:

$$\sigma_{\omega_s\Omega_s} = B_s^0 g\cos\theta/I_l. \quad (25)$$

At $g = g_f$, formula (25) determines the cross section of SpRS by polar phonons. Integrating with respect to the frequencies, we obtain

$$\sigma_{\Omega_s}^f = \int_0^\infty \sigma_{\omega_s\Omega_s}^f d\omega_s = \sigma^0 M, \quad \sigma^0 = \frac{1}{v_0}\left(\frac{\omega_s}{c}\right)^4\frac{n_s}{n_l}\eta.$$

The quantity σ^0 coincides with the expression for the cross section for scattering by dipole-inactive phonons that do not interact with the field[15]. The presence of dipole activity leads to the appearance of an additional factor M, which is determined by formulas (18) and (19). This factor differs from unity, owing to the activity of the anomalous longitudinal polariton waves. Being strongly dependent on the angle, the factor M influences considerably the SpRS intensity. In particular, it leads to the characteristic "front-back asymmetry" in scattering in the σ' plane. For example at $\beta_0 = 0$ or $\pi/2$ the value of M, in the cases described by formulas (22) and (23), is respectively $\sin^2\theta_p \approx \cos^2(\theta/2)$ and $\sin^2(\theta_p - \theta)$

[8]The region of angles corresponding to the line p_f is determined by the condition $\sin(\theta/2) > (\omega_f/2\omega_l n_l)(s/\beta)^{1/2}$. It is usually satisfied already at $\theta \sim 10$–15°.

[9]We recall that the angle θ is defined inside the medium.

$\approx \cos^2(3\theta/2)$, whereas σ^0 is symmetrical. It would be difficult to register the resultant asymmetry experimentally.

We present also a formula for the cross section of spontaneous Raman scattering by longitudinal phonons, integrated over the frequencies, for $\varphi \gg 1$[10]:

$$\sigma_{q_s}{}^{L} = \sigma_{q_s}{}^{f}\frac{\Lambda}{1-\Lambda}\frac{[1+A(1-x_L{}^2)]^2}{x_L}. \qquad (26)$$

We point out one more important consequence of the results, with[8] also taken into account. The intensity of SRS with a polarization $\mathbf{e}_s$ intermediate between $\mathbf{e}_s^{(1,2)}$, in view of the statistical independence of the mutually perpendicular noise oscillations of the field, is determined by the formula

$$B_s = B_s{}^0[\pi_1(e^{g_1 l}-1)+\pi_2(e^{g_2 l}-1)],\quad \pi_{1,2} = (\mathbf{e}_s, \mathbf{e}_s^{(1,2)})^2.$$

The result of its application differs significantly from that obtained by using formula (24) in which the given $\mathbf{e}_s$ is substituted. This difference is greater the larger gl. To the contrary, it vanishes when $gl \ll 1$, i.e., in the case of spontaneous Raman scattering.

Finally, let us discuss the ratio of the amplitudes of the transverse and longitudinal polariton waves. As is clear from (5), outside the phonon resonance the polariton wave is in the main transverse and (normal) longitudinal for the lines p and p_L, respectively. However, as the line p approaches resonance with increasing θ, the values of $A_p^{(1,2)}$ and $A_p^{(3)}$ for this line become comparable, owing to the growth of ϵ_p''. With further increase of θ, when μ increases appreciably (so that $\mu^2 \gg \epsilon_p''$) and the line p goes over into p_f, we have $A_p^{(1,2)} \to 0$ and the amplitude of the longitudinal wave becomes dominant. Thus, in the region of sufficiently large θ the contribution of the parametric processes, as already indicated in the introduction, is connected with excitation of only the anomalous longitudinal polariton wave. If this is forbidden by the selection rules, there remain only the pure phonon processes described by the tensor γ_{ijkm}.

It is also useful to note that owing to the identity

$$\eta_1+\eta_2+\eta_3 = \eta,\quad \eta_\sigma = \Big[\sum_\nu \alpha^{(\nu)}(\mathbf{e}_p^{(\sigma)}, \mathbf{e}_\nu)\Big]^2,\quad \sigma = 1,2,3,$$

from which follows the formula $M = 1 - \eta_3/\eta = (\eta_1 + \eta_2)/\eta$, we can express g_p and g_f only in terms of the unit vectors of the transverse-polariton polarization. In particular, the result for the integral intensity of the Stokes spontaneous Raman scattering radiation is such as if the scattering were to occur only by the transverse polaritons, under the additional condition $\mathrm{Im}\,\epsilon_p = \mathrm{Im}\,\chi_{ijk} = \mathrm{Im}\,\gamma_{ijkm} = 0$. A non-trivial consideration is here the fact that not only $\mathrm{Im}\,\epsilon_p$ but also $\mathrm{Im}\,\chi_{ijk}$ and $\mathrm{Im}\,\chi_{ijkm}$ fail to exert any influence here. It would of course be a mistake to attempt to obtain results for other quantities at $M \neq 1$ while ignoring the anomalous longitudinal polariton waves. An example is the spectrum of g_p in the region of small θ, in which, if no account is taken of the longitudinal waves, a large extra maximum at the frequency ω_f would appear in addition to the polariton maximum. On going over to large θ, this extra maximum would merge with the polariton maximum, which moves towards ω_f, and the resultant value of g_f would differ from (21) in that there would be no factor M, i.e., the value of g_f would turn out to be the same as for the nonpolar phonons.

[10]If $\tilde{\gamma}$ is independent of the frequency, the lines p_f and p_L have Lorentz shapes with equal half-widths. We note also that Faust et al.[17] give a result analogous to (26), but without the factor $\Lambda/(1-\Lambda)$; their result is therefore valid only if $\Lambda = 1/2$. In the experiment described in[17] the condition $\Lambda = 1/2$ is satisfied, so that the value $A \approx -1.9$ obtained for GaP is correct.

2. CASE OF UNIAXIAL CRYSTALS

We proceed now to investigate SRS by polariton in uniaxial crystals. We confine ourselves to a scattering geometry in which only one of the two possible polarizations of the Stokes radiation is active (the o- or e-wave). This covers most of the chosen geometries of greatest interest. We seek the Stokes field in the form $\mathbf{E}_s(\mathbf{r}, t) = \mathbf{e}_s A_s \exp[i(\mathbf{k}_s \cdot \mathbf{r} - \omega_s t)] + \text{c.c.}$, $e_s = 1$, and the polariton field in the same form (1) as before. We choose the unit vectors $\mathbf{e}_p^{(1,2)}$ in the following manner: $\mathbf{e}_p^{(1)}$ is perpendicular to the plane σ_0 passing through $\mathbf{w}$ and the optical axis C of the crystal, while $\mathbf{e}_p^{(2)}$ lies in the plane σ_0.

The shortened equations for the amplitudes $A_{s,p}$ are obtained in standard fashion[9], but unlike in the preceding section it is necessary to take into account the anisotropy of the dielectric tensor ϵ_p. We denote the principal values of ϵ_p along the C axis and in the plane Σ perpendicular to it by $\epsilon_\parallel$ and $\epsilon_\perp$, respectively. We seek the solution of the amplitude equations, as before, in the form $A_s = B_s e^{\kappa z}$, $A_p^{(\sigma)*} = B_\sigma e^{\kappa z}$ ($\sigma = 1, 2, 3$), assuming that B and κ are independent of z. We write out immediately the algebraic equations satisfied by the quantities B:

$$\begin{gathered}\rho_1 B_1 - \rho_0\chi_1 B_s = 0,\qquad \rho_2 B_2 + \rho_3 B_3 - \rho_0\chi_2 B_s = 0,\\ \Delta B_3 - \rho_3 B_2 + \rho_0\chi_3 B_s = 0,\qquad (ik_s{}^z\varkappa + 2\pi q_s{}^2\gamma|A_l|^2)B_s\\ + 2\pi q_s{}^2 A_l(\chi_1 B_1 + \chi_2 B_2 + \chi_3 B_3) = 0.\end{gathered} \qquad (27)$$

Here

$$\rho_0 = 4\pi A_l{}^*,\quad \rho_1 = \mu^2 - \varepsilon_\perp{}^*,\quad \rho_2 = \mu^2 - \varepsilon_\perp{}^*\cos^2\xi - \varepsilon_\parallel{}^*\sin^2\xi,$$
$$\rho_3 = {}^1/_2(\varepsilon_\perp{}^* - \varepsilon_\parallel{}^*)\sin 2\xi, \qquad (28)$$
$$\Delta = \varepsilon_\perp{}^*\sin^2\xi + \varepsilon_\parallel{}^*\cos^2\xi,\quad \chi_\sigma = e_s{}^i e_l{}^j e_p^{(\sigma)k}\chi_{ijk}(\omega_{l_s} - \omega_p),\quad \sigma = 1,2,3;$$

ξ is the angle between $\mathbf{w}$ and the optical axis.

Equating the determinant of the system (27) to zero, we obtain κ and then $g = 2\,\mathrm{Re}\,\kappa$:

$$g = -\tilde{g}\,\mathrm{Im}\Big\{\gamma + 4\pi\Big[\frac{\chi_1{}^2}{\mu^2-\varepsilon_\perp{}^*} + \frac{\Delta\chi_2{}^2 - \rho_2\chi_3{}^2 + 2\rho_3\chi_2\chi_3}{\Delta(\mu^2 - n_{pe}{}^{*2})}\Big]\Big\}, \qquad (29)$$

where

$$\tilde{g} = 8\pi^2\omega_s I_l / c^2 n_l n_s\cos\theta,\qquad n_{pe}{}^2 = \varepsilon_\perp\varepsilon_\parallel/\Delta^*;$$

n_{pe} is the refractive index of the extraordinary polariton wave.

We present also formulas connecting $B_{1,2,3}$ with B_s; these may be useful in the estimate of the relative intensities of the polariton radiation:

$$B_1 = \frac{\rho_0\chi_1}{\rho_1}B_s,\ B_2 = \frac{\rho_0(\Delta\chi_2 + \rho_3\chi_3)}{\Delta(\mu^2 - n_{pe}{}^{*2})}B_s,\ B_3 = \frac{\rho_0(\rho_3\chi_2 - \rho_2\chi_3)}{\Delta(\mu^2 - n_{pe}{}^{*2})}B_s.$$

We introduce unit vectors that are singled out by the crystal structure and the geometry of the problem, namely **a** and **b** along the component of **w** on the Σ plane and along the optical axis, respectively. We resolve $e_p^{(2)}$ and $e_p^{\|}$ along the directions of **a** and **b**: $e_p^{(2)} = -\mathbf{a}\cos\xi + \mathbf{b}\sin\xi$, $e_p^{\|} = \mathbf{a}\sin\xi + \mathbf{b}\cos\xi$. Using these resolutions in the expressions for $\chi_{2,3}$ (28), we transform g (29) into

$$g = -\bar{g}\,\mathrm{Im}\Big[\gamma + 4\pi\Big(\frac{\chi_1^2}{\mu^2-\varepsilon_\perp^*} + \frac{\rho_a\chi_a^2+\rho_b\chi_b^2+2\rho_{ab}\chi_a\chi_b}{\mu^2-n_{pe}^{*2}}\Big)\Big]. \quad (30)$$

Here

$$\chi_a = e_s^i e_l^j a^k \chi_{ijk}, \quad \chi_b = e_s^i e_l^j b^k \chi_{ijk};$$
$$\rho_a = (\varepsilon_\|^* - \mu^2\sin^2\xi)/\Delta, \quad \rho_b = (\varepsilon_\perp^* - \mu^2\cos^2\xi)/\Delta,$$
$$\rho_{ab} = -\mu^2\sin 2\xi/2\Delta.$$

Accordingly

$$B_2 = \frac{\rho_0[(-\Delta\cos\xi+\rho_3\sin\xi)\chi_a + (\Delta\sin\xi+\rho_3\cos\xi)\chi_b]}{\Delta(\mu^2-n_{pe}^{*2})}B_s,$$

$$B_3 = \frac{\rho_0[-(\rho_3\cos\xi+\rho_2\sin\xi)\chi_a + (\rho_3\sin\xi-\rho_2\cos\xi)\chi_b]}{\Delta(\mu^2-n_{pe}^{*2})}B_s.$$

We consider further the vicinity of an isolated phonon oscillation and discuss the most important particular cases.

Let the scattering geometry be such that $\chi_1, \chi_a \neq 0$, $\chi_b = 0$. These conditions are realized in the vicinity of a doubly degenerate dipole active oscillation; such oscillations are polarized in the Σ plane. For this case, we can represent g (30) in the form

$$g = -\bar{g}\,\mathrm{Im}\Big[\gamma + 4\pi\Big(\frac{\chi_1^2}{\mu^2-\varepsilon_\perp^*} + \frac{\chi_a^2}{\zeta_a-\varepsilon_\perp^*}\Big)\Big].$$

where

$$\zeta_a = \frac{\mu^2\varepsilon_\|^*\cos^2\xi}{\varepsilon_\|^* - \mu^2\sin^2\xi} = \zeta_a' + i\zeta_a''. \quad (31)$$

We use furthermore for $\chi_{1,a}$ and γ the formulas (9)–(11) of the microscopic theory, which are valid in an arbitrary anisotropic crystal. The quantity χ_1^2, as is clear from (9), is proportional to η_1. Adding to it the quantity $\eta_b = [\sum_\nu \alpha^{(\nu)}(\mathbf{b}, \mathbf{e}_\nu)]^2$, which is equal to zero (we recall that $\mathbf{b} \perp \mathbf{e}_\nu$, $\nu = 1, 2$), we use formulas (14) in which we replace $e_p^{(2)}$ and $e_p^{\|}$ by the vectors **b** and **a**, respectively. As a result we get

$$g = \frac{g_0}{1+\varphi^2}\Big[\frac{(\bar\varphi_1+\tau_1)^2 M}{1+\tau_1^2} + \frac{(\bar\varphi_a+\tau_a)^2\Lambda}{1+\tau_a^2}\Big]. \quad (32)$$

Here

$$\bar\varphi_{1,a} = \varphi + \beta x A_{1,a}(1+\varphi^2), \quad A_1 = \Big(\frac{2\pi v_0\hbar\omega_f}{s_f}\Big)^{1/2}$$
$$\times \chi_1^0 \Big/ \sum_\nu \alpha^{(\nu)}(e_p^{(1)}, e_\nu), \quad \chi_1^0 = e_s^i e_l^j e_p^{(1)k}\chi_{ijk}^0(\omega_l, -\omega_p);$$

and a similar formula, with $e_p^{(1)}$ replaced by **a**, holds for A_a. The quantities β and x are defined in accordance with (13), and g_0 in accordance with (19), with allowance for the fact that the index ν runs in this case through two rather than three values, since the oscillations ω_f is doubly degenerate. Further,

$$\tau_1 = \frac{\mu^2-\varepsilon_\perp'}{\varepsilon_\perp''}, \quad \tau_a = \frac{\zeta_a'-\varepsilon_\perp'}{\zeta_a''+\varepsilon_\perp''}, \quad M = \frac{\eta_1}{\eta} = 1-\Lambda, \quad \Lambda = \frac{\eta_a}{\eta},$$
$$\eta_a = \Big[\sum_\nu \alpha^{(\nu)}(\mathbf{a}, \mathbf{e}_\nu)\Big]^2.$$

The analysis of (32) is similar to that used in the preceding section for formula (19). The first term describes a line whose frequency position at fixed θ corresponds to the condition $\tau_1 = \varphi^{-1}$. This condition defines, in the coordinates (k_p, ω_p) or (θ, ω_p), the dispersion curve of the o-polaritons that might be produced in the scattering process in the absence of scattering and wave mismatch. We denote this curve by $\Pi_\perp$, and the line itself by $p_\perp$.[11] For this line $B_1 \neq 0$ and $B_{2,3} \approx 0$.

The maxima of g, corresponding to the second term of (32), are located, generally speaking, in the nonresonant region[12] and correspond to the condition $\tau_a \approx 0$, which is approximately equivalent here to the condition $\zeta_a - \epsilon_\perp = 0$ or $\Delta\Delta_e(\epsilon_\| - \mu^2\sin^2\xi)^{-1} = 0$, where $\Delta_e = \mu^2 - n_{pe}^2$. Putting $\Delta_e = 0$, we obtain the line p_e for scattering by extraordinary polaritons. At $\xi = 90°$ we can put $\Delta = 0$, i.e., $\epsilon_\perp = 0$, thus defining the line $p_{L\perp}$. In the case $0 \leq \xi \leq 90°$ at $\Delta = 0$, we have the product $\Delta\Delta_e(\xi_\| - \mu^2\sin^2\xi)^{-1} \neq 0$. Therefore, in particular, the line $p_{L\|}$ is inactive. The line $p_{L\perp}$ exists, for example, in the case of scattering in the Σ plane, and for it we have $B_{1,2} \approx 0$ and $B_3 \neq 0$.

Finally, the line p_e vanishes at $\xi = 90°$, for in this case we have simultaneously with $\Delta_e = 0$ also $\epsilon_\| - \mu^2\sin^2\xi = 0$. At $\xi = 0$ the line merges with $p_\perp$. In the region of intermediate ξ at sufficiently large θ, when μ increases appreciably, the line p_e is transformed into the line for scattering by "quasilongitudinal"[18] phonons, and its position is then determined approximately by the condition $\Delta = 0$. The latter follows from the relation $\Delta\Delta_e(\epsilon_\| - \mu^2\sin^2\xi)^{-1} = 0$, if $\mu^2\sin^2\xi \gg |\epsilon_\||$.

Thus, in accordance with the presence of three independent components of the amplitude of the polariton wave $A_p^{(\sigma)}$, three types of scattering line are produced in general: for ordinary, extraordinary, and longitudinal polaritons. As to the latter, we can have, just as in cubic crystals, excitation of normal longitudinal polariton waves (i.e., ordinary longitudinal phonons), and anomalous longitudinal waves maintained by the pump field near the frequencies of the mechanical phonons.

The conditions $\chi_{1,a} \neq 0$ and $\chi_b = 0$, at a suitable scattering geometry, can be satisfied in an arbitrary uniaxial crystal without an inversion center. Since $\chi_\alpha \sim \eta_\alpha$, this calls for $\eta_\alpha = 0$. For example, in crystals of class C_{3v} in a scattering geometry wherein $\mathbf{k}_l$ lies in the Σ plane, the scattering takes place in the plane passing through $\mathbf{k}_l$ and the C axis, $\mathbf{e}_l$ defines the e-wave, and $\mathbf{e}_s$ defines the o-wave, we get $\eta_\alpha = d^2\sin^2 2\beta_0$, where β_0 is the angle between $\mathbf{k}_l$ and the x axis chosen in accordance with[16], and d is the value of the nonzero component of the tensor α_{ij}[14]. In this case $M = \cos^2 2\beta_0$[6].

If the scattering takes place in the Σ plane and $\mathbf{e}_l$ corresponds as before to the e-wave and $\mathbf{e}_s$ to the o-wave, then $\eta_\alpha = d^2\sin^2(\theta+\theta_p)$ and $M = \cos^2(\theta+\theta_p)$. At large θ, when $\theta_p \approx (\pi-\theta)/2$, we have $M = \sin^2(\theta/2)$. As a result we get the characteristic front-back

[11]The analogous dispersion curve of the e-polaritons will be designated Π_e, and the corresponding scattering line p_e. In scattering by e-polaritons in the Σ plane, when $n_{pe}^2 = \epsilon_\|$, we use the notation $\Pi_\|$ and $p_\|$. Finally, the lines for scattering by longitudinal phonons, whose positions are determined by the conditions $\epsilon_\perp = 0$ and $\epsilon_\| = 0$, will be designated $p_{L\perp}$ and $p_{L\|}$, respectively.

[12]An exception is the line p_e at $\xi = 0$ (see below).

asymmetry of the phonon scattering, due to the dipole activity of the phonons.

We consider now a geometry in which $\chi_{1,a} = 0$ but $\chi_b \neq 0$. These conditions can be realized in the vicinity of nondegenerate dipole active oscillations polarized along the C axis. In this case

$$g = -\tilde{g}\,\mathrm{Im}\left(\gamma + \frac{4\pi\chi_b^2}{\zeta_b - \varepsilon_\parallel^*}\right),$$

where ζ_b is defined by the same formula (31) as ζ_a, except that $\epsilon_\parallel$ and $\epsilon_\perp$ are replaced by ξ and $\pi/2 - \xi$. In analogy with (32), we also obtain

$$g = \frac{g_0}{1+\varphi^2}\frac{(\tilde{\varphi}_b + \tau_b)^2}{1+\tau_b^2}, \quad \tilde{\varphi}_b = \varphi + \beta x A_b(1+\varphi^2),$$

$$A_b = \frac{\chi_b^0}{\alpha}\left(\frac{2\pi v_0 \hbar\omega_f}{s_f}\right)^{1/2}, \quad \tau_b = \frac{\zeta_b' - \varepsilon_\parallel'}{\zeta_b'' + \varepsilon_\parallel''}.$$

The maxima of g are determined by the condition $\tau_b = \varphi^{-1}$.

The analysis of the scattering spectrum is similar to the preceding one; we therefore confine ourselves to brief remarks in the discussion of the present case. In the nonresonant region, the condition that determines the maximum of the scattering line is approximately equivalent to $\Delta\Delta_e(\epsilon_\perp - \mu^2 \cos^2 \xi)^{-1} = 0$. The result is the line p_e, which corresponds to the condition $\Delta_e = 0$, and in the case of $\xi = 0$ we get the line $p_{L\parallel}$, which corresponds to $\Delta = 0$, i.e., $\epsilon_\parallel = 0$. For the line $p_{L\parallel}$ we have $B_1 \approx 0$, $B_2 = 0$, and $B_3 \neq 0$. There is no p_e line at $\xi = 0$, and in the case of scattering in the Σ plane ($\xi = 90°$) the p_e line goes over into $p_\parallel$. The behavior of the latter is equivalent to the behavior of the line $p_\perp$. At large θ we have for this line $g = g_0(1 + \varphi^2)^{-1}$.

Cases of excitation of purely transverse polaritons in anisotropic crystals were considered in[4]. It follows from the present results that such cases are realized in the vicinity of the phonon frequencies if the phonon oscillation is nondegenerate; for degenerate oscillations this is realized at geometries satisfying the condition $\eta_1 = \eta$, i.e., $M = 1$ and $\Lambda = 0$. The results obtained in[4] are therefore applicable in the resonant region only in these cases. In the nonresonant region, these limitations do not exist, since no longitudinal polaritons are excited here, with the exception of the region of the lines $p_{L\perp,\parallel}$.

The results obtained in this section, which pertain to uniaxial crystals, can be easily generalized also to the case of biaxial crystals.

The author is indebted to Professor R. V. Khokhlov and Professor F. V. Bunkin for a discussion of the results.

[1] R. Loudon, Proc. Phys. Soc. Lond. **82**, 393 (1963); P. N. Butcher, R. Loudon, and T. P. McLean, Proc. Phys. Soc. Lond. **85**, 565 (1965); Y. R. Shen, Phys. Rev. A **138**, 1741 (1965); F. De Martini, Nuovo Cimento **51**, 16 (1967).

[2] C. H. Henry and C. G. B. Garrett, Phys. Rev. **171**, 1058 (1968).

[3] V. V. Obukhovskiĭ, V. L. Strizhevskiĭ, and G. É. Ponat, Nelineĭnye protsessy v optike (Nonlinear Processes in Optics), Nauka, Novosibirsk, 1970, P. 109.

[4] V. L. Strizhevskiĭ, V. V. Obukhovskiĭ, and G. É. Ponat, Zh. Eksp. Teor. Fiz. **61**, 537 (1971) [Sov. Phys.-JETP **34**, 286 (1972)].

[5] V. L. Strizhevskiĭ, G. É. Ponat, and Yu. N. Yashkir, Preprint, Theoret. Phys. Inst. Ukr. Acad. Sci., Kiev, 1971. Strizhevskiĭ, Opt. Spektrosk. **31**, 831 (1971).

[7] V. M. Agranovich and V. L. Ginzburg, Kristallooptika s uchetom prostranstvennoĭ dispersii i teoriya éksitonov (Spatial Dispersion in Crystal Optics and the Theory of Excitons), Nauka, 1965.

[8] V. L. Strizhevsky and V. V. Obukhovsky, Phys. Stat. Sol. (in print).

[9] S. A. Akhmanov and R. V. Khokhlov, Problemy nelineĭnoĭ optiki (Problems of Nonlinear Optics) AN SSSR, 1964.

[10] N. Bloembergen, Nonlinear Optics, Benjamin, 1965.

[11] V. M. Faĭn and Ya. I. Khanin, Kvantovaya radiofizika (Quantum Radiophysics), Sov. Radio, 1965.

[12] V. L. Strizhevskiĭ and V. V. Obukhovskiĭ, in Nelineĭnaya optika (Nonlinear Optics), Nauka, 1968, p. 423. V. V. Obukhovskiĭ and V. L. Strizhevskiĭ, Zh. Eksp. Teor. Fiz. **57**, 520 (1969) [Sov. Phys.-JETP **30**, 285 (1970)].

[13] V. L. Strizhevskiĭ and E. I. Kondilenko, Opt. Spektrosk. **30**, 238 (1971).

[14] M. M. Sushchinskiĭ, Spektry kombinatsionnogo rasseyaniya molekul i kristallov (Raman Spectra of Molecules and Crystals), Nauka, 1969.

[15] V. L. Strizhevskiĭ, Fiz. Tverd. Tela **3**, 2929 (1961) [Sov. Phys.-Solid State **3**, 2141 (1962)]; Fiz. Tverd. Tela **4**, 1492 (1962)

[16] J. F. Nye, Physical Properties of Crystals, Oxford, 1957.

[17] W. L. Faust and C. H. Henry, Phys. Rev. Lett. **17**, 1265 (1966); W. L. Faust, C. H. Henry, and R. H. Eick, Phys. Rev. **173**, 781 (1968).

[18] R. Loudon, Adv. Phys. **13**, 423 (1964); T. C. Damen, S. P. S. Porto, and B. Tell, Phys. Rev. **142**, 570 (1966).

Translated by J. G. Adashko
168