THEORY OF STIMULATED RAMAN SCATTERING OF LIGHT BY POLARITONS

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A general fluctuation-dissipation theory of stimulated Raman scattering (SRS) by polaritons is developed. The theory is based on application of the abbreviated-equation method for the scalar amplitudes of scattered Stokes and polariton waves. Sources responsible for wave scattering are introduced. These may be quantum fluctuations of the specific polarization of the medium. Formulas describing the frequency-angular distribution of the Stokes and polariton waves are derived under the assumption that the medium is transparent at the exciting and Stokes frequencies. The cases of weak and strong absorption at polariton frequencies are analyzed in detail. The vicinity of phonon resonance is also considered. Formulas are found which express the Stokes wave gain factor in this region in terms of macroscopic parameters of the medium. It is shown that for an isolated phonon oscillation the frequency position of the SRS line peak corresponds to the dispersion curve that the polaritons would have during scattering if absorption and wave mismatching were disregarded. The general theory is applied to zinc oxide, quartz, and lithium niobate crystals. Several features of SRS in these crystals are revealed as a result of numerical calculations. For lithium niobate, for which experimental data are available, the theory agrees qualitatively with the experiments.

THE present paper is devoted to the theory of simulated Raman scattering (SRS) of light by polaritons in crystals in the absence of resonators. SRS from polaritons was recently registered experimentally[1-3]. The theoretical aspects of this phenomenon were considered in[4,5]. One of the most important questions, however, namely the calculation of the scattering intensity, has remained uninvestigated.

We have developed a general theory of SRS on polaritons, and its exposition is the main purpose of the present article. Its main results reduce to a determination of the frequency-angle distribution of the scattering intensity of the Stokes and polariton waves at the exit face of the crystal. We also obtain a simple general expression for the gain (see Sec. 4), which is valid in a much wider range than the results of other investigations. The analysis is carried out in the approximation of a given stationary pump field, which is approximated by a linearly polarized plane monochromatic wave. The medium is assumed to be nonmagnetic, weakly anisotropic, and transparent at the pump frequency ω_l and at the Stokes frequency ω_S. At the same time, account is taken of the absorption at the polariton frequency, which is usually much more significant (if we disregard the question of the SRS threshold). We consider the case of excitation of transverse polaritons. The results are then used to analyze concrete cases.

1. GENERAL FLUCTUATION-DISSIPATION THEORY

Assume that in a plane-parallel layer of volume V, bounded by the planes $z = 0$ and $z = l$ and occupied by a crystalline medium without inversion center, there acts a classical pump field propagating along the z axis

$$\mathbf{E}_L(\mathbf{r},t) = \mathbf{A}_l e^{i(k_l z - \omega_l t)} + \text{c.c.}, \quad \mathbf{A}_l = \mathbf{e}_l A_l, \quad e_l = 1;$$
$$k_l = \omega_l c^{-1} n_l.$$

To get around inessential complications, we assume henceforth the following model. The scattering layer is placed in an unbounded medium, in which all characteristics are the same as in the layer, but there is neither nonlinearity nor absorption. This enables us, in particular, to avoid the transition from scattering angles in the crystal to external angles, to introduce scattering directions without allowance for absorption, etc.

The Stokes and polariton fields $\mathbf{E}_{S,p}(\mathbf{r}, t)$ will be represented in the form of Fourier integrals in terms of the variables x, y, and t, introducing positive-frequency and negative-frequency parts:

$$\begin{gathered}\mathbf{E}_{s,p}(\mathbf{r},t) = \mathbf{E}_{s,p}^{(+)}(\mathbf{r},t) + \mathbf{E}_{s,p}^{(-)}(\mathbf{r},t), \quad \mathbf{E}_{s,p}^{(-)} = \mathbf{E}_{s,p}^{(+)+}, \\ \mathbf{E}_{s,p}^{(+)}(\mathbf{r},t) = \int\limits_0 d\omega_{s,p} \int d^2\mathbf{k}_{s,p}^{\tau} \mathbf{A}_{s,p}(z, \mathbf{k}_{s,p}^{\tau}\, \omega_{s,p}) \cdot \\ \times \exp[i(\mathbf{k}_{s,p}^{\tau}\mathbf{r}^{\tau} - \omega_{s,p} t)], \quad \mathbf{k}_{s,p}^{\tau} = (k_{s,p}^{x}, k_{s,p}^{y}), \quad \mathbf{r}^{\tau} = (x, y).\end{gathered} \tag{1}$$

All the field quantities pertaining to the Stokes and the polariton waves are operator quantities (in the Heisenberg representation). In connection with the limited dimensions of the nonlinear medium along the z axis, there is no Fourier expansion with respect to this variable.

We assume that the only interacting waves are those with fixed linear polarizations defined by the unit vectors $\mathbf{e}_{S,p}$. Accordingly, we seek the solutions of Maxwell's equations in the form $\mathbf{A}_{S,p} = \mathbf{e}_{S,p} A_{S,p}$. Instead of the scalar amplitudes $A_{S,p}$ we introduce new unknowns $\xi_{S,p}$ defined by

$$\begin{gathered}A_s(z, \mathbf{k}^{\tau}{}_s, \omega_s) = \xi_s(z, \mathbf{k}^{\tau}{}_s, \omega_s)\exp[i(k^z{}_s + \Delta k/2)z], \\ A^+{}_p(z, -\mathbf{k}^{\tau}{}_s, \omega_p) = \xi^+{}_p(z, -\mathbf{k}^{\tau}{}_s, \omega_p)\exp[-i(k^z{}_p + \Delta k/2)z], \\ k_{s,p}^z = [k_{s,p}^2 - (k_s^{\tau})^2]^{1/2}, \quad \Delta k = k_l - k_s{}^z - k_p{}^z.\end{gathered} \tag{2}$$

Here $k_S = \omega_S n_S/c$ is the length of the wave vector at the Stokes frequency in the unpumped medium, and $k_p = \omega_p n_p^0/c$, where $n_p^0 = \sqrt{\epsilon_p', \epsilon_p'} = \text{Re}\, \epsilon_p$. Introducing

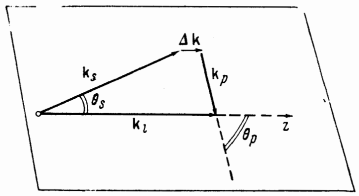

FIG. 1

the angles $\theta_{s,p}$ between the z axis and the vectors $\mathbf{k}_s = (k_s^\tau, k_s^z)$ and $\mathbf{k}_p = (k_p^\tau, k_p^z)$, respectively (see Fig. 1), we can write $k_{s,p}^z = k_{s,p}\cos\theta_{s,p}$. The angles $\theta_{s,p}$ are connected by the relation $k_s \sin\theta_s = k_p \sin\theta_p$. We have taken into account the fact that the wave-vector components that are tangential with respect to the faces are conserved, and that the problem is stationary, and therefore only waves for which $\mathbf{k}_p^\tau = -\mathbf{k}_s^\tau$ and $\omega_p = \omega_l - \omega_s$ interact.

An essential feature of our problem is the need for introducing sources responsible for the occurrence of the scattering. Such sources are the quantum fluctuations $d_{s,p}$ of the per-unit polarization of the medium $\mathbf{P}_{s,p}(\mathbf{r}, t)$, which do not depend on the pump. The polarization can be represented in the form $\mathbf{P}_{s,p} = \mathbf{d}_{s,p} + \kappa_{s,p}\mathbf{E}_{s,p} + \mathbf{P}_{s,p}^{NL}$, where $\kappa_{s,p}$ is the linear polarizability of the medium and $\mathbf{P}_{s,p}^{NL}$ is the nonlinear part of the polarization, which connects the waves $\mathbf{E}_{s,p}$.

The amplitudes ξ_s and ξ_p^+ satisfy a system of two coupled equations, which is obtained by using the standard procedure of abbreviated equations[8,9]

$$\partial\xi_s/\partial z = \chi R_s \xi^+_p + Q_s\xi_s + f_s, \quad \partial\xi^+_p/\partial z = \chi R^*_p \xi_s + Q_p\xi^+_p + f^+_p. \tag{3}$$

We have introduced here the notation

$$R_{s,p} = \frac{2\pi i\omega_{s,p}^2 A_l}{k_{s,p}c^2\cos\theta_{s,p}}, \quad \chi = e_s^i e_l^j e_p^k \chi_{ijk}(\omega_l, -\omega_p),$$
$$Q_s = \Gamma_s - i\frac{\Delta k}{2}, \quad \Gamma_s = \frac{2\pi i\gamma\omega_s^2|A_l|^2}{k_s c^2\cos\theta_s}, \quad \gamma = e_s^i e_s^j e_l^k e_l^m \gamma_{ijkm}, \tag{4}$$
$$Q_p = i\frac{\Delta k}{2} - \alpha_p, \quad \alpha_p = \frac{\omega_p^2\varepsilon_p''}{2c^2k_p\cos\theta_p},$$

$\chi_{ijk}(\omega_l, -\omega_p)$ and $\gamma_{ijkm}(\omega_s, \omega_l, -\omega_l)$ are the corresponding nonlinear polarizabilities. Finally,

$$f_{s,p} = \frac{2\pi i\omega_{s,p}^2(\mathbf{e}_{s,p}\mathbf{d}_{s,p})\exp[-i(k_{s,p}^z + \Delta k/2)z]}{c^2k_{s,p}\cos\theta_{s,p}}.$$

The quantities ξ_s and d_s correspond to the arguments $(z, \mathbf{k}_s^\tau, \omega_s)$, while ξ_p^+ and d_p^+ correspond to the arguments $(z, -\mathbf{k}_s^\tau, \omega_p)$. In the case when $\epsilon_p'' \ll \epsilon_p'$, the parameter $2\alpha_p$ has the meaning of the absorption coefficient at the frequency ω_p. We use also the symmetry property $\chi_{ijk}(\omega_l, -\omega_p) = \chi^*_{kji}(\omega_l, -\omega_s)$[9].

We shall need in what follows only the following mean values of the quadratic combinations of the operators $d_{s,p}$, which were obtained in[6,7]:

$$\langle d^+_{pi}(z', \mathbf{k}'^\tau_p, \omega'_p)\, d_{pj}(z, \mathbf{k}^\tau_p, \omega_p)\rangle = 0,$$
$$\langle d_{pi}(z', \mathbf{k}'^\tau, \omega_p')\, d_{pj}^+(z, \mathbf{k}_p^\tau, \omega_p)\rangle = \tag{5}$$
$$= \frac{\hbar\varepsilon''_{pij}}{16\pi^4}\delta(z - z')\,\delta(\mathbf{k}_p^\tau - \mathbf{k}_p'^\tau)\,\delta(\omega_p - \omega_p')$$

(we neglect the thermal excitations), and analogously for d_s.

The general solution of the inhomogeneous system of equations (3) can be written as the sum of the general solution $\tilde\xi_s$, $\tilde\xi_p^+$ of the corresponding homogeneous system and the particular solution ξ_s' and $\xi_p'^+$ of the inhomogeneous system. The equations for $\tilde\xi$ are of the form

$$\tilde\xi_s = C_s^{(1)}e^{q_1 z} + C_s^{(2)}e^{q_2 z}, \quad \tilde\xi_p^+ = C_p^{(1)}e^{q_1 z} + C_p^{(2)}e^{q_2 z},$$

where

$$q_{1,2} = \tfrac{1}{2}[\Gamma_s - \alpha_p \pm \sqrt{(\Gamma_s + \alpha_p - i\Delta k)^2 + 4\chi^2 R}],$$
$$R = 8\pi^3\omega^2_s\omega^2_p I_l / c^5 n_l k_s k_p \cos\theta_s\cos\theta_p, \tag{6}$$

and $C_{s,p}^{(1,2)}$ are integration constants, of which obviously only two are independent. It is convenient to take the particular solution in the form

$$\xi_s' = \exp(Q_s z)\int_0^z \exp(-Q_s z_1) f_s(z_1)\,dz_1 + \exp(q_1 z)\int_0^z dz_1$$
$$\times\exp\{-(2q_1 - Q)z_1\}\int_0^{z_1} dz_2 L(z_2)\exp\{(q_1 - Q)z_2\}; \quad Q = Q_s + Q_p,$$
$$L(z) = \chi R_s f_p^+ + \chi^2 R_s R_p \exp(Q_s z)\int_0^z \exp(-Q_s z_1) f_s(z_1)\,dz_1, \tag{7}$$
$$\xi'^+_p = (\chi R_s)^{-1}[\partial\xi'_s/\partial z - Q_s\xi'_s - f_s].$$

To determine the constants $C_{s,p}^{(1,2)}$ it is necessary to make use of the boundary conditions at the entrance into the medium. We stipulate that at $z = 0$[1)] the quantities $\xi_{s,p}$ and $\xi_{s,p}^+$ coincide with the operators $\xi_{s,p}^0$ and $\xi_{s,p}^{0+}$, which are independent of the pump, for the production and annihilation of photons $(\mathbf{k}_{s,p}, \omega_{s,p})$ outside the scattering layer, i.e., at $z = -0$. The latter are statistically independent with respect to $\xi_{s,p}'$.

Using the well-known results of quantization of a long-wave electromagnetic field in a transparent dispersive crystalline medium[10], it is easy to find the following correlators, which we shall find useful later on:

$$\langle \xi_{s,p}^0(0, \mathbf{k}'^\tau, \omega_{s,p}')\,\xi_{s,p}^{0+}(0, \mathbf{k}_{s,p}^\tau, \omega_{s,p})\rangle = \frac{\hbar\omega_{s,p}^2\delta(\mathbf{k}_{s,p}'^\tau - \mathbf{k}_{s,p}^\tau)\,\delta(\omega_{s,p}' - \omega_{s,p})}{4\pi^2c^2k_{s,p}\cos\theta_{s,p}}. \tag{8}$$

At the same time, the correlators of the type $\langle \xi_{s,p}^{0+}\xi_{s,p}^0\rangle$ vanish. Taking the boundary conditions into account, we obtain

$$\xi_s = \frac{\chi R_s\xi_p^{0+} - (q_2 - Q_s)\xi_s^0}{q_1 - q_2}e^{q_1 z} + \frac{\chi R_s\xi_p^{0+} - (q_1 - Q_s)\xi_s^0}{q_2 - q_1}e^{q_2 z} + \xi_s'. \tag{9}$$

Our end purpose is to calculate the frequency-angle distribution of the radiation on emerging from the medium. This distribution can be described by introducing the luminosity of the exit face at the frequencies of the Stokes wave, $\Phi_s = \langle S_z\rangle$. Here S_z is the component of the Poynting vector taken on the outside of the exit face, i.e., at points $z = l + 0$. The Poynting vector can be set in correspondence with the operator

$$\mathbf{S} = \frac{c}{4\pi}[\mathbf{E}_0^{(-)}\mathbf{H}_0^{(+)}] + \text{h.c.} \tag{10*}$$

1)During the scattering process, polariton waves traveling in both the forward ($\theta_p < 90°$) and in the backward ($\theta_p > 90°$) directions can be excited. In the latter case the condition $\xi_p^+ = \xi_p^{0+}$ must be imposed at $z = l$. We assume, for concreteness, that $\theta_p < 90°$. The results remain valid also when $\theta_p > 90°$, provided $\cos\theta_p$ is replaced in all formulas by $|\cos\theta_p|$, and z is replaced in the expressions for the fields and for the polariton-wave intensities by l-z.

*$[\mathbf{E}_0^{(-)}\mathbf{H}_0^{(+)}] \equiv \mathbf{E}_0^{(-)} \times \mathbf{H}_0^{(+)}$.

(The subscript zero denotes here the field outside the layer). Using the fact that the dispersion law outside the layer is such that the Fourier components of the field $\mathbf{E}_0(\mathbf{r}, t)$ can be represented in the form $\mathbf{E}_0(\mathbf{k}, \omega) = \mathbf{E}_0'(\mathbf{\Omega}, \omega)\delta(k - k_S)$, $\mathbf{\Omega} = \mathbf{k}/k$, and taking into account the boundary conditions, the stationary character of the problem, and its homogeneity in the xy plane, we can readily reduce Φ_S to the form

$$\Phi_s = \int_0^\infty d\omega_s \int d\Omega_s \cos\theta_s B_s(\Omega_s, \omega_s). \tag{11}$$

Here $d\Omega_S$ is the solid-angle element in $\mathbf{k}_S$-space and $B_S(\mathbf{\Omega}_S, \omega_S)$ is the spectral density of the surface brightness at the exit face,

$$B_s(\Omega_s, \omega_s) = \frac{cn_s k_s^2 \cos\theta_s}{2\pi} \varphi_s(\Omega_s, \omega_s). \tag{12}$$

The function φ_S is defined by the condition

$$\langle A_s^+(l, \mathbf{k}_s'^{\tau}, \omega_s') A_s(l, \mathbf{k}_s, \omega_s)\rangle = \varphi_s(\Omega_s, \omega_s)\delta(\mathbf{k}_s'^{\tau} - \mathbf{k}_s^{\tau})\delta(\omega_s' - \omega_s). \tag{13}$$

To find the explicit form of the function φ_S, we use the solution obtained above for A_S, which is determined by formulas (2), (6), (7), and (9), and also the values of the nonvanishing correlators (5) and (8). As a result we get

$$\varphi_s(\Omega_s, \omega_s) = \frac{\hbar\omega_p^2 |\chi R_s|^2}{4\pi^2 c^2 k_p \cos\theta_p} \left\{ \left| \frac{e^{q_1 l} - e^{q_2 l}}{q_1 - q_2} \right|^2 + \frac{\alpha_p}{\beta}\left(\frac{e^{gl} - 1}{g\mu^*} + \frac{e^{gl} - e^{\mu^* l}}{\mu^* \beta^*} + \frac{e^{\mu l} - 1}{\mu\nu} + \frac{e^{\mu l} - e^{-\nu l}}{\nu\beta^*} \right) \right\}, \tag{14}$$

$$g = 2\operatorname{Re} q_1, \quad \beta = Q - 2q_1, \quad \mu = Q - 2i \operatorname{Im} q_1, \quad \nu = -g - 2\operatorname{Re}\beta.$$

It is appropriate to call g the gain

We have thus completed the calculation of the spectral density of the surface brightness. Formula (14) reflects two physical mechanisms responsible for the frequency width of the scattering lines, namely the dissipation processes and the influence of the wave detuning, connected with the finite thickness of the nonlinear layer.

We must, however, note the following. In the derivation of the abbreviated equations (3) we neglect the second derivatives of the scalar amplitudes ξ_S and ξ_p^+. To do so it is necessary, in particular, to satisfy the inequalities $|q_{1,2}| \ll \omega_p c^{-1} |\epsilon_p|^{1/2}$. These conditions may be violated[5] near the phonon resonance at sufficiently strong absorption, if $\epsilon'' \gtrsim \epsilon'$. Therefore the theory developed in the present section is, generally speaking, not applicable here. We confine ourselves first to a consideration of the nonresonant region, when $\omega_f - \omega_p$ greatly exceeds the half-width of the spontaneous scattering γ_f. It should also be noted that failure to take the second derivatives into account here, as follows from the results of Sec. 4 (see formula (20)), imposes here a limitation also on the magnitude of the wave detuning Δk:

$$|\Delta k| \ll |\operatorname{Re}\chi^2 / \operatorname{Im}\chi^2| \alpha_p.$$

In the nonresonant region we can neglect the contribution of the imaginary part of the tensor χ_{ijk}, and also the contribution of the tensor γ_{ijkm}. At the same time, the relation between the absorption coefficient $2\alpha_p$ and the gain g remains arbitrary. It is of interest to distinguish and investigate separately two limiting cases: $g \gg 2\alpha_p$ (weak absorption at polariton frequencies) and $g \ll 2\alpha_p$ (strong absorption at polariton frequencies).

2. CASE OF WEAK ABSORPTION AT POLARITON FREQUENCIES

In the region of weak absorption, taking the foregoing into account, we obtain from (6)

$$q_1 = -q_2 = q = \sqrt{D^2 - (\Delta k/2)^2}, \quad D = \chi\sqrt{R}.$$

A detailed analysis of the dependence of D on ω_p and θ_S can be found in[11].

Additional simplification results also from the fact that in (14) we can neglect the second term in the curly brackets. As a result we obtain

$$B_s(\Omega_s, \omega_s) = \frac{\hbar\omega_s^4 \omega_p n_s \chi^2 I_l}{n_p n_l c^5 \cos\theta_p} \left| \frac{\operatorname{sh} qz}{q} \right|^2. \tag{15}$$

This expression as a function of ω_S for fixed Ω_S has a sharp maximum at the point ω_S^0, which is the root of the equation $\Delta k(\omega_S^0) = 0$. All the remaining quantities in (15), which depend on ω_S, can be taken at the point ω_S^0, and Δk can be expanded about this point in an approximation linear in ω_S. After this we obtain

$$B_s(\Omega_s, \omega_s) = B_0(\Omega_s)\Lambda(Dl, \omega_s), \tag{16}$$

where $B_0(\Omega_S)$ is the light flux in a unit solid angle, integrated over the frequencies, in spontaneous Raman scattering (SpRS)[12]. The SRS line shape is determined by the factor

$$\Lambda(Dl, \omega_s) = \frac{1}{\pi} \left| \frac{\operatorname{sh}\sqrt{D^2 l^2 - \eta^2}}{\sqrt{D^2 l^2 - \eta^2}} \right|^2, \quad \eta = \frac{|\zeta| l(\omega_s^0 - \omega_s)}{2}, \tag{17}$$

$$\zeta = \left(\frac{\partial \Delta k}{\partial \omega_s} \right)_{\omega_s^0}.$$

Let us also obtain a formula for the effective SRS line width $\Delta\omega$, defined as the width at which the intensity decreases by a factor e: $\Delta\omega = 0.64\sqrt{Dl}\Delta_0\omega$, where $\Delta_0\omega = 2\pi(|\zeta| l)^{-1}$ is the corresponding effective width of the spontaneous Raman scattering line[13]. We present also formulas for the emission at the polariton frequencies:

$$B_p(\Omega_p, \omega_p) = \frac{\omega_p u_p}{\omega_s u_s} B_s(\Omega_s, \omega_s), \quad \Phi_p = \frac{\omega_p^0}{\omega_s^0}\Phi_s.$$

3. CASE OF STRONG ABSORPTION AT POLARITON FREQUENCIES

In the region of strong absorption, i.e., at $g \ll 2\alpha_p$, we obtain approximately

$$\sigma = \frac{2\alpha_p \chi^2 R}{\alpha_p^2 + (\Delta k)^2}, \quad Q = -\alpha_p, \quad \beta = \mu = -\alpha_p + i\Delta k, \quad \nu = 2\alpha_p.$$

In this case, the predominant term in the general formula (14) for φ_S is the second term in the curly brackets, in which, in turn, the first term with $g\mu^*$ in the denominator, predominates (it is assumed that $\alpha_p l \gg 1$). As a result we get

$$B_s(\Omega_s, \omega_s) = B_s^0(\omega_s)(e^{gl} - 1), \quad B_s^0(\omega_s) = \hbar\omega_s^3 n_s^2 / 8\pi^3 c^2. \tag{18}$$

Here $B_S^0(\omega_S)$ is the spectral density of the flux of electromagnetic radiation of fixed polarization in a unit solid angle from a unit visible surface, due to quantum noise with one photon per mode.

The obtained formula has a simple meaning: the

Stokes-frequency quantum noise, B_s^0 which exists in the unpumped medium, becomes amplified in the pump field to a level $B_s^0 e^{gl}$. Since the observed quantity is the excess of radiation over the quantum-noise level, it is necessary to subtract B_s^0 at the exit from the nonlinear layer.

If $gl \ll 1$, we arrive at the case of spontaneous Raman scattering. Then $B_s = B_s^0 gl$. Unlike the case of weak absorption, the line width of the SRS from polaritons (in analogy with SRS from phonons[14,15]) is smaller by a factor $\sqrt{g_0 l}$ than the spontaneous Raman scattering line width (g_0 is the value of g at the center of the SRS line; it is assumed that $g_0 l \gg 1$). Thus, the dissipation mechanism of SRS line broadening and the mechanism due to the influence of the wave detuning, which in accord with (16) and (17) is responsible for the line broadening in the transparency region, exert different influences on the formation of the SRS line contour.

For the polariton radiation we obtain

$$B_p(\Omega_p, \omega_p) = \frac{g\omega_p u_p}{2\alpha_p \omega_s u_s} B_s(\Omega_s, \omega_s), \quad \Phi_p = \frac{g\omega_p{}^0}{2\alpha_p \omega_s{}^0}\Phi_s. \tag{19}$$

The obtained formulas complete the solution of our problem.

4. SRS FROM POLARITONS UNDER CONDITIONS OF PHONON RESONANCE

Let us discuss further the situation in SRS from polaritons in the vicinity of the phonon resonance. As already noted above, when solving Maxwell's equations in this region it is necessary to take into account[5] the second derivatives of the amplitudes of the Stokes and polariton waves.

We shall pay principal attention to the calculation of the gain. Since this calculation does not require the introduction of sources, we consider a homogeneous system of Maxwell's equations. We seek a solution in the form

$$\mathbf{E}_{s,p} = \mathbf{e}_{s,p} A_{s,p} \exp[i(\mathbf{q}_{s,p}\mathbf{r} - \omega_{s,p}t)] + \text{h.c.},$$

assuming that $A_{s,p}$ does not depend on $\mathbf{r}$.

We separate in $\mathbf{q}_{s,p}$ the parts $\mathbf{k}_{s,p}$ corresponding to the absence of pumping, $\mathbf{q}_{s,p} = \mathbf{k}_{s,p} + \boldsymbol{\kappa}_{s,p}$. Here $\mathbf{k}_{s,p}$ are the wave vectors in the unpumped medium[2)], and the vector $\boldsymbol{\kappa}_{s,p}$, which is parallel to the z axis, takes the influence of pumping into account. The system of equations for the coupled waves $\mathbf{E}_{s,p}$ has solutions of damped type and of the type that grow exponentially in space[5]; we are interested in the latter. For these κ_s can be regarded as a small quantity. Changing over to the homogeneous algebraic system of equations for the amplitudes $A_{s,p}$, equating its determinant to zero, and assuming the absorption at the polariton frequency to be so strong that $2|w^z \kappa_s| \ll \omega_p^2 c^{-2} |\epsilon_p|$ ($\mathbf{w} = \mathbf{k}_l - \mathbf{k}_s$), we obtain an equation linear in κ_s (for details see[16]). Solving this equation, we obtain the gain

$$g = -2\,\mathrm{Im}\,\varkappa_s = \frac{8\pi^2\omega_s I_l}{c^2 n_c n_s \cos\theta_s}\left[4\pi\frac{\mathrm{Re}\,\chi^2 - \tau\,\mathrm{Im}\,\chi^2}{\varepsilon_p''(1+\tau^2)} - \mathrm{Im}\,\gamma\right], \tag{20}$$
$$\tau = [w^2 - \varepsilon_p'(\omega_p/c)^2]/\varepsilon_p''(\omega_p/c)^2.$$

2) In contrast to the preceding sections, in which the symbol $\mathbf{k}_p$ was used for the real part of the wave vector, $\mathbf{k}_p$ will stand below for the total wave vector $\mathbf{k}_p = \mathbf{k}_p' + i\mathbf{k}_p''$ at the polariton frequency.

The obtained formula completely determines the gain $g = g(\omega_p, \theta_s)$ in the form of a function of phenomenological parameters of the nonlinear medium, which in general are known.

Let us consider further the case of an isolated phonon oscillation with frequency $\omega_f - i\gamma_f/2$ and obtain, under certain simplifications the frequency-angle position of the maximum of g. It is convenient to separate first from g the factor g_0, given by

$$g_0 = \frac{16\pi^2\omega_s I_l \omega_f \eta_2 M}{c^2 n_l n_s v_0 \omega_p \hbar\gamma_f \cos\theta_s}, \quad M = \frac{\eta_1}{\eta_2},$$
$$\eta_1 = \Big[\sum_\nu \alpha^{(f\nu)}(\mathbf{e}_\nu, \mathbf{e}_p)\Big]^2, \quad \eta_2 = \sum_\nu [\alpha^{(f\nu)}]^2$$

which has a smooth frequency-angle dependence. Here $\alpha^{(f\nu)} = e^i \alpha_{ij}^{(f\nu)} e_l^j$, $\alpha_{ij}^{(f\nu)}$ is the spontaneous Raman scattering tensor per cell of volume v_0, and the index ν numbers the mutually degenerate oscillations of a given frequency (if such exist). The remainder of g will be denoted by Ψ, so that $g = g_0\Psi$. Using the explicit form of the tensors ϵ_{ij}, χ_{ij}, and γ_{ijkl}[9,12], and introducing the symbol $\varphi = (\omega_t^2 - \omega_p^2)(\nu_j\omega_p)^{-1}$, we get

$$\Psi = \frac{(\tilde\varphi + \tau)^2}{(1+\varphi^2)(1+\tau^2)} + \frac{M^{-1} - 1}{1+\varphi^2}, \quad \tilde\varphi = \varphi + \tilde A_f(1+\varphi^2),$$
$$\tilde A_f = A_j\frac{\gamma_f\omega_p}{\omega_f{}^2}, \quad A_f = \chi_0\left(\frac{2\pi v_0\hbar\omega_f}{b_f}\right)^{1/2}\Big[\sum_\nu \alpha^{(f\nu)}(\mathbf{e}_\nu, \mathbf{e}_p)\Big]^{-1} \tag{21}$$

where χ^0 is the nonresonant part of χ[17] and b_f is the oscillator strength of the transition $0 \to f$. The factor g_0 can be taken at the point of the maximum of the factor Ψ (with the exception of the cases when $\omega_p \sim \gamma_f$).

We confine ourselves henceforth to scattering geometries at which $M = 1$. In addition, assuming that $|A_f| \sim 1$[17] and $\gamma_f/\omega_f \ll 1$, we neglect in the vicinity of the phonon resonance the contribution of the nonresonant processes, putting $\tilde\varphi \cong \varphi$. Using the conditions $\Psi_\tau' = 0$ and $\Psi_\varphi' = 0$, we obtain the absolute extrema of Ψ. Both these conditions are equivalent to the condition $D_1 D_2 = 0$, $D_1 = 1 - \varphi\tau$, $D_2 = \varphi + \tau$. From this we get $\tau_1 = \varphi^{-1}$, $\tau_2 = -\varphi$. The maximum of g is reached on the curve $\tau = \varphi^{-1}$. Then $\Psi = 1$ and g_0 determines the maximum gain.

We introduce further the quantity $\tilde n_p = wc/\omega_p$. Solving the equation $\tau = \varphi^{-1}$ with respect to $\tilde n_p^2$, we get

$$\tilde n_p{}^2 = \varepsilon_\infty + b_f\omega_f{}^2/[\omega_f{}^2 - \omega_p{}^2], \tag{22}$$

where ϵ_∞ is the high-frequency limit of ϵ_p. This means that the frequency-angle position of the maximum of g is described by the dispersion curve of the polaritons that would be produced in the scattering without allowance for the wave detuning or for the attenuation. This curve also describes the position of the conditional maxima of g at fixed θ_s or ω_p.

In concluding this section, we note that D. N. Klyshko obtained, simultaneously and independently of us, an expression for the complex dielectric constant of a pumped medium, leading to a formula for g that coincides with (20) at $M = 1$ (compare[18] and[19]).

5. CONCRETE EXAMPLES. COMPARISON WITH EXPERIMENT

We now apply our general results to the calculation of concrete cases of SRS from polaritons in crystals

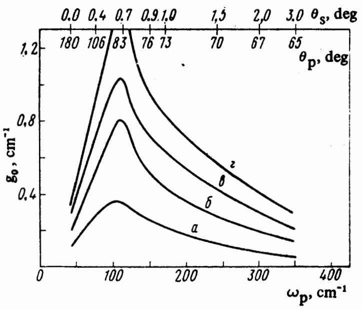

FIG. 2. Frequency-angle dependence of the maximum gain g_0 in a ZnO crystal at different exciting-field intensities. The values of $|A_l|^2$ (in cgs esu) are as follows: a–0.5×10^6, b–1.0×10^6, c–1.5×10^6, d–2.0×10^6.

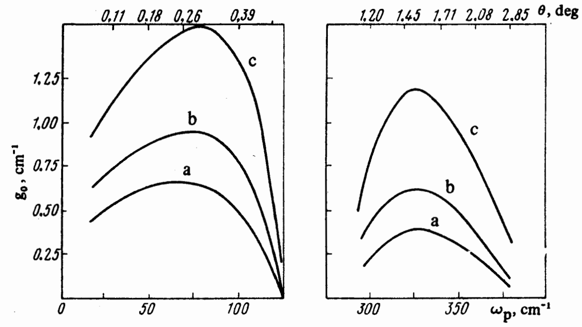

FIG. 3. Frequency-angle dependence of g_0 in an α-quartz crystal. Type o → o + o interaction. The different curves correspond to different values of the parameter ρ: a–3×10^{-6}, b–5×10^{-6}, c–10^{-5}. The ordinate scale is decreased by a factor of 2.

of zinc oxide, quartz, and lithium niobate. It is assumed that the excitation is by means of a ruby laser.

Case of ZnO. The scattering geometry corresponds to that used in[20], namely, $\mathbf{k}_l \parallel \mathbf{x}_0$[3)], $\mathbf{e}_l \parallel \mathbf{y}_0$, $\mathbf{k}_s$ lies in the $\mathbf{x}_0\mathbf{z}_0$ plane, the Stokes wave is extraordinary and the polariton wave ordinary. The values of the required parameters are $A_f = 1.8$[21,22], $\omega_f = 407\ \mathrm{cm}^{-1}$, $\gamma_f = 10.3\ \mathrm{cm}^{-1}$, $\epsilon_\infty = 4.00$, $\epsilon_{st} = \epsilon_p(\omega_p = 0) = 8.15$[12].

The crystal ZnO has a number of specific features, which make its study of interest. Notice must be taken first of the large range of variation of ω_p with changing θ_s: according to the results of[20], the frequency at the maximum of the spontaneous-scattering line changes by almost 10 times in the angle interval $\theta_s = 0-4°$. Further, in the interval $\theta_s = 0-2°$ ($\omega_p \sim 50-100\ \mathrm{cm}^{-1}$) the absorption is quite small. Thus, at $\omega_p = 47\ \mathrm{cm}^{-1}$ ($\theta_s = 0°$) we have $\alpha_p \cong 0.1\ \mathrm{cm}^{-1}$. This favors polariton emission. The scattering lines in the region of weak absorption are very narrow: the effective widths are of the order of several hundredths of a cm^{-1}.

Far from phonon resonance, we have for g_0 the formula

$$g_0 = \sqrt{4D^2 + \alpha_p^2} - \alpha_p. \tag{23}$$

Figure 2 shows the results of calculations of the gain g_0 at the center of the line (i.e., at $\Delta k = 0$). Unfortunately, in view of the small values of g_0, the use of ZnO is promising only if a resonator is employed.

Case of quartz. All three waves are ordinary. The values of α_p and the dispersion data needed for the calculation of g_0 were taken from[23,24]. Figure 3 shows the $g_0(\omega_p)$ plot calculated by us. There exist three sufficiently stable maxima of g_0 at ω_p equal to approximately 70, 170, and 330 cm^{-1}. The calculations were based on formulas that are valid outside the phonon resonances, and therefore the maxima corresponding to the latter are not seen in Fig. 3. It should also be noted that χ depends on ω_p, but this dependence can be approximately neglected in the vicinity of each of the maxima. Of course, the values of χ in the region of different maxima can be different. In particular, one can expect a considerable decrease of χ on going from frequencies $\omega_p < 128\ \mathrm{cm}^{-1}$ into the region $\omega_p > 128\ \mathrm{cm}^{-1}$, since the sign of the contribution made by the oscillation at 128 cm^{-1} to χ is reversed in this case. This contribution is proportional to $(b_f\sigma_f)^{1/2}$ (σ_f is the scattering cross section), which in this case is sufficiently large. Therefore the conditions for the appearance of a maximum near 170 cm^{-1} are apparently less favorable than for the maxima in the vicinity of 70 and 330 cm^{-1}. The vicinity of the maximum at 170 cm^{-1} is not shown in Fig. 3. The parameter employed in the problem was the quantity

$$\rho = 32\pi^5 I_l \chi^2 (cn_l)^{-1}$$

An experimental investigation of SRS in crystalline quartz was carried out by Aref'ev et al.[25]. Six SRS lines were observed in the frequency interval 60–470 cm^{-1}, namely 69, 129, 200, 262, 335, and 467 cm^{-1}. Two of them (128 and 467 cm^{-1}) are regarded by the authors as the Stokes frequencies, and the remainder are interpreted as secondary-scattering lines. Attention is called, however, to the rather good agreement of the 69 and 335 cm^{-1} lines with the polariton maxima on Fig. 3. It is likewise of interest that the lines 200 and 262 cm^{-1} are close to the frequencies of the other fundamental oscillations in accordance with the data of[26]. However, there are not sufficient data at present for a final determination of the origin of the observed lines; additional investigations, primarily experimental ones, are necessary. In any case, one can hope to make use of the polariton maxima shown in Fig. 3 for the excitation of polariton processes.

The case of $LiNbO_3$. Two variants of the geometry of the problem were considered here: a) $\mathbf{e}_s = \mathbf{e}_p = (0, 1, 0)$[4)]–o-waves, the scattered light is detected in the x_0z_0 plane; b) $\mathbf{e}_s = \mathbf{e}_p = (0, 0, 1)$–e-waves, the scattered light is detected in the x_0y_0 plane. In both cases $\mathbf{k}_l \parallel \mathbf{x}_0$ and $\mathbf{e}_l = (0, 0, 1)$. The polariton bands considered[5)] were 152, 236, 265, 322, 363, 431, and 586 cm^{-1}, corresponding to E-type oscillations (geometry a) and also the bands 248 and 628 cm^{-1}, corresponding to A_1 oscillations (geometry b). The values of the parameters needed for the calculations were taken from[27]. The case of $LiNbO_3$, in view of the large nonlinearity of this crystal, is of considerable interest in connection with the problem of the

3)Here and below x_0, y_0, and z_0 denote the principal axes of the tensor ϵ_{ij}.

4)Components along the axes x_0, y_0, and z_0 are indicated.

5)The marking of the polariton bands corresponds to the frequencies of the phonons into which the polaritons go over at large values of θ_s.

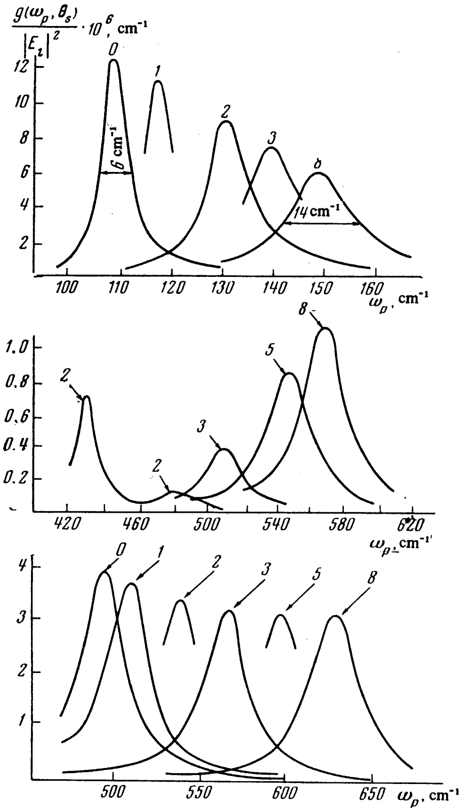

FIG. 4. Spectral form of the gain $g(\omega_p, \theta_s)$ of $LiNbO_3$ crystal for the polariton bands 152 and 586 cm^{-1} (symmetry E) and also 628 cm^{-1} (symmetry A_1). E_l—amplitude of pump field. Numbers at curves—values of θ_s in degrees.

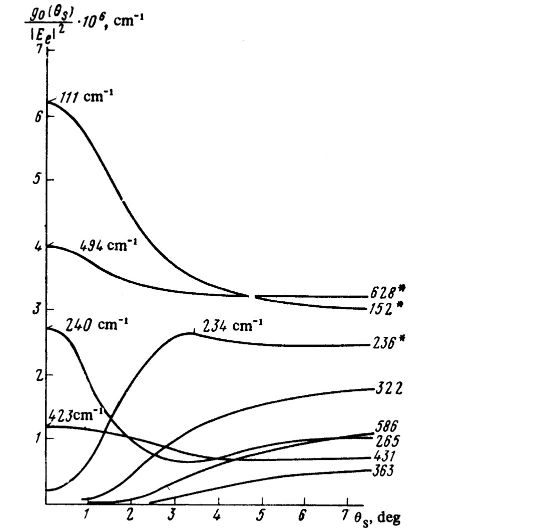

FIG. 5. Angular dependence of the maximum gain g_0 of the $LiNbO_3$ crystal. The scale of g_0 for the bands 152, 236, and 628 cm^{-1}, marked with asterisks, is reduced by a factor of 2.

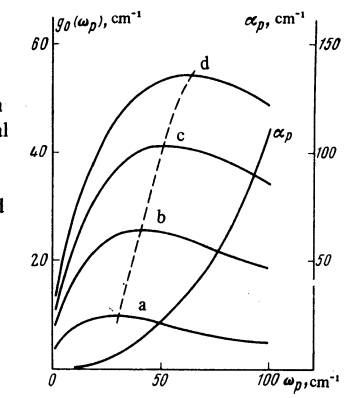

FIG. 6. Frequency dependence of the gain g_0 and of the absorption coefficient α_p of the $LiNbO_3$ crystal in the long-wave part of the IR band. Type e → e + e interaction. Different branches of g_0 correspond to different pump-field intensities. The values of $|A_l|^2$ (in cgs esu) are as follows: a—10^5, b—4×10^5, c—8×10^5, d—1.2×10^6. The dashed line passes through the maxima of g_0.

development of generators of infrared and visible radiation that could be smoothly tuned in a relatively wide frequency interval, and it is precisely in this crystal that a Stokes and a polariton wave were registered simultaneously and reliably[3]. Some of the results of the calculations based on the use of formula (20)[6)] are shown in Figs. 4—6. This is the first time that such calculations have been made.

Figure 4 shows the dependence of $g(\omega_p, \theta_s)$ for polariton bands corresponding to the 162, 236, 265, 586, and 628 cm^{-1} oscillations. We see that the $g(\omega_p, \theta_s)$ spectrum contains a number of lines, the width of which varies from several units to several dozen cm^{-1}. With decreasing θ_s, the effective widths can either decrease or increase. The greatest narrowing for $g(\omega_p, \theta_s)$ (by a factor 2.2, if we compare the cases $\theta_s = 0°$ and $\theta_s = 180°$), takes place for the polariton band 152 cm^{-1}, which extends over the region 111—152 cm^{-1}. Figure 5 shows the dependence of the maximum gains on θ_s. The most favorable for generation, generally speaking, are bands for which the maxima of $g_0(\theta_s)$ lie at $\theta_s = 0$. These are the bands 152, 265, 431, and 628 cm^{-1}. Particularly large values of g_0 are possessed by the polariton bands 152 and 628 cm^{-1}. It is not surprising, therefore, that it is precisely for these that SRS from polaritons in $LiNbO_3$ was registered in[1-3]. No SRS has been observed so far for the other bands shown in Fig. 5.

There are cases when the maximum on the $g_0(\theta_s)$ curve lies at $\theta_s \neq 0°$. These include the 236 cm^{-1} band (the maximum corresponds to $\theta_s = 3.5°$, $\omega_p = 234$ cm^{-1}), and the lowest type-A_1 band, the 248 cm^{-1} band. For the latter, Fig. 6 shows a plot of $g_0(\omega_p)$ at different pump levels. We see that the position and magnitude of the maximum depend strongly on the pump. When I_l increases by one order of magnitude, the maximum shifts towards increasing θ_s and ω_p by approximately 0.5° and 60 cm^{-1}, respectively. The fact that according to the experimental data[3] the polariton scattering in the 248 cm^{-1} band was observed only at $\theta_s \neq 0°$ agrees with the placement of the maxima on the curves of Fig. 6.

We indicate finally that for all the bands with the exception of the initial section $\theta_s < 1°$ of the 363 cm^{-1} band, the maxima of g_s lie on the $\omega_p(\theta_s)$ curve calculated without allowance for the damping and the detuning, with accuracy 1 cm^{-1} (the interval used in the numerical calculations). The deviations for the 363 cm^{-1} band are small, ~2—4 cm^{-1}.

Thus, the scanty experimental data presently available on SRS from polaritons agree qualitatively with

[6)] In view of the large absorption in $LiNbO_3$ (at room temperature), the condition $g \ll 2\alpha_p$ is valid in practically the entire investigated frequency region. The only exception is the region of the lower polariton branch at $\omega_p < 100$ cm^{-1}. The calculation of g_0 for this region, the results of which are shown in Fig. 6, was based on formula (23).

the predictions of the theory. The results obtained here may be useful when choosing the optimal conditions for realizing the SRS effect from polaritons.

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