PARAMETRIC LUMINESCENCE IN CRYSTALS INVOLVING POLARITON EXCITATION

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Parametric luminescence of light[7,8] in crystals involving polariton excitation, which can also be interpreted as Raman scattering by polaritons, is considered. The dependence of the intensity I_S of such scattering on the scattering angle θ and other parameters of the problem is investigated. It is shown for GaP and ZnO crystals that the polariton nature of the excitation may result in the appearance of a strong dependence of I_S on θ small values of θ. Thus in ZnO the scattering intensity changes by approximately 10 times in the region between $\theta \sim 0-4°$. Some characteristic features of Raman light scattering by longitudinal optical phonons are also investigated for small values of θ. The theory is in qualitative agreement with the experimental data.[3,4]

It is well known that the electromagnetic waves interact strongly with phonons that are active in the absorption spectra of crystals, provided that both have nearly equal frequencies and wave vectors.[1] The results of the interaction are excitations of mixed electromagnetic-mechanical nature,[2] called polaritons. They can be regarded as a particular case of photons in a medium (quanta of the macrofield energy).

The polariton character of the excitations becomes manifest in the Raman scattering (RS) spectra of light in piezoelectric crystals, in the form, for example, of the recently discovered kinematic effect of the decrease of the Stokes frequency shift of the scattered wave with scattering angle θ (in the case of oscillations that are active simultaneously in the absorption spectra and in RS of light[3-5]). This decrease occurs in the region of small scattering angles ($\theta \sim 0-6°$), when the quasimomentum $\hbar\mathbf{k}$ transferred to the crystal by the exciting radiation in the visible region becomes commensurate with the quasimomentum of the electromagnetic wave in the vicinity of the resonant frequency of the lattice vibrations. The observable change of displacement is quite appreciable in this case: a change of the Stokes shift by a factor of 2.5 was observed in [4].

The question of the angular shift of the scattering frequency was discussed in detail in [3-6]. The present paper is devoted to a theoretical investigation of the angular dependence of the intensity of RS by polaritons. Since the polariton is essentially a photon in a medium, we can treat the RS as a particular case of parametric luminescence (PL),[7] when the latter is accompanied by polariton excitation,[8-10] or else as spontaneous decay of a photon ω_L of exciting radiation, say from a laser, into two photons: a Stokes photon ω_S and a polariton ω_p.

We note that besides the study of the RS of light, an investigation of processes of this kind is very important also for the description of stimulated Raman scattering (SRS) by polaritons, since the probability of spontaneous scattering at a given angle determines directly the behavior of the SRS. This question is of particular interest in connection with the possibility of producing a sufficiently intense[1)] tunable coherent source of infrared or submillimeter radiation, since SRS by polaritons is accompanied by the emission of the appropriate frequencies.

1. ANALYSIS METHOD

The already mentioned spontaneous decay is due to the interaction

$$W = -\frac{1}{c}\int \mathbf{J}(\mathbf{r})\mathbf{A}(\mathbf{r})\,dv$$

between the Coulomb system and the transverse macrofield in the medium, which is quantized in accordance with [11,12] (div A = 0), and is described in third order perturbation theory. The light flux $B(\Omega)$ at the frequency ω_S, scattered in a unit solid angle near the direction $\Omega = \mathbf{k}_S/k_S$ can be represented in the form

$$B(\Omega, \sigma_p) = \frac{2\pi\hbar V I_L}{n_L c^5}\,\frac{\omega_s^4 \omega_p n_s \gamma_s \gamma_p \gamma_L v_s v_p |\chi(\sigma_p)|^2}{n_p[v_s\gamma_s - v_p\gamma_p\cos\psi]}, \qquad (1)$$

$$\chi(\sigma_p) = e_s^i(\mathbf{k}_s)e_p^j(\mathbf{k}_p)e_L^k(\mathbf{k}_L)\varkappa_{ijk}(\mathbf{k}_s, \omega_s; \mathbf{k}_p, \omega_p). \qquad (2)$$

Here n_i, e_i, and γ_i are respectively the refractive index, the polarization unit vector, and the cosine of the angle between the group and phase velocities of the i-th wave; I_L is the intensity of the exciting (laser) radiation, ψ is the angle between the wave vectors $\mathbf{k}_S$ and $\mathbf{k}_p$, κ_{ijk} is the tensor of nonlinear susceptibility of the medium, σ_p is the polarization of the produced polariton, and V is the volume of the scattering section of the body. In the derivation of (1) we took into account the fact that the energy and momentum are conserved in the elementary decay act:

$$\omega_L = \omega_s + \omega_p, \quad \mathbf{k}_L = \mathbf{k}_s + \mathbf{k}_p. \qquad (3)$$

Formula (1) is a generalization of the result of Klysh-

1)The ratio of the powers of the SRS and the polariton radiation when ω_p is quite removed from the frequency of the optical phonon ω_f (when the decay becomes small) has an order of magnitude $\sim \omega_p/\omega_s \sim 10^{-3}$.

ko[8, 9] to include the arbitrary case of anisotropic crystals; this is our starting formula.

As ω_p approaches ω_f—the frequency of the infrared-active photon f, the PL goes over into RS on polaritons. Accordingly, the investigation consists of the application of the general formula (1) to this case. We use here explicit expressions for κ_{ijk},[13–15] and assuming that ω_L and ω_s lie in the visible part of the spectrum, we retain in these expressions only the terms containing frequency denominators with ω_p. This makes it possible to connect κ_{ijk} with the RS tensor $\alpha_{ij}^{(f)}$:[16, 17]

$$\alpha_{ij}^{(f)}(\omega_L, \omega_s) = \frac{V^2}{\hbar\omega_L\omega_s\sqrt{N}} \sum_l \left[\frac{J_{0l}^j J_{lf}^i}{\omega_{l0}-\omega_L} + \frac{J_{0l}^i J_{lf}^j}{\omega_{l0}+\omega_s}\right], \tag{4}$$

which describes the "ordinary" RS, i.e., RS without allowance for the polariton effects (N—number of unit cells in the volume V, J_{nm}—matrix elements of the current-density operator, taken at the origin between the state satisfying the quasimomentum conservation law). An analysis of this connection makes it possible to separate explicitly the influence of the polariton effect.

2. RAMAN SCATTERING BY POLARITONS

The explicit connection between κ and α reduces to the following:

$$\varkappa_{ikj}(\omega_s, \omega_p) = \frac{\sqrt{N}}{i\hbar\omega_p} \sum_f \left[\frac{J_{f0}^j}{\omega_f-\omega_p}\alpha_{ki}^{(f)}(\omega_L, \omega_s) + \frac{J_{0f}^j}{\omega_f+\omega_p}\alpha_{ik}^{(f)}(\omega_s, \omega_L)\right]. \tag{5}$$

We shall henceforth confine ourselves to an isolated phonon line ω_f, in the vicinity of which ω_p is located, so that only the mutually degenerate states take part in the summation over f. To number these states, we introduce an additional index ν. In the transparency region, the tensor $\alpha_{ij}^{(f\nu)}$ can be regarded as symmetrical and the difference between the frequencies ω_L and ω_s can be neglected.[16, 17] Then, taking into account the relation $\mathbf{J}_{l0} = -\mathbf{J}_{0l} = -i\omega_{l0}\mathbf{P}_{l0}$ we can represent (5) in the form

$$\varkappa_{ijk} \cong \varkappa_{ijk}^{(f)} = \frac{2\sqrt{N}}{\hbar} \sum_{f\nu} \frac{\omega_f P_{f\nu,0}^j \alpha_{ik}^{(f\nu)}(\omega_L)}{\omega_f^2-\omega_p^2}. \tag{6}$$

We introduce further into consideration the energy flux $B_0^f(\Omega)$ in a unit solid angle, emitted in ordinary RS, when the crystal goes over during the scattering process to a final state f. The expression for $B_0^f(\Omega)$ can be readily derived by a semi-phenomenological method in second order perturbation theory (see also [11, 16]), namely

$$B_0^f(\Omega) = N\left(\frac{\omega_s}{c}\right)^4 \gamma_L \frac{n_s}{n_L} \sum_\nu |e_s^i(\mathbf{k}_s)\alpha_{ij}^{(f\nu)}(\omega_L)e_L^j(\mathbf{k}_L)|^2 I_L. \tag{7}$$

Substituting (6) in (1) and taking into account the character of the polarization of the Coulomb exciton,[18–20] and also making allowance for the relation[2)]

$$n^2 = \frac{k^2c^2}{\omega^2} = (e_i\varepsilon_{ij}^{\perp}e_j), \tag{8}$$

where[18]

$$\varepsilon_{ij}^{\perp}(\omega) = \varepsilon_{0ij}^{\perp} + 8\pi V \sum_f P_{f0}^i P_{f0}^j \omega_f \hbar^{-1}(\omega_f^2-\omega^2)^{-1},$$

we obtain in the case of anisotropy crystals

$$B^f(\Omega, \sigma_p) = B_0^f(\Omega)\Pi_f(\Omega, \sigma_p), \tag{9}$$

$$\Pi_f(\Omega, \sigma_p) = \frac{\omega_f\,\partial n_p/\partial\omega_p}{n_p + \omega_p\,\partial n_p/\partial\omega_p - c(v_s\gamma_s)^{-1}\cos\psi}. \tag{10}$$

In the case of cubic crystals it is necessary to sum (9) additionally with respect to σ_p, in view of the fact that both polarizations σ are on par. The result then coincides with (9) and (10), if σ_p is omitted from the latter equations.

Thus, owing to allowance for the polariton effect, the quantities B^f and B_0^f differ from each other by a factor Π_f, which we shall call the polariton factor. At sufficiently large scattering angles θ (roughly speaking, when $\sin(\theta/2) \gg \omega_f/\omega_L$), we have $\Pi_f \to 1$, since $\omega_p \to \omega_f$ and consequently $B^f \to B_0^f$.[3)]

As to the quantity $\cos\psi$ in (10), it can also be expressed in terms of the macroscopic parameters. In the numerical calculations it is more convenient, however, to deal with the following function:

$$Z = n_p\cos\psi = c\omega_p^{-1}(k_L\cos\theta - k_s),$$

where θ is the scattering angle in the crystal, i.e., the angle between $\mathbf{k}_L$ and $\mathbf{k}_s$. Assuming that $\omega_{L,s} \gg \omega_{p,f}$, we get

$$Z = n_p\cos\psi = n_L + \omega_L\frac{\partial n_s}{\partial\omega_s} - \frac{\omega_L}{\omega_s}\left(\Delta n_{Ls} - 2n_L\sin^2\frac{\theta}{2}\right), \tag{11}$$

$\Delta n_{Ls} = n_L(\omega_L) - n_s(\omega_L)$. The quantity Δn_{Ls} can be different from zero if the exciting radiation and the radiation scattered in the anisotropic crystal have different polarizations.

Formulas (9)–(11) are the final formulas and can be used directly for the investigation of various particular cases.

Let us examine in greater detail the case of cubic crystals. Using (3), and also the fact that now $\Delta n_{Ls} = 0$, as well as the well-satisfied inequality $v_s \gg c\omega_p \times (2n_L\omega_L)^{-1}$, we can rewrite Π_f in the form

$$\Pi_f = \frac{\omega_f\,\partial\varepsilon_p/\partial\omega_p}{\omega_p\partial\varepsilon_p/\partial\omega_p + 8(\omega_L\omega_p^{-1}n_L)^2\sin^2(\theta/2)}, \quad \varepsilon_p = n_p^2. \tag{12}$$

In the low-frequency region we can put for the dielectric constant[1]

$$\varepsilon_p \equiv \varepsilon_p(\omega_p) = \varepsilon_\infty + \omega_f^2(\varepsilon_{st} - \varepsilon_\infty)(\omega_f^2-\omega_p^2)^{-1}, \tag{13}$$

where ϵ_{st} and ϵ_∞ are the static and high-frequency (relative to ω_f) dielectric constants.

As a result, the problem of finding $\Pi_f(\theta)$ has been reduced practically to the determination of the $\omega_p(\theta)$ dependence. The latter can be determined from the energy and momentum conservation laws (3).[3, 4] We note also that in the case of forward scattering ($\theta = 0$) we have $\Pi_f = \omega_f/\omega_p$ and that for the lower polariton branch $\Pi_f > 1$.

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2)The fact that the poles of n, as can be seen from (8) correspond to the energies of the Coulomb excitons ω_f [18], signifies that the frequencies of the lines observed in the phonon spectra coincide with the resonant frequencies of the refractive index.

3)We confine ourselves to a discussion of scattering by the lower polariton branch.

3. BEHAVIOR OF $B_0^f(\Omega)$ AND SCATTERING BY LONGITUDINAL PHONONS IN THE REGION OF SMALL θ

As seen from (9), the scattering intensity is determined by the products of two quantities, namely, B_0^f and the polariton factor $\Pi_f(\Omega, \sigma_p)$ (10). Since the properties of the latter have already been discussed above, we shall investigate now the behavior of B_0^f in the region of small scattering angles. This quantity determines completely the intensity in those cases when the polariton effects are completely missing and the factor Π_f in (9) should be simply omitted in the entire region of angles θ (for example, in the scattering by longitudinal optical phonons).

It is important to note, however, that in considering scattering at small angles it is necessary to take exact account of the geometrical relations between the vectors of the waves that take part in the scattering process. Thus, in the region of large θ we can neglect the difference between the lengths of the vectors $\mathbf{k}_L$ and $\mathbf{k}_S$ (since $k_L - k_S \ll q$, $q = |\mathbf{k}_L - \mathbf{k}_S|$). Assuming, in particular, that $\psi = (\pi - \theta)/2$. This can no longer be done at small values of θ, for here $q \sim k_L - k_S$. This circumstance was not reflected in earlier works on RS in crystals.[21]

Let us examine, for example, scattering by a transverse optical phonon in a cubic crystal, using the setup geometry corresponding to Fig. 1, in a coordinate system whose axes coincides with the twofold axes of the crystal C_2 if $\mathbf{k}_L \parallel OX$ and $\mathbf{e}_L \parallel OZ$. Scattering is observed and is polarized in the XY plane ($\mathbf{e}_L \perp \mathbf{e}_S$) plane. In this case the intensity of scattering at a distance R, without allowance for the polariton effects, is

$$I_s^0(\theta) = B_0^f(\theta)/R^2 = M|\alpha_{yz}^{(fx)}|^2 \sin^2\theta \frac{[\cos\theta - \sqrt{b^2 + 4\sin^4(\theta/2)}]^2}{b^2 + 4\sin^2(\theta/2)}$$
$$M = N\left(\frac{\omega_s}{c}\right)^4 \frac{n_s}{n_L}\frac{I_L}{R^2}, \qquad b = b(\theta) = \frac{\omega_p}{\omega_s}\frac{c}{n_s v_s} \ll 1. \tag{14}$$

At large θ ($\theta \gg b$), the expression for $I_S(\theta)$ goes over into the approximate formula obtained in [7, 21]:

$$\tilde{I}_s(\theta) = M|\alpha_{yz}^{(fx)}|^2 \cos^2(3\theta/2),$$

whereas at small θ appreciable differences occur.

Retaining the same geometry of experiment as before, we obtain the intensity of scattering by a longitudinal optical phonon:

$$I_s(\theta) = m|\varkappa_{xyz}|^2 \frac{[\cos\theta\sqrt{a^2 + 4\sin^4(\theta/2)} + \sin^2\theta]^2}{a^2 + 4\sin^2(\theta/2)},$$
$$a = c\omega_f'' (n_s v_s \omega_s)^{-1} \ll 1. \tag{15}$$

At large scattering angles ($\theta \gg a$), (15) goes over into the well known result[6, 15]

$$\tilde{I}_s(\theta) = m|\varkappa_{xyz}|^2 \sin^2(3\theta/2), \tag{15'}$$

In the region of small angles ($\theta \lesssim a$), however, the functions I_S and $\tilde{I}_S$ behave in essentially different manners.

Finally, let us assume that $\mathbf{k}_L$ is parallel to the three-dimensional diagonal of the crystal-lattice cube, and the light scattered by the transverse phonon is observed in a plane passing through $\mathbf{k}_L$ at one of the edges of the cube, with $\mathbf{l}_S$ lying in the same plane and $\mathbf{l}_L$ perpendicular to it. In this case we have

$$I_s^0(\theta) = m|\varkappa_{xyz}|^2 \sin^2(\theta - \alpha), \quad \cos\alpha = 1/\sqrt{3},$$

i.e., in the dependence of I_S and $\Delta\theta$ can be neglected in the small angle interval I_S θ.

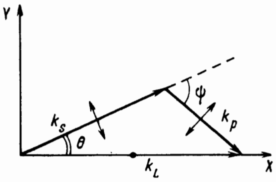

FIG. 1

4. DISCUSSION OF RESULTS AND COMPARISON WITH EXPERIMENT

Experimental investigations of polariton effects were carried out on crystals GaP,[3] ZnO,[4] and α-quartz,[5] the RS being excited with gas lasers. Principal attention was paid there to the kinematic aspect of the problem, namely to the determination of the dependence of the observed Stokes shift $\omega_\beta = \omega_L - \omega_S$ of the scattered frequency ω_S on the scattering angle θ in the region of small θ. As regards the intensity $I_S(\theta)$, proper attention has not been paid heretofore to its investigation at small angles θ, probably because of the difficulties connected with registration of the scattered radiation, so that we can speak more readily only of preliminary results. This makes the problem of comparing the theory with experiment more difficult. Nonetheless, it is possible to advance in this connection a number of considerations, which may perhaps be useful also from the point of view of future experiments.

We consider first the cubic crystal GaP (class T_d). In [3] they investigated scattering in this crystal near the directions [100] and [111]. It was observed in the case of the [100] direction that scattering by transverse phonons is missing in the angle region $\theta > 1°$, and for longitudinal phonons in the region $\theta < 1°$. This agrees with formulas (14) and (15). Borrowing from [22, 23] the values of the required parameters, we obtain[4)] $a = 1.8°$ and $b = 1.4°$, so that the region of the transition values of θ, in general, corresponds to that considered in [3]. Figure 2 shows (in relative units) a plot of $I_S(\theta)$ obtained by us theoretically, by means of formula (15), for the scattering in GaP by a longitudinal phonon $\omega_f = 404\ \text{cm}^{-1}$. The angles θ correspond to Stokes radiation in the crystal prior to its emergences to the outside. The same figure shows for comparison (dashed line) curve plotted in accordance with formula (15')[6, 21]). We see that this curve does not hold in the region of small θ.

Inasmuch as in the case of transverse oscillations $I_S^0 \to 0$ near the [100] direction, we have investigated the polariton effects for the [111] direction. Here $I_S^0(\theta) \simeq$ const and the change of $I_S(\theta)$ is due entirely to the factor $\Pi_f(\theta)$. To calculate this factor we have used (12) and (13), putting $\epsilon_{st} = 10.18$, $\epsilon_\infty = 8.46$, $n_L = 3.32$, λ_L

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4)In view of the fact that $\omega_{L,\,S}$ are close to the absorption poles we cannot identify v_S with c/n_S, since these quantities differ here by a factor of several times. The value of v_S was obtained by us by using (with interpolation) the results of dispersion measurement [22].

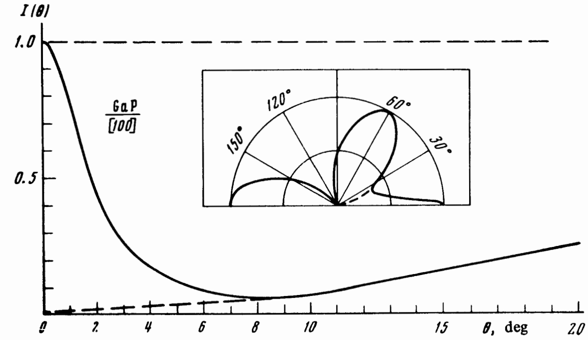

FIG. 2. Angular dependence of the intensity of scattering by a longitudinal phonon (ω''_f = 404 cm^{-1}) in a GaP crystal.

= 6328 Å, and taking the $\omega_p(\theta)$ dependence from [3]. The results are shown in Fig. 3, from which it is seen that $I_s(\theta)$ grows quite appreciably (~25%) in the region of small θ. For the sake of comparison, we show also the $\omega_p(\theta)$ curve.

From among the experimentally investigated anisotropic crystals ZnO and α-quartz,[4,5] it is preferable to use ZnO (class C_{6v}) for a comparison of theory with experiment, since the signal-to-noise ratio is somewhat higher on the spectrograms of [4]. At the experimental geometry used in [4], I_s^0 is practically independent of θ in the region $\theta \sim 0-6°$, so that it is sufficient to consider the factor $\Pi_f(\theta)$.

In the case when the laser radiation propagates in a crystal as an ordinary (o) wave, and the Stokes radiation is detected as an extraordinary (e) wave, the polariton appearing in the RS spectra is ordinary (o). Therefore, to find n_p, we can use (13), in which ϵ_{st} and ϵ_∞ are replaced by $\epsilon_\perp^{st}$ and $\epsilon_\perp^\infty$. Substituting (11) and (13) in (10) and neglecting the difference between γ_s and unity at small θ, we obtain

$$\Pi_f(\theta) = \frac{\omega_p(\varepsilon_\perp^{\text{ст}} - \varepsilon_\perp^\infty)}{\omega_f[1-(\omega_p/\omega_f)^2]^2[\varepsilon_{\perp p} + {}^1/_2\,\omega_p\,\partial\varepsilon_{\perp p}/\partial\omega_p - A]}, \qquad A = \frac{c}{v_s^e} Z(\theta). \tag{16}$$

This formula can be calculated numerically, as was

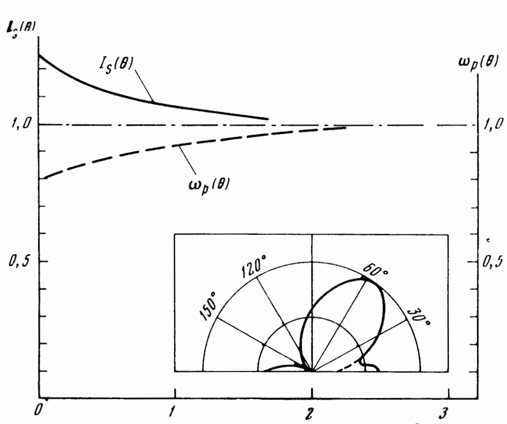

FIG. 3. Angular dependence of the intensity of scattering by a transverse polariton in a GaP crystal in the region of small θ. The frequency unit is ω_f = 367 cm^{-1}. In the right-hand corner we show schematically the scattering indicatrix.

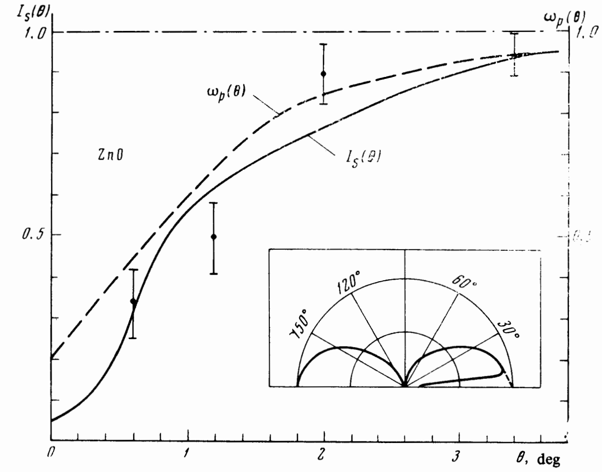

FIG. 4. $I_s(\theta)$ plot for scattering in ZnO crystal (ω_f = 407 cm^{-1}) in the region of small θ. The scattering indicatrix is shown schematically in the right-hand corner. Points – values of $I_s(\theta)$ in accordance with the spectrograms from [4].

done by us for the following values of the parameters: λ_L = 4880 Å, ω_f = 407 cm^{-1}, $n^e(\omega_L)$ = 2.0815, $n^0(\omega_L)$ = 2.0644, v_s^e = 0.3914 c. The results are shown in Fig. 4 in the form of a solid curve for $I_s(\theta)$ together with a dashed curve for $\omega_p(\theta)$. We see that in a region $\Delta\theta$ spanning ~4° there is a sharp decrease of $I_s(\theta)$ (by a factor of almost 10). From the spectrograms contained in [4], we have determined the experimental values of $I_s(\theta)$, assuming them to be proportional to the height of the maxima. We took into account here the fact that the quantity $I_s(\theta)$ investigated by us is in fact the intensity of the given RS line, observed at a fixed angle θ, and integrated over the frequencies within the limits of the broadening $\delta_f\omega$ due to the decay of the phonon. If $\delta_f(\omega)$ and the spectral width $\delta_s\omega$ of the output slit are small compared with the line width $\Delta\omega$ registered on the spectrogram,[5)] then $I_s(\theta)$ can be approximately regarded as proportional to the heights of the maxima. The values of $I_s(\theta)$ obtained in this manner for θ equal to 0.6, 1.2, 2, and 3.4° are also shown in Fig. 4, and on the whole agree with the theory.

In conclusion we note that the angle interval $\Delta\theta$ in which polariton effects are appreciable could be much larger if RS were excited in the infrared region, for example, by radiation from a CO_2 laser. We note also that the region of applicability of the theory developed in Sec. 2 can be expanded by taking into account, besides the resonant processes also the contribution made to χ by the nonresonant term $\chi_0 = e_s^i e_p^j e_L^k \kappa_{ijk}^0$. In this case (9) acquires an additional factor

$$K_f = \left[1 + A\left(1 - \frac{\omega_p^2}{\omega_f^2}\right)\right]^2, \qquad A = \frac{\chi_0 \hbar\omega_f}{2\sqrt{N}\sum\limits_\nu \alpha^{(f\nu)}(\mathbf{P}_{f\nu}\mathbf{e}_p)}$$

which may turn out to be appreciable at sufficiently large χ_0 (for example, in semiconductors with a narrow

[5)] As a result of the polariton effects in the region of small θ, the value of $\Delta\omega$ is determined mainly by the aperture of the input slit and increases with decreasing θ. In the ZnO crystal [4] we have $\Delta\omega \sim$ 100 – 30 cm^{-1} in the angle region $\theta \sim 0.6 - 4°$.

forbidden band) with increasing distance away from resonance.

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