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Scattering of Light by Polaritons in a Biaxial Crystal ($\alpha-HIO_3$)

D. N. Klyshko, V. F. Kutsov, A. N. Penin, and B. F. Polkovnikov
Moscow State University

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The scattering of light by polaritons in a biaxial crystal has apparently been observed for the first time. Measurement of the scattering angle as a function of the frequency enables us to determine the dispersion of the two principal values of the dielectric permittivity tensor in the range from 800 to 6500 cm^{-1}. Several "anomalous" resonances are observed in the spectrum; their intensity falls off abruptly with increasing values of the scattering angle θ, and the position of the angular maximum of the intensity is determined by a dispersion law having an anomalous region. A simple formula, expressing the intensity in terms of the macroscopic tensors characterizing the linear, quadratic, and cubic polarizabilities, enables us (in the single-pole approximation for the latter) to explain the anomaly by the existence of a frequency-independent quadratic polarizability, which is due to the electronic nonlinearity of the crystal. This nonlinearity enables us to observe the spectrum of the fluctuations of the infrared macroscopic field, which are related to the dipole-active vibrations, even for a zero deformation potential.

RAMAN scattering of light by polaritons[1] is the most effective method for experimentally investigating the interaction between the vibrations of a crystal lattice and a macroscopic electromagnetic field. Such scattering processes have been observed in a number of cubic and uniaxial piezocrystals (see, for example,[2-6] and the references cited there). The results of an investigation of the biaxial crystal, alpha-iodic acid, are described below.[1)] This substance has recently attracted attention in connection with the possibility of using it in devices to double the frequency of light and in other nonlinear optical devices.[7-9] More than 20 optical vibrations have been classified in the infrared and Raman spectra of iodic acid.[10]

1. THE EXPERIMENTAL RESULTS

From the experimental point of view, it is most convenient to observe the scattering at small angles by using photographic techniques,[3,4,11] in which the slit of the spectrograph is placed in the focal plane of the lens, which is positioned coaxially with the laser beam after the sample (a telescope is used in order to increase the angular aperture). In this connection a frequency-angular spectrum is recorded on the photographic plate—that is, the intensity of the scattered light as a function of (ω_S, ϑ) (here ω_S is the frequency of the observed Stokes radiation, that is, the frequency of the signal, and ϑ is the angle between the signal wave vector $\mathbf{k}_S$ and the pump wave vector $\mathbf{k}_L$ inside the crystal). The intensity of the spectrum is a maximum along the synchronism curves $\omega_S(\vartheta)$ which are uniquely related (outside the regions of resonance absorption) to the dispersion relation $\Omega(\mathbf{q})$ of the polaritons by the synchronism condition:

$$\mathbf{k}_L = \mathbf{k}_s + \mathbf{q}. \tag{1}$$

Examples of such spectrograms obtained by using an argon laser (λ_L = 4880 Å) are shown in Fig. 1. The vectors $\mathbf{k}_L$ and $\mathbf{e}^L$ (the unit polarization vector of the pump field) were arranged in the xy plane of the crystal (see, for example, [7] for the definition of the crystal axes). The angle θ_L between $\mathbf{k}_L$ and the y axis amounted to 38°. The analyzer only transmitted light polarized in the plane ($\mathbf{k}_L$, z). The photograph shown in Fig. 1a was obtained upon also positioning the spectrograph slit in the xy plane; in this connection all three of the vectors in Eq. (1) lie in this plane, and one can keep the "uniaxial" classification of the types of polarization—namely, the division into ordinary (o) and extraordinary (e) waves. The crystal has D_2 symmetry;[7,10] therefore the only nonvanishing components of the quadratic polarizability tensor, which determines the intensity of the scattered light, are the off-diagonal components β_{xyz}. Hence it follows that the scattering shown in Fig. 1a is caused by e-polaritons (associated with vibrations of the type $B_{x,y}$). The weak "tail" in the interval between 800 and 900 cm^{-1} (the white arrow shown in Fig. 1a) is probably due to o-polaritons (vibrations of the type B_z) in spite of this selection rule. Figure 1b shows an example of a spectrogram obtained upon removing the spectrograph slit (the vectors $\mathbf{k}_S$ and $\mathbf{q}$) from the xy plane. Here the "uniaxial" classification loses its meaning, and the two types of polaritons are simultaneously observed over a broad range (in the interval from 2.3 to 3.0 microns, the intersection of the spectrogram by the horizontal line ν = const gives four values of the scattering angle ϑ).

The asymmetry of the synchronism curves in Fig. 1 with respect to the sign of ϑ is caused by the angular dispersion of the index of refraction of the e-polaritons:

$$n_e(\theta) = cq_e(\theta)/\omega = (\varepsilon_x^{-1}\cos^2\theta + \varepsilon_y^{-1}\sin^2\theta)^{-1/2} \tag{2}$$

(θ denotes the angle between $\mathbf{q}$ and the y axis, $\omega \equiv \omega_L - \omega_S$, and $\epsilon_{x,y}$ are the principal values of the dielectric permittivity tensor). This asymmetry makes it feasible to measure the two values $\vartheta_\pm$ at a given frequency, and this in turn allows us, by means of Eq. (1), to determine the two values of $\theta_\pm$ and $q_e(\theta_\pm)$, and then by using the system (2) of two equations we are able to determine the principal values ϵ_x and ϵ_y ($n_e(\omega_L)$ and $n_o(\omega_S)$ were determined according to the data given in [7]). The results of such a procedure are shown in Fig. 2. The errors are primarily due to errors in the reading and calibration of ϑ and are of the order of 1%. However, the errors increase sharply (up to ~10%) in the vicinity

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[1)] The authors thank L. N. Rashkovich for providing the sample.

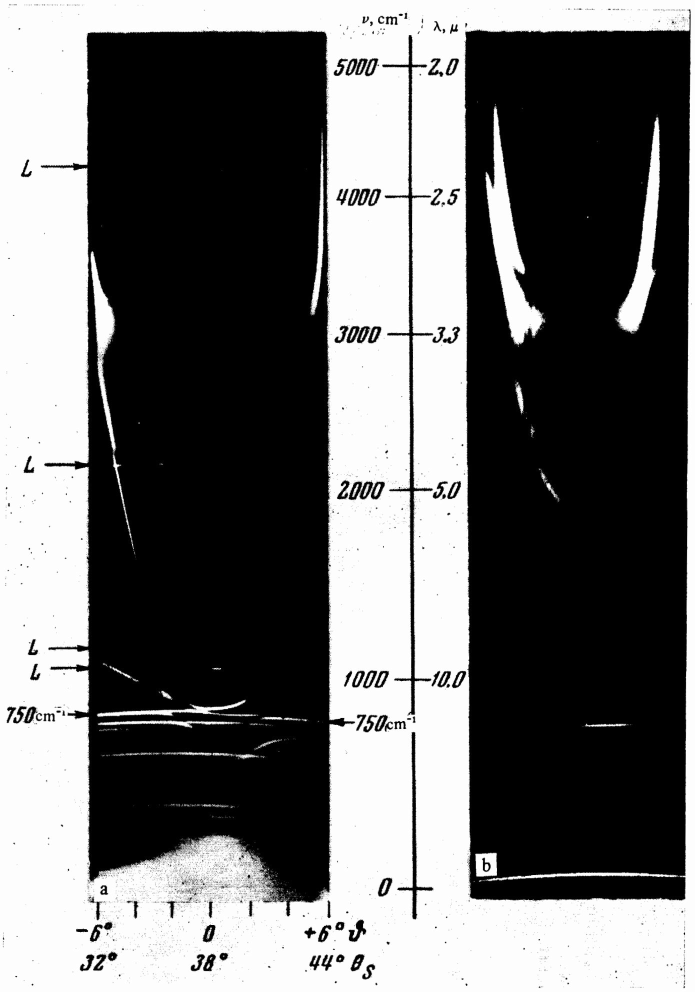

FIG. 1. Spectra of polariton scattering in α-HIO_3. ϑ denotes the scattering angle relative to the wave vector of the pumping field (recalculated to the interior of the crystal). θ_S is the angle relative to the y axis of the crystal. Figure 1a corresponds to the case when the spectrograph slit and the xy plane coincide (the arrows with the label L indicate parasitic lines of the argon laser); Fig. 1b corresponds to the case when the slit is removed from the xy plane by ~10°.

of 1000 cm^{-1} due to the accidental proximity of $\theta_\pm$ to 90° (which gives $q_+ \approx q_-$), and the errors also increase in the interval from 800 to 900 cm^{-1} due to the steepness of the synchronism curves. The values of ϵ_z are determined under the assumption that the branch of the spectrum referred to above is the one associated with o-polaritons.

Let us consider certain characteristic features of Fig. 1a. The asymmetry in the intensities of the right and left branches is connected with the influence of θ on the effective interaction constant (outside the regions of resonant absorption):

$$\beta = \beta_{ijk} e_i{}^s e_j{}^L e_k \neq \beta_{2xy} \cos \bar{\theta}_L \sin \bar{\theta} + \beta_{2zyx} \sin \bar{\theta} \sin \bar{\theta}_L$$
$$\bar{\theta}_L \equiv \theta_L + \rho_L, \quad \bar{\theta} \equiv \theta + \rho$$

(here $\rho = \pi/2 - (\widehat{\mathbf{k} \cdot \mathbf{e}})$ is the angle of anisotropy, which is determined by ϵ_x, ϵ_y, and θ; in the resonance region $\rho(\theta)$ may reach 90°—thus it is probable that the sinuous line at 750 cm^{-1} (see Fig. 1a) is caused by scattering by quasi-longitudinal polaritons). The "dip" in the intensity of the spectrum around 1300 cm^{-1} is caused by, just as in the case of lithium niobate[3] and possibly in the case of ADP,[4] a "linearization" of the crystal as a consequence of the mutual cancellation of the electron and the electron-lattice parts of β. Thus, the above "dip" in the scattering is primarily due to the purely electronic nonlinearity.

An interesting feature of the spectrum is the presence (see Fig. 1a) of three "anomalous" resonances (two weak resonances at 1600 and 2200 cm^{-1} and a strong and a diffuse resonance at 3000 cm^{-1}), which abruptly disappear in the region of large values of $|\vartheta|$. These resonances appear in the synchronism curves $\vartheta(\omega)$ (Fig. 1) and consequently on the dispersion curves $\epsilon(\omega)$ (Fig. 2) in the form of regions with anomalous dispersion. The resonance at 3000 cm^{-1} is obviously related to the OH-bond (the lines observed in [10] in the Raman spectrum at 1160 and 2940 cm^{-1} were attributed to OH-vibrations).

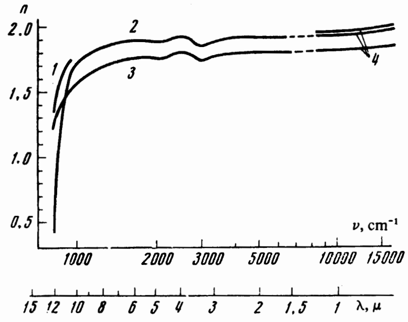

FIG. 2. Dispersion of the dielectric permittivity tensor for α-HIO_3 in the infrared region, measured with the aid of spectrograms similar to the ones shown in Fig. 1a. Curve 1 is for $n = \sqrt{\epsilon_z}$, curve 2 is for $n = \sqrt{\epsilon_y}$, curve 3 is for $n = \sqrt{\epsilon_x}$, and curve 4 refers to the data from [7].

The numerous resonances in the region between 600 and 800 cm^{-1} lead to a rather complicated spectral shape (here there are five B-type vibrations according to [10]), which is aggravated by the angular dispersion (2). In polar coordinates the graph of this dependence is an ellipsoid for $\epsilon_{x,y} > 0$ and a pair of hyperbolas in the frequency intervals in which either ϵ_x or $\epsilon_y < 0$. Here the interpretation of the spectrum is also hindered by the superimposed 180-degree spectrum, which arises due to the reflection of the pumping wave and the signal from the crystal's boundaries. We note several "normal anti-intersections" of the horizontal phonon lines with the parabolic "photon" curves $\omega(\vartheta)$. In contrast to the anomalous intersections, here (see the resonance at 750 cm^{-1}) the "repulsion" of the branches of the spectrum is quite evident (so that $\omega(\vartheta)$ corresponds to the dispersion law without taking absorption into consideration), the intensity does not change noticeably with increasing values of $|\vartheta|$, and a local dip occurs on the photon branches.

Thus, the resonances observed in the polariton spectrum of iodic acid can be divided into two basic types: normal, having a smooth scattering curve, and anomalous, in which the intensity of the coherent scattering appreciably exceeds the incoherent, isotropic part.[2] On the other hand, it is precisely the anomalous case which seems to be natural, since ϵ' is finite as a consequence of absorption and the condition for synchronism

$$c|k_L - k_s|/\omega \equiv n = \sqrt{\varepsilon'(\omega)} \qquad (1a)$$

cannot be satisfied for large values of ϑ. From the point of view of elementary excitations, the absorption only slightly vertically smears the dispersion relation $\Omega(q)$, without restricting the value of q, and the synchronism condition must have the form

$$n = cq(\omega)/\omega \equiv \sqrt{\varepsilon(\omega)_{\text{id}}}, \qquad (1b)$$

where $q(\Omega)$ denotes the function whose inverse is $\Omega(q)$ (here we neglect the anisotropy). One can easily resolve the contradiction if we consider the (ω, q)-spectrum of the equilibrium fluctuations of the transverse macroscopic field, which is proportional to the function

$$f(\omega, n)_{\text{equil}} = \operatorname{Im}[n^2 - \varepsilon(\omega)]^{-1}. \qquad (3)$$

The maximum of f with respect to n^2 is given by condition (1a), and with respect to ω its maximum is given by condition (1b) (one can verify the latter statement by assuming, for example, that $\epsilon = \epsilon_\infty + (\Delta\epsilon/(\omega - \omega_0 - i\Gamma))$ and $\epsilon_{id} = \epsilon_\infty + (\Delta\epsilon/(\omega - \omega_0))$. In the absence of absorption, f has the shape of a delta function and both conditions agree. Thus, the validity of conditions (1a) or (1b) depends on the experimental procedure. Unfortunately, these simple considerations quantitatively describe only the spectrum of the anomalous resonance ($f_{\text{equil}} \to 0$ for $n \to \infty$).

2. A MACROSCOPIC DESCRIPTION OF THE SCATTERING BY POLARITONS

Resonant polariton scattering has been theoretically investigated repeatedly in both the stimulated[12,14–16] and in the spontaneous[17–19] versions. Here we are interested in qualitative conclusions permitting us to classify the fundamental observable effects and the corresponding parameters under typical experimental conditions. It follows from the phenomenological analysis given below that the difference between normal and anomalous resonances is described by the parameter $b = \Delta\beta/\beta_\infty$, which is the ratio of the amplitude of the resonant part of the quadratic polarizability to the constant part. The mechanical vibrations are not active in Raman scattering when b = 0, and in this case we see only the fluctuations of the macroscopic field, which are transformed into the Stokes region owing to β_∞.[20]

Let the polarization of the medium, which appears in the presence of the macroscopic field **E**, be given by $(\alpha E + \beta E^2 + \gamma E^3)/4\pi$, where $\alpha + 1 = \epsilon$ is the linear dielectric constant and β and γ are the quadratic and cubic polarizabilities multiplied by 4π. It is not difficult to show that the dielectric constant of the medium for the Stokes wave is increased by the following amount under the influence of a monochromatic pumping wave $e^L E_L \exp[i(\mathbf{k}_L \cdot \mathbf{r} - \omega_L t)]$ + c.c.:

$$\widehat{\Delta\varepsilon}(\omega_s, k_s) = \left(\hat{\gamma} + \frac{\hat{\beta^2}}{n^2\cos^2\rho - \varepsilon^*}\right)|E_L|^2 \equiv \hat{\tilde{\gamma}}|E_L|^2,$$

$$\varepsilon \equiv \varepsilon(\omega, k_L - k_s)_{ij} e_i e_j, \quad (\beta^2)_{mn} \equiv \beta(\omega_L - \omega)_{mij} e_i^L e_j \beta(\omega_L - \omega_s)^*_{kln} e_k e_l^L,$$

$$\hat{\gamma}_{mn} = \hat{\gamma}(\omega_L - \omega_L + \omega_s)_{mijn} e_i^L e_j^L. \qquad (4)$$

This expression is valid for sufficiently strong absorption at the frequency ω—such that the mean free path of a polariton is much smaller than a) the dimensions of the sample and b) the characteristic length associated with parametric amplification. Condition (a) eliminates the case of parametric scattering from consideration (see, for example, [20]), and condition (b) is practically always satisfied. It is also assumed that all polarization unit vectors are real, and the medium is assumed to be transparent at the signal and pump frequencies.

The relation between the spectrum of the scattered light in the far zone and $\Delta\epsilon$ differs from the relation between the spectrum of thermal radiation and $\epsilon(\omega_S)$ only by replacing the number of photons $N(\omega_S)$ in the Stokes mode by $N(-\omega) = -[N(\omega) + 1]$. Thus, in the case of spontaneous scattering the spectral intensity of the light (erg/sr) emitted by a volume V of the medium is given by

$$\frac{d^2P(\omega_s, \mathbf{k}_s/k_s)}{d\omega\, d\Omega} = \hbar\omega_s[N(\omega) + 1]\frac{n_s V}{\cos^2\rho_s \lambda_s^3}(-\Delta\varepsilon''), \qquad (5)$$

where $\Delta\epsilon''$ is the imaginary part of the convolution of $\widehat{\Delta\epsilon}$ with the unit vectors of the observed mode and $\lambda_S = 2\pi n_S/k_S$. The coefficient of gain can also be expressed in terms of $\tilde{\epsilon}$—the contraction of $\hat{\epsilon} + \widehat{\Delta\epsilon}$ with the eigenvectors—in the usual way

$$g_s = 2 \operatorname{Im} \tilde{\varepsilon}^{1/2}\omega_s / c\cos\rho_s \qquad (6)$$

(in the case of o-waves Eq. (6) agrees with the result obtained by Strizhevskiĭ et al.[16]). Formulas (4) and (5) (after introducing the correction due to Fresnel reflections on the boundaries of the sample) describe the re-

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[2] Claus[5,6] has observed (by a photoelectric method) an analogous phenomenon in the spectrum of $K_3Cu(CN)_4$. The weakness of the Raman branches was first explained by selection rules, [5] and later by the two-photon nature of the process[6] (in connection with this, the effect was called a "polariton Fermi resonance"; also see [12,13]).

lation between the observed spectrum and the macroscopic parameters α, β, and γ.

Outside the regions of resonant absorption, one does not have to take γ into account in Eq. (4) and one can assume β to be a real, slowly varying function of the frequency so that the spectrum has a sharp maximum, which is determined by the condition that the absolute value of the denominator in (4) will be a minimum. This condition is equivalent to (1) (thus, one can easily verify that for the e-wave $\sqrt{\epsilon}/\cos\rho$ coincides with (2)). One can express the frequency width of the maximum[20] in terms of ϵ'' and the group velocities u_S of the signal and u of the polariton (in the infrared band $u \ll u_S$ and it is essentially equal to the reciprocal of the polariton's lifetime).

In the resonant region it is necessary to take γ and the dispersion of β into account. For simplicity we shall ignore the tensor nature of the quantities. In the vicinity of an isolated "mechanical" resonance[21] and in the absence of spatial dispersion one has

$$\begin{gathered}\varepsilon=\varepsilon_\infty+\Delta\varepsilon/z, \quad \beta=\beta_\infty+\Delta\beta/z^*, \\ \gamma=\gamma_\infty+\Delta\gamma/z^*, \quad z=x-i, \quad x=(\omega_0-\omega)/\Gamma.\end{gathered} \tag{7}$$

Having substituted (7) into (5), one can verify that the resonant Raman term $\Delta\gamma/z^*$ is cancelled out due to the relation $(\Delta\beta)^2 = \Delta\epsilon\Delta\gamma$ between the residues,[14] a relation which follows from perturbation theory (this relation also holds for the corresponding components of the tensors). However, if this cancellation did not occur, then according to (5) the phonon line $\omega = \omega_0$ would horizontally cut the entire region of small angles, and the polariton branch $\omega(\vartheta)$ would branch off downwards from it. Such "connecting crosspieces" are commonly seen on spectrograms, but they are usually explained by 180 degree scattering or else by near longitudinal resonances.[3)]

Consequently the observable spectrum (normalized to the intensity at the center of the Raman line) is determined by the function

$$\begin{gathered}f(x,y)\equiv\frac{\tilde{\gamma}''}{\Delta\gamma}=\frac{(1+a/by)^2}{1+(x-a/y)^2}=\frac{\varepsilon_\infty}{a}\left(y+\frac{a}{b}\right)^2 f_{\text{equil}}, \\ y\equiv\frac{n^2}{\varepsilon_\infty}-1, \quad a\equiv\frac{\Delta\varepsilon}{\varepsilon_\infty^2}=\frac{\omega_l-\omega_0}{\Gamma}, \quad b\equiv\frac{\Delta\beta}{\beta_\infty},\end{gathered} \tag{8}$$

where ω_l is the frequency of the longitudinal resonance. Here the variable y primarily depends on ϑ, and in a rough approximation is proportional to the increase $\vartheta - \vartheta_0$. The following useful approximation is more accurate: $k \approx \vartheta k_L$, from where it follows that

$$y\approx(ck_L\vartheta)^2/\varepsilon_\infty\omega^2-1,$$

which is valid under the conditions $\omega/u_S k_L \ll |\vartheta| \ll 1$ which are often satisfied. The coordinate lines y = const in the (ϑ, ω) plane are almost vertical: $\omega = ck_L\vartheta[\epsilon_\infty(1 + y)]^{-1/2}$. The line y = 0 coincides with the asymptotic synchronism curve $\omega(\vartheta)$ for $|x| \gg 1$. The parameter a characterizes the polariton effect: The distance between the hyperbolas xy = a is equal to $2\sqrt{2a}$.

Neglecting the weak dependence of y on ω, according to (8) the condition for a frequency maximum has the form

$$y=a/x, \quad n^2=\varepsilon_\infty+\Delta\varepsilon/x\equiv\varepsilon_{\text{id}}, \tag{9}$$

that is, it is determined by the dispersion law without taking the absorption into account.[15-17,19] We have f = 0 on the line y = $-a/b$; for $ab^2 > 1$ this local dip is located on the phonon part of the hyperbola xy = a; for the opposite inequality the dip is located on the photon branch.

The condition for an angular maximum (in the same approximation),

$$y=a/[x+1/(x+b)] \tag{10}$$

coincides with (9) only if b = ∞ (a normal resonance). However, in the case b = 0 (an anomalous resonance) instead of (10) we have

$$y=\frac{x}{1+x^2}, \quad n^2=\varepsilon_\infty+\Delta\varepsilon\frac{x}{1+x^2}=\varepsilon' \tag{11}$$

—this is the usual relation between the frequency and the wavelength far away from sources. Here Raman scattering is not present, and the polariton repeats the equilibrium spectrum (3). Thus, even a completely inactive vibration in the 90-degree spectrum ($\Delta\gamma = \Delta\beta = 0$) can be observed in the longitudinal spectrum due to its contribution to ϵ ($\Delta\epsilon \neq 0$) and because of β_∞—that is, due to the nonlinearity created by the other vibrations or by the electronic resonances (it is precisely the latter case which occurs for the three anomalous resonances shown in Fig. 1). We note that essentially this same effect—the appearance of a weak vibration on the polariton branch of a neighboring strong vibration—was treated in a numerical example by Strizhevskiĭ et al.[12] under the name of a "polariton Fermi resonance" (see footnote [2)]). We note also that the resonance at 3000 cm^{-1} is an intermediate resonance ($b \approx -1$), which follows from its pronounced asymmetry with respect to the sign of y (for certain values of a, b, and x the dependence of the function (9) on y has the form of a step function).

In conclusion let us once again emphasize the advantage of using photographic methods of investigating polaritons—namely, simplicity, easy visualization, and the large amount of information. The application of electron-optical devices to amplify the luminosity should sharply increase the sensitivity of the method (the exposure time in our experiments was on the order of one hour for a 50 milliwatt laser).

In principle the method can also be used to investigate exciton polaritons in piezocrystals with the aid of their excitation by two-photon processes. By making small and obvious changes, the formulas cited above will describe the selective decrease in the brightness of the radiation from a supplementary source of white light upon its passage through a sample which has been excited by a laser. The synchronism curves, similar to the ones shown in Fig. 1, will here appear "in negative image." The first experiment on two-photon excitation of exciton polaritons belonging to the upper branch in CuCl (apparently using photoelectric detectors) was recently described by Fröhlich et al.[22] The observation of the lower branch will also be possible in the case of doubly-refractive crystals. Finally, let us note the possibility of observing "inverted" polariton scattering.

[3)] According to [16] this cancellation does not occur for o-polaritons because of degeneracy. But since the mechanical excitons are not the solutions of the complete linear Hamiltonian, their branch of the dispersion curve cannot appear in the spectrum.

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