

Raman scattering by coherently excited phonon polaritons

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The spectra of the Raman scattering by phonon polaritons excited by the beam from a tunable CO₂ IR laser have been studied experimentally. Scattering spectra in wave-vector space have been found. They have been used to determine the polariton dispersion $k_p(\nu)$ and the dispersion of the polariton absorption, $\alpha_p(\nu, k_p')$, of a lithium niobate crystal. Evidence of a polariton Fermi resonance is found in the frequency interval 930–1090 cm⁻¹. This resonance broadens the spectra, while having essentially no effect on the polariton dispersion law. The possibilities of carrying out spectroscopy based on the Raman scattering of light by coherently excited polaritons and the advantages of such spectroscopy over spontaneous scattering are also discussed.

1. INTRODUCTION

The Raman scattering of light by coherently excited (hot) polaritons results from Stokes or anti-Stokes scattering of a test beam by polaritons excited by external IR light. The process may also be thought of as optical mixing of visible and IR light (the generation of sum or difference frequencies) in a medium which has a quadratic nonlinearity when the IR light falls in a region of strong lattice absorption of the crystal and converts into polaritons.

The first experiments on optical mixing under such conditions were carried out in a GaP crystal.¹ Those experiments provided the first opportunity to measure the dispersion of the nonlinear quadratic susceptibility in the region of a lattice resonance and to relate the cross sections for Raman scattering by longitudinal and transverse optical phonons, on the one hand, to the nonlinear-optics properties of a crystal, on the other. Since then, Raman scattering by hot polaritons has also been observed in RbClO₃ (Ref. 2) and SiO₂ (Ref. 3) crystals.

In this paper we report a comparison of the spectroscopic method based on the Raman scattering of light by fluctuational polaritons (spontaneous Raman scattering) and the spectroscopic method based on the Raman scattering by hot polaritons. We report an experimental study of the spectra of Raman scattering by hot polaritons of the upper dispersion branch of a lithium niobate crystal.

2. RAMAN SCATTERING BY HOT AND FLUCTUATIONAL POLARITONS

Because of the significant polariton dispersion $\omega_p(\mathbf{k})$ at the center of the Brillouin zone (Ref. 4, for example), the intensity of the Raman scattering by polaritons is a function of both the frequency¹⁾ $\omega = \omega_l - \omega_s$ and the wave vector $\mathbf{k} = \mathbf{k}_l - \mathbf{k}_s$: $I = I(\omega, \mathbf{k})$, where $\mathbf{k}_l, \mathbf{k}_s, \omega_l, \omega_s$ are the wave vectors and frequencies of the test light and the scattered light, respectively. As a result, a study of Raman scattering by polaritons presents an opportunity to study the spectra both in ω space, $I = I(\omega, \mathbf{k} = \text{const})$, and in \mathbf{k} space, $I = I(\mathbf{k}, \omega = \text{const})$. In many cases, \mathbf{k} polariton spectroscopy has a significant advantage over ω spectroscopy.²⁾ A study of the shape and a determination of the widths of the lines of these spectra make it possible to directly measure the polariton dispersion and damping and to thereby completely reconstruct the complex dielectric constant $\varepsilon(\omega, \mathbf{k})$ in the

IR range. The conventional methods of phonon spectroscopy (e.g., IR spectroscopy) are capable of measuring only the frequency dependence $\varepsilon(\omega)$.

In a study of the shape and a measurement of the widths of lines by the method of spontaneous Raman scattering by polaritons, it is necessary to record a signal when both the spectral width of the spectrometer and the scattered-light collection angle are small ($\Delta\varphi \lesssim 0.1^\circ$) in order to achieve the resolution required in ω and \mathbf{k} spaces. However, as the spectral width and the collection angle are reduced, the signal of interest is significantly attenuated, so extracting the information required becomes more difficult. The net result is that there have been exceedingly few measurements of polariton line widths.

The use of Raman scattering by hot polaritons is preferable for such measurements since in this case the collection angle (the resolution in \mathbf{k} space) is automatically set by the diffraction-level divergence of the laser sources which provide the test beam and the IR beam, while the spectral resolution is set by the laser linewidths. Furthermore, the scattered light in hot-polariton Raman scattering may be several orders of magnitude brighter than the spontaneous scattered light, as we will see below. This circumstance also improves the opportunities for extracting quantitative data from the recorded spectra.

In order to compare the brightness of the Raman scattering by fluctuational scattering and hot polaritons it is convenient to work from a relation which describes a parametric amplification of light ($\omega_l = \omega_s + \omega_p$) when the medium is subjected not only to the seed field due to the fluctuational polaritons but also to auxiliary light at the frequency of the intermediate wave (the polaritons), which falls in a region of strong absorption ($\alpha_p l \gg 1$, where α_p is the absorption coefficient for a polariton wave, and l is the length of the scattering medium in the propagation direction of the interacting waves). Assuming that there is no linear absorption at the frequencies ω_l and ω_s' and following the standard procedure of nonlinear optics, we find the following expression for the spectral brightness B_s of the scattered light (the energy carried across a unit surface per unit time in a unit solid angle and in a unit spectral interval), assuming weak (linear) parametric amplification:

$$B_s = B_{s0} (1 + \bar{N}_p + N_p / \alpha_p l) g l. \quad (1)$$

Here

$$B_{\omega_0} = \hbar \omega_0^3 n_0^2 / 8\pi^3 c^2 \quad (2)$$

is the spectral brightness of the fluctuations of the electromagnetic vacuum, and \tilde{N}_p is the number of quanta in the mode of the fluctuational field which is generated by the surrounding medium in the scattering layer: $\tilde{N}_p = [\exp(\hbar\omega_p/kT) - 1]^{-1}$, where N_p is the number of photons in the polariton illumination mode at the entrance face of the crystal, and g is the parametric gain, given by

$$g = \beta \frac{\alpha_p}{\Delta k^2 + (\alpha_p/2)^2} \quad (3)$$

Here $\Delta \mathbf{k} = \mathbf{k}_l - \mathbf{k}_s - \mathbf{k}'_p \equiv \mathbf{k} - \mathbf{k}'_p$, $\mathbf{k}_p = \mathbf{k}'_p + i\mathbf{k}''_p$ is the complex polariton wave vector,

$$\beta = \frac{8\pi^3}{c^2} \frac{\omega_s \omega_p}{n_l n_s n_p} |\chi|^2 I_l \quad (4)$$

I_l is the intensity of the test wave, and $\chi = \chi'_{ijk} e_i^s e_j^l e_k^p$ is a convolution of the quadratic-nonlinear-susceptibility tensor with the polarization unit vectors of the corresponding waves.

With $N_p = 0$, expression (1) describes the spontaneous Raman scattering of a test wave ω_l , while the term which stems from the presence of N_p describes the scattering component due to the coherent (hot) component of the polariton wave which arises because of the external IR illumination. Under the condition $N_p/\alpha_p l \gg \tilde{N}_p + 1$, relation (1) becomes the well-known expression for the generation of difference frequencies ($\omega_s = \omega_l - \omega_p$), if the frequency ω_p falls in a region of strong absorption.

It can be seen from (1) that the polariton absorption reduces the efficiency of the IR illumination. Nevertheless, the improvement η_Ω , defined as the ratio of the brightness of the scattering by hot polaritons to the total intensity of the spontaneous scattering into a solid angle $\Delta\Omega$, which is equal to the angular divergence of the illumination laser, can reach a significant level. From (1) and (2) we find

$$\eta_\Omega \approx \frac{I_p}{B_{p0} \Delta\Omega \Delta\omega_p \alpha_p l}, \quad B_{p0} = \frac{\hbar \omega_p^3 n_p^2}{8\pi^3 c^2} \quad (5)$$

where I_p is the intensity of the IR beam, and $\Delta\omega_p$ is the spectral width of a polariton scattering line into a unit solid angle, given by

$$\Delta\omega_p = \alpha_p / |u_p^{-1} - u_s^{-1} \cos \psi|. \quad (6)$$

Here u_p and u_s are the group velocities of the polariton wave and of the scattered wave, respectively, and ψ is the angle between \mathbf{k}_s and u_p . For a numerical estimate we adopt $I_p = 1 \text{ W/cm}^2$, $\nu = \omega/2\pi c = 1000 \text{ cm}^{-1}$, $\Delta\Omega = 10^{-4} \text{ sr}$, $n_p = 3$, $l = 1 \text{ cm}$, $\alpha_p = 500 \text{ cm}^{-1}$, and $\Delta\nu_p = 20 \text{ cm}^{-1}$. We find $\eta_\Omega \approx 2 \cdot 10^2$.

The line of the Raman scattering of hot polaritons in \mathbf{k} space has a Lorentzian profile according to (1) and (3), with a width at half-maximum which is equal to the absorption coefficient of the polaritons, $\alpha_p = 2k_p''$, and with a maximum determined by the matching condition $\Delta \mathbf{k} = 0$. By varying the quantity $\mathbf{k} \equiv \mathbf{k}_l - \mathbf{k}_s$ at various fixed frequencies of the IR beam we can thus record \mathbf{k} spectra and in this manner directly measure the dispersion of the real and imaginary parts of the polariton wave vector.

In experiments on spontaneous Raman scattering by polaritons, k is usually varied by scanning the scattering angle (Ref. 5, for example). Another method—applicable only

to anisotropic crystals—might be to make use of the angular dispersion of a wave vector, e.g., that of the test light or that of the scattered light. A method of this sort, which we have used in the present study, is convenient in that the \mathbf{k} spectra are recorded by measuring the power of the scattered light while the test crystal is rotated, but without a change in the relative arrangement of the wave vectors of the interacting waves. Furthermore, the rate of change of k with the angular position of the crystal is significantly smaller than that during scanning of the scattering angle.

We might add that the possibility of a precise measurement of α_p also permits use of Raman scattering by hot polaritons to be used to measure the spectral brightness of IR sources relative to the spectral brightness of the electromagnetic vacuum. In contrast with a "parametric photometer," however, whose physical principles are described in detail in Ref. 6, the lowest brightness which can be measured by the method of hot-polariton Raman scattering should be larger by a factor of about $\alpha_p l$.

3. EXPERIMENTAL PROCEDURE

Raman scattering by hot polaritons was studied in two cases: with the help of cw and periodic-pulse lasers. Figure 1 shows the layout of the experimental apparatus using periodic-pulse lasers. Polaritons are excited by the light from a transverse-excitation atmospheric-pressure pulsed CO_2 laser (1 in Fig. 1; a TEA laser) whose frequency is discretely tunable (over the interval $930\text{--}1090 \text{ cm}^{-1}$). The typical pulse from this laser is short peak with a length on the order of 200 ns followed by a tail which lasts up to $1 \mu\text{s}$. The energy in the peak amounts to 20–50% of the total energy in the pulse, which in turn is 100 mJ (the energy depends on the lasing wavelength; the value cited here corresponds to the maximum value). As the test beam we use the second harmonic ($\lambda_l = 0.532 \mu\text{m}$) of an yttrium aluminum garnet (YAG) laser (4) in Q-switched operation with a pulse repetition frequency of 10 Hz. The energy in a test-light pulse 10 ns long is on the order of 10 mJ. The pulses from the CO_2 and YAG lasers are synchronized in time; the beams from these lasers are brought together by the plate (7) of a BeO single crystal, which transmits the test beam but reflects the IR light. The Stokes scattering is detected in the forward direction with respect to the test and IR light; i.e., the three vectors \mathbf{k}_l , \mathbf{k}_s , and \mathbf{k}_p are parallel. The scattered light is separated from the test beam by a Glan prism (10) and an interference filter (11) (after this step, the light is easily

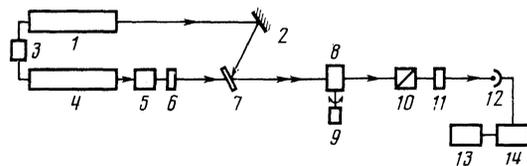


FIG. 1. The experimental layout. 1—Tunable TEA CO_2 laser; 2—mirror; 3—electronic synchronization of the output pulses of the CO_2 and YAG lasers; 4—YAG laser; 5—device which doubles the frequency of the beam from the YAG laser; 6—optical filter which transmits the second harmonic ($0.532 \mu\text{m}$) and absorbs the fundamental frequency of the YAG laser; 7—BeO crystal plate; 8—the test crystal; 9—stepping motor for rotating the test crystal; 10—Glan prism; 11—interference filter which transmits the scattered light; 12—photomultiplier; 13—chart recorder; 14—device for averaging the pulse heights.

observed visually in an undarkened room) and measured by an FEU-79 photomultiplier (12). The output signal from the photomultiplier is averaged and fed to a chart recorder (13). The k spectra are recorded by measuring the power of the scattered light as a function of the angle θ , between the optical axis of the crystal under study (8) and the triad of wave vectors of the interacting waves. This angle is varied by rotating crystal 8 with the help of a stepping motor (9).

In the experiments with cw lasers, the polaritons are excited by the beam from an LG-74 CO₂ laser, while the test beam is produced by an argon laser ($\lambda_l = 0.5145 \mu\text{m}$). The scattered light is analyzed by a Raman spectrometer with a slit width of 4 cm^{-1} . The ω spectra are recorded at various fixed positions of the test crystal (at fixed values of the angle θ).

4. EXPERIMENTAL RESULTS

As the sample we selected a crystal of lithium niobate, LiNbO₃, which is an efficient and widely used material in applied nonlinear optics (see Ref. 7, for example). Several studies have been carried out on the polariton dispersion and phonon spectra of this crystal.⁸⁻¹² It follows, in particular, from those studies that the interval over which the frequency of the CO₂ laser is tuned corresponds to the region of the upper dispersion branch of polaritons, which lies near the highest-frequency longitudinal optical phonon. In order to satisfy the matching condition $\Delta k = 0$ in this part of the spectrum, one must choose a polarization for the test beam which corresponds to the extraordinary wave (e) in the crystal, while the scattered light corresponds to the ordinary wave (o). In this case the scattering should be by ordinary polaritons (of symmetry E), i.e., by polaritons whose frequencies do not depend on the direction of the wave vector in the crystal. This arrangement is convenient for recording k spectra by rotating the crystal at a fixed arrangement of the wave vectors of the interacting waves.

Figure 2 shows the dispersion of ordinary polaritons in the frequency interval of interest here, according to a calculation based on the relation

$$\frac{k_p^2(\nu)}{(2\pi\nu)^2} = \epsilon_\infty + \sum \frac{f_i \nu_i^2}{\nu_i^2 - \nu^2 - i\nu\Gamma_i}. \quad (7)$$

Here ϵ_∞ is the high-frequency dielectric constant, and f_i , ν_i , and Γ_i are the oscillator strengths, frequencies, and attenuation coefficients of transverse optical phonons. The summation is over all the branches of optical phonons of symmetry E . Data for this calculation were taken from Refs. 11 and 12. Figure 2 also shows the values of the angles θ calculated for $\lambda_l = 0.5145 \mu\text{m}$ from the matching position $\Delta k = 0$. For this particular scattering geometry, the matching condition takes the form

$$\Delta k = \Delta k(\nu, \theta) = k_l^e(\nu_l, \theta) - k_s^o(\nu_l - \nu) - k_p^o(\nu), \quad (8)$$

where

$$k_l^e(\nu_l, \theta) = \frac{2\pi\nu_l n_l^o n_l^e}{[(n_l^o)^2 \sin^2 \theta + (n_l^e)^2 \cos^2 \theta]^{1/2}},$$

and n_l^o and n_l^e are the refractive indices for the ordinary and extraordinary rays, respectively, at the frequency of the probe wave, ν_l . The data on the refractive indices required for the calculation were taken from Ref. 13. In nonlinear-optics terms, the functional dependence $\nu(\theta)$ is essentially a

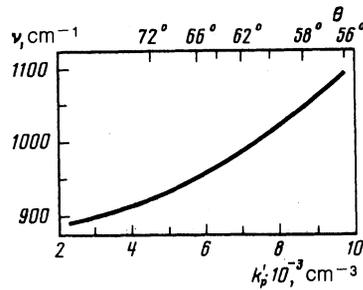


FIG. 2. Calculated polariton dispersion curve $k_p'(\nu)$ and corresponding tuning curve $\theta(\nu)$ for the parametric process $\nu_s = \nu_l - \nu$, $k_s^o k_l^e k_p^o$ in the collinear propagation of the interacting waves; $\nu = \omega/2\pi c$, $\lambda_l = 0.5145 \text{ mm}$.

tuning curve for the parametric process $\nu_l = \nu_s + \nu$. In accordance with this curve, we used a lithium niobate plate cut at an angle of 64° (between the normal to the surface of the plate and the optic axis of the crystal) in the experiments.

Figure 3 shows ν spectra of the Stokes forward scattering, recorded with the help of cw lasers on a Raman spec-

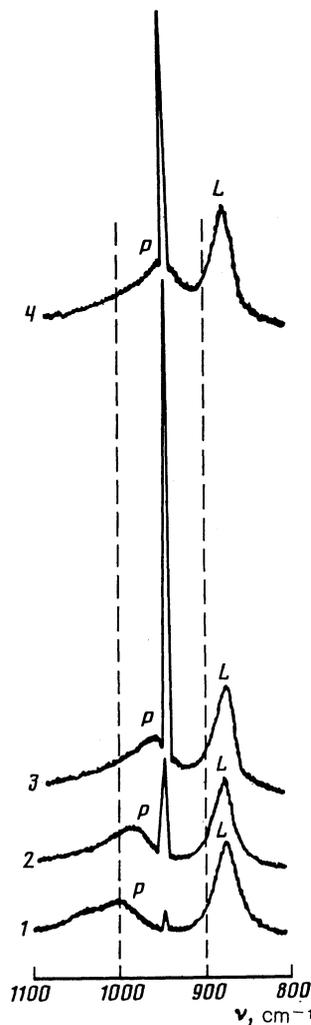


FIG. 3. ν spectra of the Raman scattering of light by polaritons recorded during exposure to IR light at the frequency 944 cm^{-1} at various angles θ : 1— 61° ; 2— 64° ; 3— 67° ; 4— 69.8° .

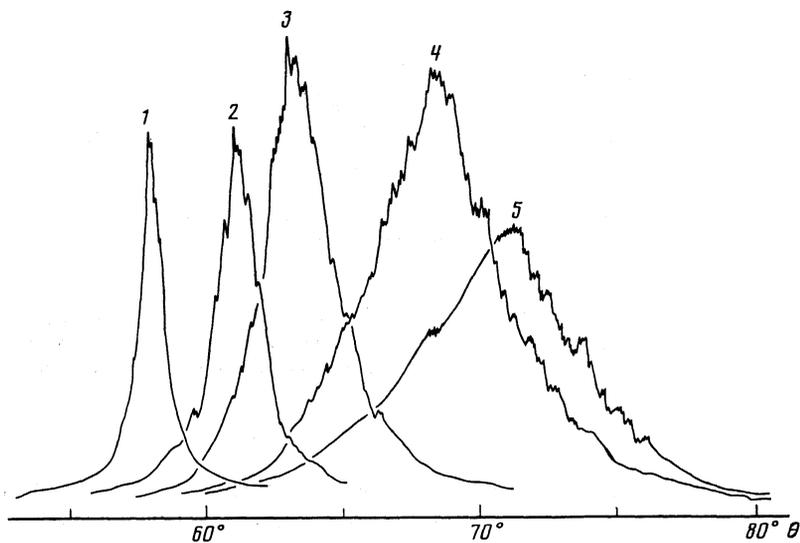


FIG. 4. k spectra of the Raman scattering of light by hot polaritons recorded at various fixed frequencies of the IR light: 1—1087 cm^{-1} ; 2—1031; 3—978; 4—946; 5—931 cm^{-1} . The wavelength of the probing beam is $\lambda_l = 0.532$ mm.

trometer for various angles θ . The angle in which the scattered light was collected was determined by the speed of the spectrometer, $F/6.5$.

The L and P bands correspond to spontaneous Raman scattering of light by extraordinary longitudinal optical phonons [of mixed symmetry: $A_{LO} + E_{LO}$, $\nu_{LO}(A) = 874$ cm^{-1} and $\nu_{LO}(E) = 881$ cm^{-1}] and by ordinary polaritons of symmetry E , respectively. We see that the position of polariton band P depends on θ , as would follow from the curves in Fig. 2. In addition to these bands, the spectra of Fig. 3 reveal a narrow line, whose width is determined by the instrumental function of the spectrometer, with a Stokes shift equal to the frequency, 944 cm^{-1} , of the IR beam. This line corresponds to scattering by hot polaritons (it disappears if there is no IR beam). The θ dependence of the amplitude of this line is described by relation (3). These spectra thus clearly illustrate the significant enhancement of the Raman scattering during coherent illumination of the polaritons. When the matching condition $\Delta k = 0$ holds exactly, and at an intensity on the order of 0.5 W/cm^2 of the CO_2 laser beam the improvement in the amplitude of the signal with respect to spontaneous Raman scattering is about 10^2 , in agreement with the estimate above.

Experiments on the k spectra of the Raman scattering

of light by polaritons were carried out in an apparatus with pulsed lasers (Fig. 1). In this case the spontaneous scattering was vastly weaker than the scattering by hot polaritons, so this spontaneous scattering is not seen in the recorded spectra. Figure 4 shows k spectra recorded at various fixed frequencies of the CO_2 laser. We see that the lines broaden significantly with decreasing frequency of the IR light. This broadening is a consequence of an increase in the phonon component in the polaritons as their frequency approaches the phonon resonance. From these spectra we determined the wave vectors and attenuation of the coherently excited polaritons participating in the scattering, by making a least-squares fit of the spectral lines with a Lorentzian profile, in accordance with (1), (3), and (8). As a result we measured the behavior $k_p'(\nu)$ and $\alpha_p(\nu, k_p')$, shown in Fig. 5. The solid lines here are calculations from Eq. (7), carried out with the help of experimental data on the Raman scattering by optical phonons.^{11,12}

It can be seen from Fig. 5 that the results of the measurements of the dispersion $k_p'(\nu)$ from the spectra of the Raman scattering by hot polaritons agree with the calculations to within the measurement error ($\delta k_p' = \pm 200$ cm^{-1}). However, the discrepancies between the measured values of the polariton absorption $\alpha_p(\nu, k_p')$ and the calcu-

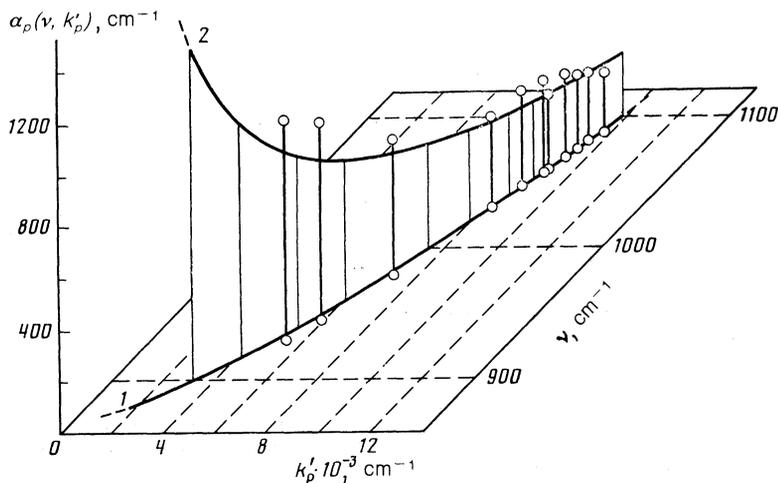


FIG. 5.—Polariton dispersion $k_p'(\nu)$; 2—dispersion of the absorption of polaritons, $\alpha_p(\nu, k_p')$. Circles) results of measurements from the k spectra of the Raman scattering of light by hot polaritons; solid lines) results of a calculation based on data on the Raman scattering by optical phonons.

lated values exceed the measurement error of $\pm 50 \text{ cm}^{-1}$ in certain regions. In the frequency interval $1000\text{--}1100 \text{ cm}^{-1}$ the maximum discrepancy is about 1.5 times the experimental error, while in the interval $900\text{--}1000 \text{ cm}^{-1}$ the discrepancy exceeds the experimental error by a factor of three. These discrepancies are a manifestation of anharmonic effects,¹⁴⁻¹⁶ which were ignored in the calculation of the dispersion of the dielectric constant [see (7)], because the necessary data were not available. The need to incorporate anharmonic effects in a description of the polariton spectra of the lithium niobate crystal is confirmed by the second-order spectra (which correspond to higher harmonics and composite modes of the fundamental lattice vibrations), which we observed during recording of the IR phonon spectra in the interval $900\text{--}1100 \text{ cm}^{-1}$. The polariton Fermi resonance which occurs in the lithium niobate crystal in the frequency region $900\text{--}1100 \text{ cm}^{-1}$ thus leads to a slight broadening of the polariton spectra, while having essentially no effect on the polariton dispersion.

5. ESTIMATE OF THE DENSITY OF NONEQUILIBRIUM POLARITONS

During IR illumination, a steady-state, nonequilibrium polariton density is established in the crystal (this is a steady-state density because the polariton lifetime τ_p is much more shorter than the duration of the IR laser pulse). This nonequilibrium polariton density is given by

$$n = Q\tau_p / \hbar\omega_p,$$

where Q is the power transferred to the polaritons per cubic centimeter through absorption of the IR light.

To estimate the occupation numbers N (the numbers of polaritons in a mode) we need to know the number of excited modes, ρ ($n = \rho N$), which we can estimate from

$$\rho = (2\pi)^{-3} k_{\text{IR}}^2 \Delta k_{\text{IR}} \Delta\Omega \approx (2\pi)^{-3} \Delta k_{\parallel} (\Delta k_{\perp})^2 \approx V^{-1},$$

where $V \approx S/\alpha_p$ is the volume of the effective interaction of the IR light with the medium, k_{IR} is the average wave vector of the IR illumination beam, Δk_{\parallel} is the spread in the component of k_{IR} along the propagation direction, which satisfies $\Delta k_{\parallel} \approx \alpha_p$, and $(\Delta k_{\perp})^2$ is on the order of the cross-sectional area of the beam, S .

Making the further assumption that all of the incident IR light is absorbed uniformly in the crystal over a distance $1/\alpha_p$, we find

$$N = \tau_p P_{\text{IR}} / \hbar\omega_p,$$

where P_{IR} is the power of the IR beam. For an estimate we will see parameter values corresponding to the conditions and results of the experiments which were carried out: $P_{\text{IR}} = 100 \text{ kW}$, $\nu_p = 10^3 \text{ cm}^{-1}$ ($\hbar\omega_p \approx 2 \cdot 10^{-20} \text{ J}$), $\tau_p = (\alpha_p u_p)^{-1} = 4 \cdot 10^{-13} \text{ s}$ [for lithium niobate, according to the experimental results (Fig. 5), we have $\alpha_p \approx 400 \text{ cm}^{-1}$ and $u_p \approx 0.64 \cdot 10^{10} \text{ cm/s}$ at $\nu_p = 10^3 \text{ cm}^{-1}$], and $V = 2.5 \cdot 10^{-4} \text{ cm}^3$. As a result we find $N = 2 \cdot 10^{12}$ quanta/mode and $n = NV^{-1} = 8 \cdot 10^{15}$ quanta/ cm^3 . At room temperature, the equilibrium polariton population \tilde{N}_p at the frequency 10^3 cm^{-1} would be about $7 \cdot 10^3$ quanta/mode.

6. CONCLUSION

This study has shown that the Raman scattering of light by coherently excited (hot) polaritons is an effective spec-

troscopic method and has several advantages over the method of spontaneous Raman scattering. These advantages are the high brightness of the scattered light, the high resolution in both ω space and k space, and the selectivity of the excitations which are detected. This method opens up the possibility of measuring the spectral brightness of sources of radiation in the mid- and far-IR ranges in units of the spectral brightness of the electromagnetic vacuum. There is the further possibility of studying nonequilibrium excitations and, in particular, directly measuring the mean free path of polaritons.

In the experiments, we studied the spectra of the Raman scattering of light by hot polaritons of the upper dispersion branch of a uniaxial lithium niobate crystal, using both pulsed and cw lasers. In the course of the experiments we made use of the angular dispersion of the wave vector of the test beam in an anisotropic crystal, so that—in contrast with the conventional approach⁵—we were able to record the k spectra of ordinary polaritons at a fixed relative arrangement of the wave vectors of the interacting waves. Working from the k spectra which we recorded, we measured the polariton dispersion $k_p'(\nu)$ and the polariton absorption dispersion $\alpha_p(\nu, k_p')$ over the frequency range $930\text{--}1090 \text{ cm}^{-1}$. The results show, in particular, that a polariton Fermi resonance is manifested in this region; this resonance has only a slight effect on the polariton dispersion, but it has a significant effect on the dispersion of the absorption; i.e., it broadens the polariton scattering spectra.

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¹For definiteness, we will discuss only the Stokes component of the scattering.

²The spectra of spontaneous Raman scattering by polaritons in ω and k spaces have been discussed in many places in the literature (see, e.g., Ref. 5 and the bibliography there).

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