NEW MANIFESTATION OF POLARITON FERMI RESONANCE IN RAMAN SCATTERING OF LIGHT IN $LiIO_3$ CRYSTAL

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We have observed and investigated new manifestations of Fermi resonance in the spectrum of spontaneous Raman scattering of light by polaritons in an $LiIO_3$ crystal. They occur in the region of intersection of the upper polariton branch with the branches of three closedly adjacent "weak" polar oscillations, and consist, in particular, in the appearance of a threefold splitting of the dispersion curve of the scattering polaritons.

Manifestations of polariton Fermi resonance in the spectra of spontaneous Raman scattering (SRS) of light were first observed in the crystal $K_3Cu(CN)_4$ [1] and then in the crystal α-HIO_3 [2]. We have registered a triple polariton Fermi resonance in the SRS spectrum on the upper polariton branch of the $LiIO_3$ crystal and investigated its evolution with changing scattering geometry.

The scattering was excited by the λ_ℓ = 4882 Å line of an argon laser of 200 mW power. The single-crystal $LiIO_3$ (6 mm thick) was cut in such a way that its optical axis made an angle 23 ± 1.5° with the normal to the opposite faces. The extraordinary pump wave propagated in the xz plane at an angle α_ℓ to the z axis. The SRS spectra were registered by a well-known photographic procedure [2] with simultaneous sweep over the frequency ω_s and the scattering angle θ. The entrance slit of the spectrometer was parallel to the y axis. Its spectral width was 15^{-1} cm^{-} and the exposure time was about 45 minutes. The axes x, y, and z coincide with the crystallographic axes chosen in accordance with [3]. In particular, the axis is the z axis is the optical axis of the crystal. To prevent exposure of the photographic plate to the radiation of the argon plasma, the direction to the slit made a fixed angle $\theta_0 = 0.45°$ with the pump wave vector $\vec{k}_\ell$, so that scattered radiation with $\theta \geq 0.45°$ was registered. The measurements were performed at different angles α_ℓ between $\vec{k}_\ell$ and the z axis; the angles were determined accurate to 2°.

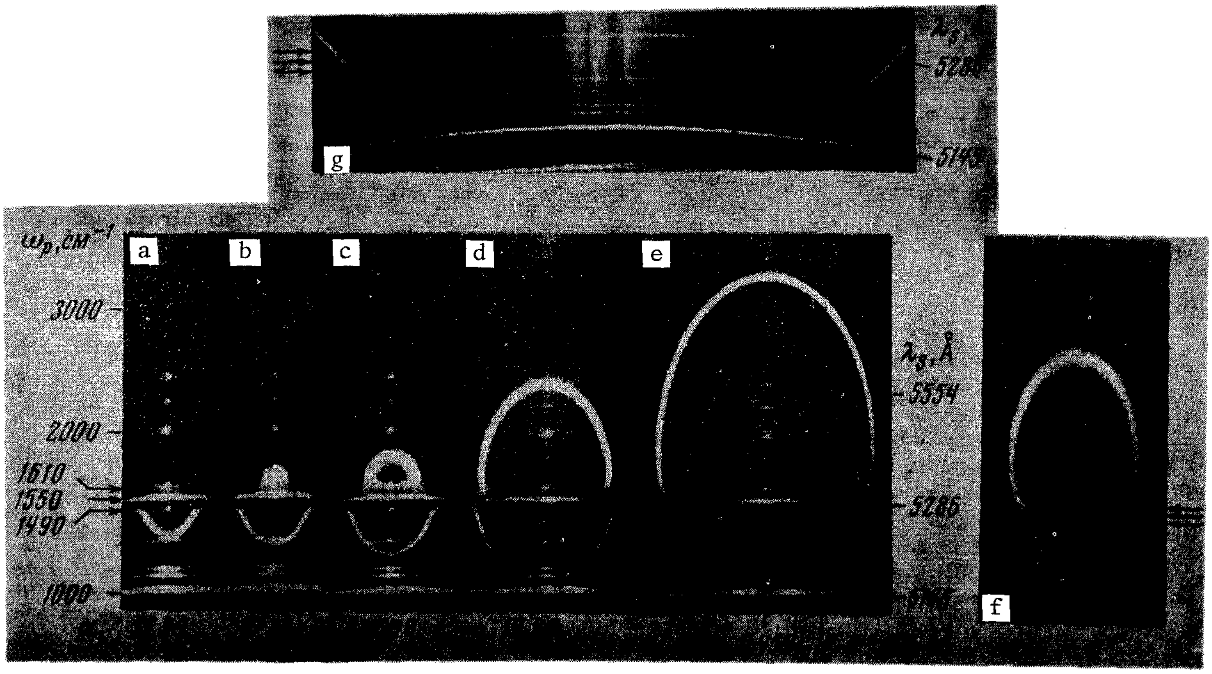

Fig. 1

Figures 1a - 1g show a characteristic series of spectrograms. The outer "ellipse" corresponds to the extraordinary polariton wave, while the internal (less bright) ellipse corresponds to the extraordinary wave. The latter is seen only on part of the spectrograms; at $\theta = \theta_0$ the scattering by extraordinary polaritons is forbidden by the selection rules. This scattering appears when θ is increased, but remains weak. The decrease of the intensity in the lower part of the ellipses in the vicinity of the polariton frequency $\omega_p = \omega_\ell - \omega_s = 1000\ \mathrm{cm}^{-1}$ is due to mutual cancellation of the purely electronic and electron-ion contributions to the quadratic nonlinear polarizability χ (cf. [4, 5].

We see that there are three resonances (marked by arrows in Figs. 1 - 3) with weak polar vibrations at the frequencies $\omega_1 = 1490\ \mathrm{cm}^{-1}$, $\omega_2 = 1550\ \mathrm{cm}^{-1}$, and $\omega_3 = 1610\ \mathrm{cm}^{-1}$ (they are determined with approximate accuracy 20 cm^{-1}). The vibrations $\omega_{1,2,3}$ can be tentatively interpreted as second-order vibrations corresponding to the group of principal vibrations of LiIO [6] $\nu(E_2) = 765\ \mathrm{cm}^{-1}$, $\nu(E_1) = 769\ \mathrm{cm}^{-1}$, and $\nu(A) = 795\ \mathrm{cm}^{-1}$. A more exact interpretation is difficult, since the rules for the formation of the energy spectrum, corresponding to second-order vibrations in crystals of the $LiIO_3$ type, have not been sufficiently thoroughly established at present.

Unfortunately, the frequency region in question includes the argon-plasma line 5286 Å, which makes the observation difficult. All the lines outside the region of the resonance, which appear on the spectrograms on top of the ellipse, also belong to the argon plasma. Figure 1e shows one of the spectrograms obtained by excitation with the 4965 Å line, which is weaker than the 4882 Å line. There is no parasitic line in the resonance region in this case, but the decrease in the scattering intensity causes the picture quality to deteriorate.

The picture of the "ellipses" varies strongly with α_ℓ. In particular, when α_ℓ is decreased the upper part of the "ellipse" contracts into a spot and then vanishes. We note also that the Fermi resonance is most clearly pronounced for those α_ℓ at which the ellipse dimensions are commensurate with the dimensions of the resonance region. The method employed here to reveal polariton resonances with weak polar vibrations, based on choosing the appropriate region of α_ℓ variation and using an extraordinary pump wave, is quite effective. Figure 1g shows a spectrogram obtained at $\alpha_\ell = 30°$ (the spectral width of the slit was 4 cm^{-1} in this case); practically no resonances can be seen on this spectrogram. Nor did Winter [7], who used a scattering geometry with $\alpha_\ell = 90°$, succeed in observing any resonances.

Using the energy-momentum conservation laws, we calculated the dependence of $\omega_p(0)$ for the external ellipse. The contribution of the weak vibrations to the dielectric constant of the crystal was approximated by an expression of the same type as for the strong vibrations [4, 8] with allowance for the fact that the lines corresponding to weak vibrations are sharp enough; incidentally, the main laws governing the spectrum can hardly be very sensitive to the explicit form of the approximation. The oscillator strengths s_f of the weak vibrations $\omega_{1,2,3}$ were chosen such as to reconcile the sections of the theoretical and experimental $\omega_p(\theta)$ curves adjacent to the resonances. It turns out that one can put $s_f = 10^{-4}$ for all three vibrations. The values of the remaining parameters were borrowed from [6, 9]. The results of the calculations are shown by the curves of Fig. 2. The experimental data are indicated, with allowance for their errors, as individual points obtained from the spectrograms. The calculated and experimental (in parentheses) values of α_ℓ are also indicated. We see that the agreement between theory and experiment is satisfactory.

Putting $\alpha_\ell = 18.3°$ and using the formulas obtained in [8, 10], we have performed an illustrative calculation of the spectrum of the gain g that determines the scattering intensity [11]. The necessary parameters of the "weak" vibrations were chosen in such a way that their contribution to χ is 10^{-3} of the contribution of the 769 cm^{-1} fundamental vibration, and the half-widths of their proper SRS lines were assumed to be 30 cm^{-1}. The calculation results are given in Fig. 3, which shows the character and evolution of the $g(\omega_p)$ spectrum with changing θ. These results are in qualitative agreement with the experimental data.

The described regularities constitute new manifestations of the polariton Fermi resonance.

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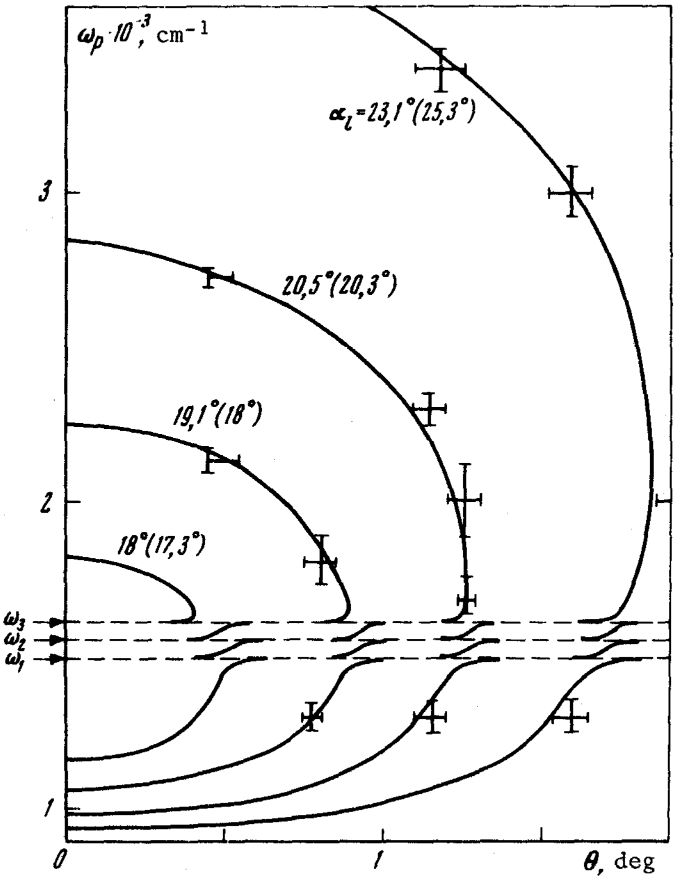

Fig. 2

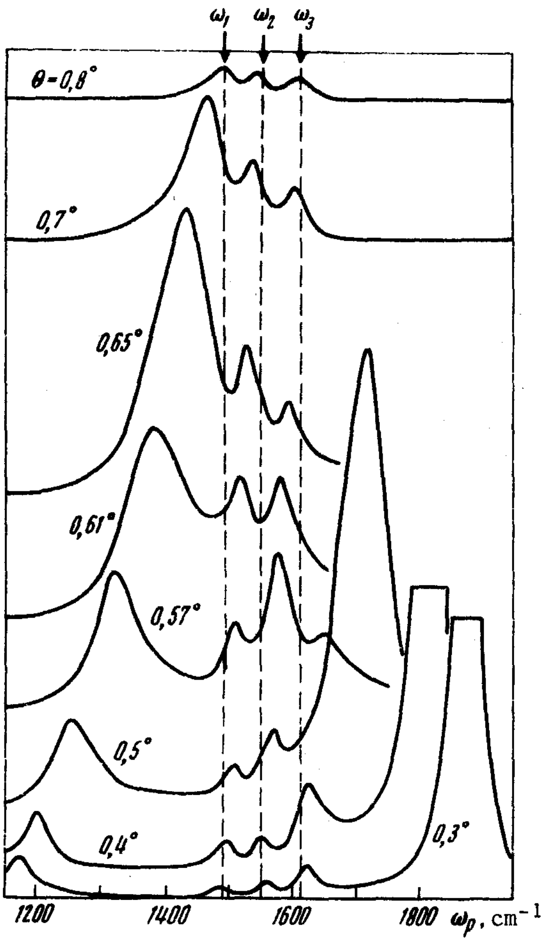

Fig. 3

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