Rayleigh scattering of polaritons in CdS crystals

A. B. Pevtsov, S. A. Permogorov, and A. V. Sel'kin
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

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The Rayleigh scattering spectra of polaritons have been studied for the first time. Elastic scattering of light in the vicinity of the $A_{n=1}$ exciton resonance of CdS crystals at $T = 2$ K is determined primarily by a single scattering of polaritons by a short-range potential.

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Recent years have seen substantial progress in research on the resonant scattering of excitonic polaritons. Attention has been focused primarily on the inelastic component of the secondary emission, which is associated with the Raman[1] and Brillouin[2] scattering mechanisms. There has been essentially no study of elastic scattering (scattering without a frequency shift).

In crystals with smooth surfaces the primary mechanism for elastic scattering is Rayleigh scattering by volume defects and impurities. The possibility of this type of scattering was discussed by Ivchenko *et al.*,[3] who derived a theory for the spectral and polarization characteristics of the elastically scattered light, taking into account the polariton effects in multiple reabsorption and re-emission processes. Recent theoretical[4] and experimental[5] research on the resonant inelastic scattering of polaritons indicates that elastic processes have an important effect on the spectra of Brillouin and Raman scattering.

In this letter we are reporting a study of the resonant Rayleigh scattering of polaritons by impurities. As samples we selected CdS crystals, whose exciton–polariton spectrum is well known. The scattering was studied in a crystal at a temperature of 2 K in the vicinity of the $A_{n=1}$ exciton resonance in the "backscattering" geometric arrangement shown at the left in Fig. 1.

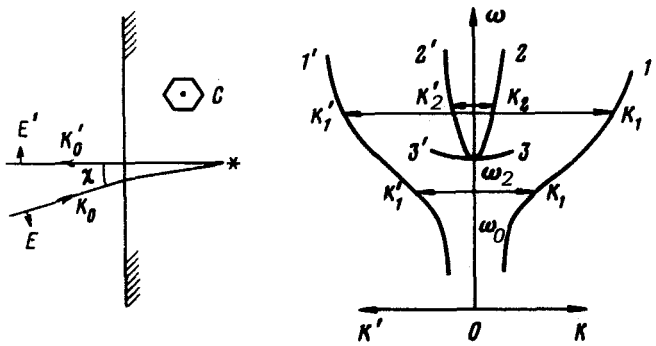

FIG. 1. Left–Elastic-scattering geometry; right–schematic diagram of the Rayleigh "backscattering" of polaritons.

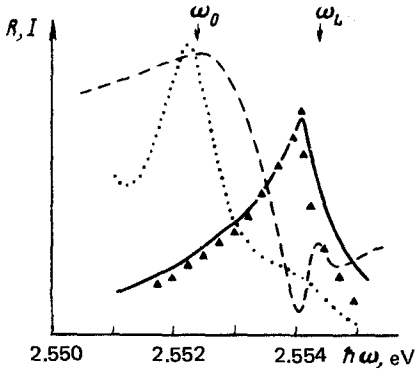

FIG. 2. Triangles–Experimental spectrum of the elastic scattering of light by a CdS crystal ($T = 2$ K) in the vicinity of the $A_{n=1}$ exciton resonance; solid curve–theoretical spectrum for Rayleigh scattering of polaritons; dashed curve–reflection spectrum for normal incidence; dotted curve–spectrum of exciton–polariton luminescence. The ordinate scale is arbitrary.

The scattering plane, which is the plane containing the wave vectors $\mathbf{K}_0$ and $\mathbf{K}'_0$ of the incident and scattered photons, was oriented perpendicular to the **C** axis (the optic axis) of the crystal and to its emitting face. Nearly monochromatic exciting light, with a linewidth ~0.5 meV, was selected by an ISP-51 monochromator from the continuous spectrum of an incandescent lamp. This exciting light was incident in the polarization $\mathbf{E} \perp \mathbf{C}$ on the crystal at a minimum angle ($\chi = 22°$) with respect to the normal to the emitting surface. The scattered light was detected along the normal in the polarization $\mathbf{E}' \perp \mathbf{C}$ by a DFS-24 spectrometer. The spectrum of the scattering line was recorded at various fixed frequencies of the exciting light in the exciton-resonance region. The maximum of the scattered line coincided in spectral position with the maximum of the excitation line over the entire range of incident frequencies, and there was no significant change in line shape.

The triangles in Fig. 2 show the experimental results for the peak intensity of the scattering line, plotted as a function of the frequency of the excitation line. Also shown here are the specular-reflection spectrum of this crystal for the case of normal incidence (the dashed curve) and the exciton–polariton luminescence spectrum as observed along the normal to the surface. This luminescence was excited in the interband absorption band by the 4416-Å line of a He–Cd laser (the dotted curve).

Comparison of the experimental curves reveals that the scattering spectrum is quite different from the luminescence spectrum. The maximum scattering intensity is at the absolute minimum of the reflection, slightly below the longitudinal frequency ω_L (the position of the short-wave peak in the reflection spectrum[6]). The maximum scattering intensity is ~0.02% of the specular-reflection intensity at the exciton resonance frequency.

We can state with assurance that the scattering spectrum in Fig. 2 is attributable to elastic processes, since the elastic-scattering intensity in CdS crystals is at least three orders of magnitude higher than the intensity of the Brillouin components.[7] On the other hand, the Brillouin shift near the frequency ω_L of the $A_{n=1}$ exciton in

CdS reaches values ~1 meV (Ref. 7). If the Brillouin component were significant in the overall scattering, this shift would have been detected in the present experiments.

The observed scattering spectra agree qualitatively with the results reported by Ivchenko *et al.*[3] for the case of single scattering by an impurity.

The single elastic scattering of polaritons in the "backscattering" arrangement is shown schematically at the right in Fig. 1. At frequencies $\omega < \omega_L$, only polaritons of branch 1 ($1 \to 1'$) participate in the scattering; in a collision with an impurity, these polaritons undergo a change in wave-vector direction, remaining on the same branch. At frequencies $\omega > \omega_L$ there may be an elastic transition from one branch to another; i. e., in addition to the scattering events $1 \to 1'$ and $2 \to 2'$ there may be transitions $1 \to 2'$ and $2 \to 1'$. Transitions involving longitudinal excitons 3 and 3′ do not contribute to the external emission in this single-scattering geometric arrangement.

By assuming (a) that the Rayleigh scattering of polaritons by a short-range potential, whose Fourier components are independent of the wave vector, is a single scattering, (b) that there is no interference between polariton states in the course of the scattering, and (c) that the "dead"-layer model is valid,[6] we find the following expression for the scattered light intensity in the geometric arrangement of Fig. 1:

$$I \sim (1-R)^2 |W|^2 N \frac{(v_1+v_2)^2 + 4v_1v_2}{\Gamma (v_1+v_2)^3}, \qquad (1)$$

where $|W|^2$ is the square modulus of the matrix element for scattering of a polariton by an impurity, N is the impurity concentration, Γ is the exciton damping constant, R is the specular reflection coefficient, and $v_i (i = 1,2)$ are the rates of energy transfer by polariton modes 1 and 2.

In calculating the scattering spectrum from Eq. (1), we used numerical values from Ref. 8 for the parameters of the $A_{n=1}$ exciton resonance, and we used measured values of R. The results of the calculation, which are represented by the solid curve in Fig. 2, correspond well to the observed scattering spectrum, thereby confirming the assumptions adopted for the calculation.

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