Resonant elastic scattering of light by fluctuations of the surface exciton potential

V. A. Kosobukin and A. V. Sel'kin
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

(Submitted 16 July 1986)
Pis'ma Zh. Eksp. Teor. Fiz. **44**, No. 8, 377–380 (25 October 1986)

The cross section and angular distribution of the resonant elastic scattering of light by CdS crystals ($T = 2$ K) have been measured in the vicinity of the $A_{n=1}$ exciton state. The scattering results from fluctuations of the exciton potential barrier, which can be approximated by an exciton-free ("dead") layer with a mean square roughness height $h = 5$ Å at the inner surface and with a correlation length $l = 0.5$ μm.

The resonant elastic scattering of light in the spectral region of an exciton state was first pointed out in Ref. 1. A mechanism of Rayleigh scattering of polaritons by defects or impurities in the crystal was proposed in an effort to explain the effect. In the present letter we report new experimental data on the elastic scattering of light

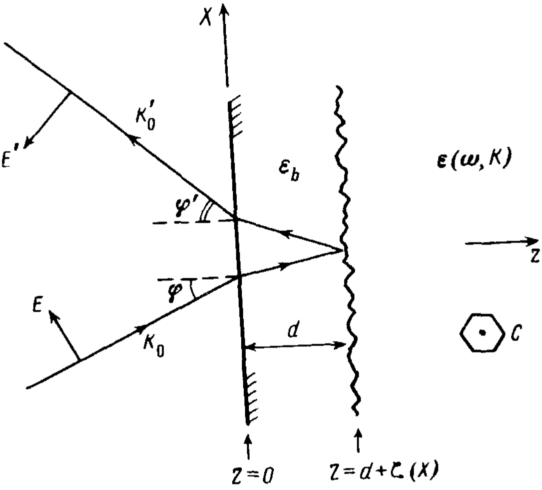

FIG. 1. Light scattering geometry. The random function $\zeta(X)$ describes the profile of the rough scattering surface of the dead layer; d is the average thickness of this layer.

near the $A_{n=1}$ exciton state of CdS crystals. These results cannot be described satisfactorily by the model proposed previously.

Figure 1 is a schematic diagram of the experimental geometry in our study of the resonant elastic scattering of light. The scattering plane, which is the XZ plane and is perpendicular to the (hexagonal) optic axis (C) of the crystal, contains the polarization vectors E and E' of the incident and scattered light. The angles of incidence and scattering, $\varphi + \varphi'$, are varied as the crystal is rotated around the C axis in such a way that the angle $\varphi + \varphi' = 18°$ remains constant. This experimental geometry corresponds to resonant conditions for the scattering of light, since the optical transition to the $A_{n=1}$ exciton state in CdS is allowed only for light with the polarization $E \perp C$. The methodological details of this experiment have been reported previously.[1,2]

In the experiments we measure the dimensionless scattering cross section $\sigma(\varphi', \varphi)$, defined as the ratio of the intensity of the light scattered at an angle φ' to the energy flux density of the exciting light, which is incident at an angle φ (Fig. 1). The measurements of σ are taken by comparing the intensities of the diffuse and specular components of the scattered light. The experimental points in the inset in Fig. 2 show the spectrum of $\sigma(4°, 14°)$; the transverse and longitudinal frequencies ω_0 and ω_L are found from an analysis of the reflection spectra.[3] The scattering spectrum is noticeably asymmetric, with a maximum near ω_L. As the angle φ is increased (or φ' decreased), the maximum value of the cross section, $\sigma_{\max}$, decreases sharply, as demonstrated in Fig. 2 by a plot of $\sigma_{\max}(\varphi', \varphi)$ versus the angle of incidence φ (the points are experimental).

The value of $\sigma_{\max}(4°, 14°) = 2.4 \times 10^{-3}$ (Fig. 2) can be found in the model[1] of single Rayleigh scattering of a polariton by a short-range impurity potential or defect potential if the exciton damping parameter (Γ') associated with the scattering is assumed to be $\Gamma' \approx 10 \cdot \Gamma$, where Γ is the total damping constant of the exciton. By virtue of its meaning, however, have $\Gamma' \leqslant \Gamma$; furthermore, we have $\Gamma' \ll \Gamma$ for single scattering (as is indicated by the weak depolarization of the light during scattering[2]). In order to

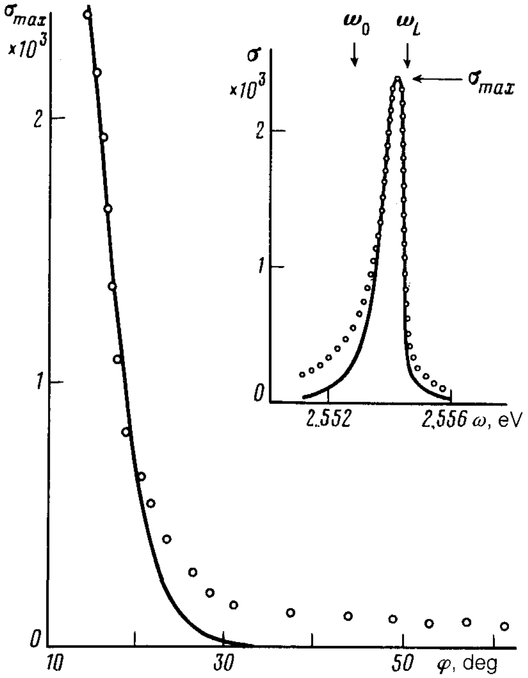

FIG. 2. The maximum value ($\sigma_{\max}$) of the cross section for elastic scattering of light by the CdS crystal ($T = 2$ K) near the $A_{n=1}$ exciton resonance as a function of the scattering angle φ under the condition $\varphi + \varphi' = 18°$ (φ' is the scattering angle). The inset shows the scattering spectrum for $\varphi = 14°$, $\varphi' = 4°$. The points are experimental, the solid curves theoretical.

explain the angular dependence of the cross section ($\sigma_{\max}$ decreases by an order of magnitude as the angle of incidence φ is varied from 14° to 28°) in the same model, we would have to assume an anomalously pronounced anisotropy of the scattering. Taking into account the large value of the refractive index ($n \approx 15$) at the frequency of the scattering maximum, we should attribute the effect to centers which are almost absolutely anisotropic. The existence of such centers does not seem likely.

These difficulties can be eliminated by relating the observed effects to a Rayleigh scattering by fluctuations of a surface exciton potential. Figure 1 explains the model used in the calculations. An external light wave is incident on the plane outer surface of the crystal ($Z = 0$) and is scattered diffusely by the rough inner surface, $Z = d + \zeta(X)$, of the exciton-free dead layer.[3] Here d is the average thickness of the dead layer, and $\zeta(X)$ is a random function describing the profile of the rough surface, with an average value $\langle \zeta(X) \rangle = 0$ over the ensemble of realizations of these functions. The dead layer is characterized by a background dielectric constant ϵ_b; the region of the crystal $Z = d + \zeta(X)$ is described by a dielectric constant $\epsilon(\omega, K)$, which depends on the wave vector K and incorporates the contribution of excitons.[3] The elements of the roughness are assumed to be small $|\zeta/\lambda| \ll 1$, where λ is the shortest length of a normal wave excited at the boundary $Z = d + \zeta(X)$, and smooth ($|\nabla \zeta| \ll 1$). It is thus sufficient to restrict the analysis to first-order perturbation theory in the roughness height in writing boundary conditions, including the additional boundary conditions.

The experimental data of Ref. 2 shows that the spectrum of diffuse scattering of light by irregularities of the boundary of a crystal (the outer boundary of the dead layer) basically reproduce the spectrum of specular reflection. The scattering spectrum, also shown in Fig. 2, is radically different from the experimental spectrum of specular reflection, but its shape is extremely close to that calculated in Ref. 3 for the spectrum of specular reflection from the boundary between semi-infinite media with ϵ_b and $\epsilon(\omega, K)$ (from the inner surface of the dead layer). It is thus important to take into account the roughness of specifically the inner surface of the dead layer, which corresponds physically to spatial fluctuations of the sharp potential relief for an exciton near the surface of the crystal.

The solid curves in Fig. 2 show results calculated by taking the roughness of the inner boundary of the dead layer into account. The correlation function of the roughness is chosen to be Gaussian[4]: $\langle\zeta(X)\cdot\zeta(X')\rangle = h^2; \exp[-(X-X')^2/l^2]$, where h is the mean square height, and l is the correlation length of the roughness. In fitting the theoretical curves to the experimental data, we varied h and l; the other parameters of the $A_{n=1}$ exciton resonance in Cds at $T = 2$ K correspond to the experimental specular-reflection spectral.[3] The angular dependence of the scattering intensity (Fig. 2) is determined by the parameter l, while $\sigma_{\max}$ (see the inset in Fig. 2) depends primarily on h. The best agreement between theory and experiment is achieved with $h = 5$ Å and $l = 0.5\ \mu$m (in which case we have $d = 70$ Å; Ref. 3). These values of the parameters h and l satisfy the initial approximations of the theory quite well.

There is some difference between the experimental and theoretical curves in Fig. 2, apparently a consequence of the fact that the theoretical description ignores additional scattering mechanisms, e.g., (a) Rayleigh scattering by defects or impurities in the crystals, (b) scattering by outer irregularities of the dead layer, and (c) scattering accompanied by the excitation of surface polaritons (in second-order perturbation theory in the roughness height). With regard to the last mechanism, we should mention that the importance of the correlation length of *natural* roughness in CdS makes it difficult to observe surface polaritons[4] in experiments on the elastic scattering of light.

---

[1] A. B. Pevtsov, S. A. Permogorov, and A. V. Sel'kin, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 419 (1981) [JETP Lett. **33**, 402 (1981)].

[2] A. V. Pevtsov and A. V. Sel'kin, Fiz. Tverd. Tela (Leningrad) **26**, 2875 (1984) [Sov. Phys. Solid State **26**, 1741 (1984)].

[3] A. B. Pevtsov and A. V. Sel'kin, Zh. Eksp. Teor. Fiz. **83**, 516 (1982) [Sov. Phys. JETP **56**, 282 (1982)].

[4] V. M. Agranovich and D. L. Mills (editors), Poverkhnostnye polyaritony (Surface Polaritons), Nauka, Moscow, 1985.

Translated by Dave Parsons