Thermally stimulated brightening of a crystal boundary in the exciton absorption region

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As the temperature of a CdS crystal is raised from 2 to 30 K, the minimum value of the exciton reflection coefficient is observed to decrease. This effect stems from the temperature dependence of the dissipative exciton damping Γ near an interior surface of an exciton-free transition layer. At $T \lesssim 50$ K the $\Gamma(T)$ dependence cannot be explained by an exciton-phonon interaction mechanism.

Back in 1958, Pekar called attention to a possible manifestation of the Brewster effect (an exact vanishing of the reflection coefficient) in the exciton part of the spectrum.[1] He pointed out that the conditions under which the effect is seen might serve as a test of the optical reflection theory which uses auxiliary waves. It follows from a recent detailed analysis[2] of the exciton Brewster effect that the reflection coefficient of an absorbing crystal cannot vanish according to the theory of Ref. 1. If, however, we assume that there is a layer at the crystal surface in which excitons are not excited[3] (a "dead layer"), then it can be shown[2] that for certain relations among the parameters of the dead layer and the exciton resonance the Brewster effect can occur even in the face of absorption (which is always present in reality). In particular, the effect might be arranged by changing the exciton damping parameter Γ, which is generally a very strong function of the crystal temperature.

We report here a study of the reflection and transmission spectra of CdS single crystals in the spectral region corresponding to the exciton state $A_{n=1}$ at various temperatures over the range $2\ \mathrm{K} \leqslant T \leqslant 60$ K. The test samples were crystalline plates about 0.3 mm thick with **C** optic axis lying in the plane of the reflecting face. The reflection spectra were measured in the light polarization $\mathbf{E} \perp \mathbf{C}$ along the propagation direction $\mathbf{K} \perp \mathbf{C}$ at nearly normal incidence. The transmission was studied in a polarization approximating $\mathbf{E} \| \mathbf{C}$ (the angle between **E** and **C** was less than 2°) in the vicinity of the absorption line of the "longitudinal exciton,"[4] A_L. Figure 1 shows some representative reflection and transmission spectra ($T = 44$ K; a—reflection; b—transmission).

As the crystal temperature is raised, the most obvious changes in the reflection spectrum occur near the minimum value of the reflection coefficient, $R_{\min}$. Initially, $R_{\min}$ decreases, and it reaches its lowest value near $T = 30$ K (see the $R_{\min}$, T plane in Fig. 2). Beyond this point, it increases monotonically. Because of the finite spectral and angular resolutions of the apparatus and also because of diffuse scattering, the coefficient $R_{\min}$ does not reach zero in an actual experiment. However, taking into account the sharp change in the phase of the reflected wave, by $\pm\pi$, as the temperature of the crystal is changed near 30 K (Ref. 2), we can assert that we are in fact seeing the Brewster effect, i.e., that $R_{\min}$ does vanish at this temperature.

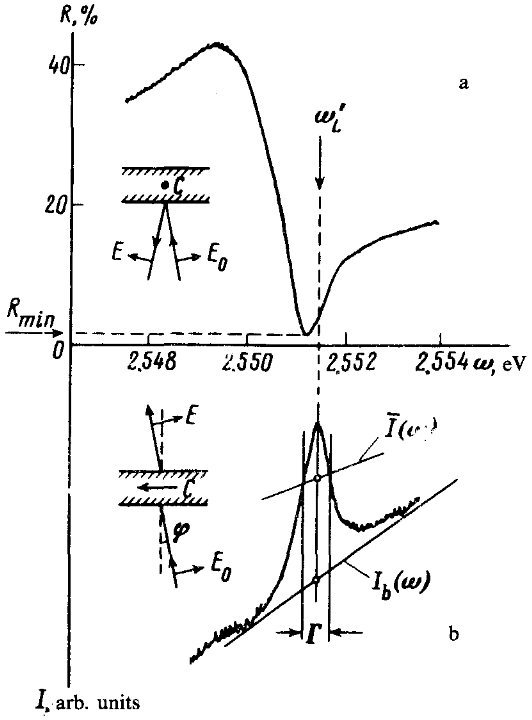

FIG. 1.

Shown in the Γ, $R_{\min}$ plane in Fig. 2 is the theoretical dependence of $R_{\min}$ on Γ (Γ^{-1} is the total exciton lifetime in the coherent state). Here we have used the Hopfield-Thomas model[3] with the parameters which we found in Ref. 2 for the exciton

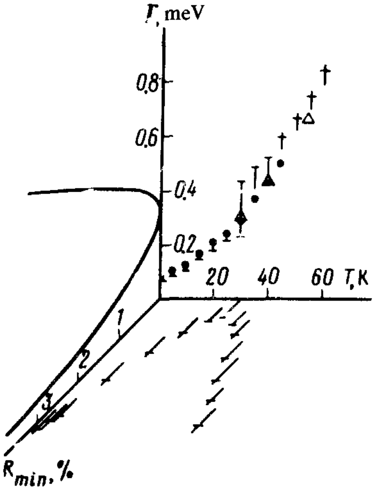

FIG. 2.

resonance $A_{n=1}$. According to these calculations, we would have $R_{\min}=0$ at $\Gamma=0.32$ meV. Working from the experimental data on $R_{\min}(T)$ and the theoretical $R_{\min}(\Gamma)$ curve in Fig. 2, we plotted Γ vs the temperature T (the Γ, T plane; the crosses). Shown in the same figure, by the triangles, are points found through a comparison of the measured temperature dependence of the Brewster angle $\phi_{BR}(T)$ with a theoretical curve of $\phi_{BR}(\Gamma)$.

Since the $\Gamma(T)$ dependence which we have plotted leans heavily on the model of Ref. 3, it is worthwhile to study this dependence by another method, which does not involve study of the reflection spectra. For this purpose we analyzed the shape of the absorption line A_L (Fig. 1b) observed in the light polarization corresponding to the excitation of a mixed mode.

It can be shown that in this configuration the spectrum of the absorption coefficient near the line A_L is Lorentzian (has the classical form) if $\Gamma \gg 2\sin\phi\cdot k_0\cdot(\hbar\omega_{LT}/2M)^{1/2}$, where ϕ is the angle of incidence, k_0 is the vacuum wave vector of the light, ω_{LT} is the longitudinal-transverse splitting, and M is the translational mass of the exciton. For the parameters of the $A_{n=1}$ exciton resonance in CdS, this condition clearly holds over the entire temperature range studied for $\phi \leqslant 2°$. In this case, Γ can be determined directly from the transmission spectra (Fig. 1b), as the energy distance between the points at which the experimental $I(\omega)$ curve intersects the line $\bar{I}(\omega)=(I(\omega_L')/I_b(\omega_L'))^{1/2}I_b(\omega)$, where $I_b(\omega)$ is a linear function of the frequency which approximates the background transmission, and ω_L' is the frequency which corresponds to the minimum intensity, $I_{\min}=I(\omega_L')$, of the light transmitted through the crystal plate.

The filled circles in Fig. 2 show the temperature dependence of Γ constructed from this analysis of the shape of the A_L transmission line. We see that there is a good agreement between the results found for $\Gamma(T)$ by the three measurement methods. We should add that our measured values of $\Gamma(T)$ are also in approximate agreement with the values found in Ref. 5 from an analysis of the integral absorption intensity of thin CdS plates in the light polarization $\mathbf{E}\perp\mathbf{C}$.

In summary, the values found for $\Gamma(T)$ from the analysis of the reflection spectra in the dead-layer model agree well with the results of independent transmission measurements. We may thus regard the observed decrease in the reflection coefficient (the brightening of the boundary) with increasing temperature as the most direct proof of the existence of a dead layer at the surface of the crystal.

Our measured $\Gamma(T)$ dependence is of interest in its own right, in connection with the effort to explain the mechanism for energy dissipation in the exciton part of the spectrum. We have attempted to carry out a quantitative analysis of this dependence on the basis of an exciton-phonon interaction mechanism, working from the results of Ref. 6 (both the piezoelectric interaction of excitons with phonons and their interaction through the strain energy were taken into account). Completely unexpectedly, we found that the experimental values of $\Gamma(T)$ are nearly two orders of magnitude higher than those predicted by the theory. Another fact which argues against an exciton-phonon scattering mechanism is the significant deviation of the experimental $\Gamma(T)$ curve from linearity.

It may be that the observed values of Γ are determined by mechanisms involving the scattering (and capture) of excitons by defects or impurities. In this case, the temperature dependence of Γ might be governed by a thermally activated tunneling of excitons through a potential barrier around the defect or impurity atom.[7]

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