Light transmission through a CdS crystal wedge with allowance for additional light waves

A. A. Demidenko, M. V. Lebedev, S. I. Pekar, M. I. Strashnikova, V. B. Timofeev, and B. E. Tsekvava

Institute of Solid State Physics, Acad. Sci. USSR; Institute of Physics, Acad. Sci. Ukr. SSR; and Semiconductor Institute, Acad. Sci. Ukr. SSR
(Submitted 5 March 1985)
Zh. Eksp. Teor. Fiz. **89**, 330–335 (July 1985)

Light transmission through a thin wedge-shaped CdS crystal near the fundamental $A_{n=1}$ exciton resonance is analyzed when additional light waves are generated. The transmission and the deflection angles of the two identically polarized + and − light waves are calculated, and the theoretical and experimental results are compared in detail.

1. INTRODUCTION

One of the early papers[1] on the theory of additional light wave (ALW) generation considered possible experiments for directly verifying the theoretical predictions regarding light refraction by thin wedge-shaped crystals. This method was first employed by Broser *et al.*[2] to analyze exciton polaritons in CdS; however, they were unable to record the simultaneous transmission of the two identically polarized light waves because the light was strongly absorbed near the resonance frequency for the dipole-allowed exciton transition, where the spatial dispersion was greatest. This difficulty was overcome by using super-thin CdS crystals and a tunable dye laser.[3] Light transmission through an absorbing wedge was examined theoretically in Ref. 4, where ALW was treated. In the present paper we describe the experiments in detail and compare the results with theoretical calculations.

2. EXPERIMENT

Figure 1 shows a sketch of the experimental setup. We used a tunable coumarin 152A dye laser pumped by pulses from a nitrogen laser. The laser pulses were 6 ns long, the pulse power was ≈ 100 W, the spectral width was 0.3 Å, and the repetition rate was 100 Hz. The laser light was elliptically polarized with vertical/horizontal amplitude ratio (ellipticity factor) ≈ 10. In order to facilitate the mounting of the crystal, we actually employed a linearly (horizontally) polarized laser beam which then passed through a $\lambda/2$-wave plate and polarizer P_1. Because the amplitudes of the ordinary and additional light waves are comparable only in a very narrow spectral interval and differ elsewhere by several orders of magnitude, it was necessary to minimize the light losses. We did this by using a lens L_1 and microscope objective O_1 to collimate and focus the laser beam to a diameter $\approx$ 1 mm. The crystal was placed in an optical cryostat cooled by superfluid helium, and the experiment was carried out at 1.8 K. The beams diffracted by the crystal were focused by a lens L_2 ($f = 80$ mm) to spots of diameter 15 μm in the focal plane F, which also contained a movable diaphragm that selected the central part of the beams. The crossed slits S were used to limit the light beam transmitted by the crystal. This was done by a lens L_2 which magnified the image of the crystal by roughly a factor of 2 in the plane of the slits. The second polarizer P_2 was used to select the desired polarization efficiently. The optical system contained a rotating prism P, which when combined with the mirror M_1, long-focusing objective O_2, and microscope M enabled us to observe to movement of the spots in the plane F. When the prism was not used, lens L_3 focused the light on the entrance slit of a DFS-12 double monochromator. The light-sensitive SIT matrix of an OMA-2 optical multichannel analyzer was located at the exit from the monochromator. This enabled us to quickly monitor the spectral composition of the light in the F plane and measure its intensity. The neutral filters F_1 attenuated the signal at the wavelengths to which the crystal was transparent, while the filters F_2 helped to view the image.

The CdS single crystal wafer used in the experiment measured 2×1 mm and was grown from the vapor phase; its cross section was a wedge whose two sides formed an edge of angle α. The C_6 hexagonal axis lay in the plane of the wafer parallel to the edge, and the wafer was 0.7 μm thick at the base of the wedge. The angle $\alpha = 2.48 \cdot 10^{-4}$ rad was measured by microscopic analysis of interference fringes of equal width parallel to the edge. The crystal was positioned freely inside a specially constructed quartz cell; stress generation in the crystal was avoided by eliminating optical contacts with the crystal holder. The cell containing the crystal was mounted on a rotating shaft which varied the incident angle θ of the light on the crystal. Large angles $\theta = 55.5°$ were used in order to make the angular measurements more accurate. We note that the experiments were carried out for laser power densities $< 10^4$ W/cm^2 low enough so that the nonlinear changes in the dielectric function of the medium near exciton resonance were negligible.

3. RESULTS AND DISCUSSION

We observed a single bright spot corresponding to the + wave (cf. Refs. 1, 5 for the definition of the + and − waves) when the laser was tuned to $\omega < \omega_L$, where $\omega_L = 2.5541$ eV is the longitudinal exciton frequency; this spot was observed in the focal plane of lens L_2 for the same

 0038-5646/85/070185-03$04.00

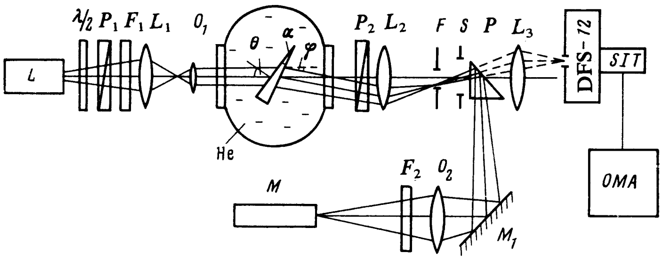

FIG. 1. Sketch of the experimental system.

polarization $\mathbf{E}\perp\mathbf{C}_6$ used throughout the experiment. The spot became less intense and moved farther away from the center of the microscope viewing field as ω approached ω_L. A second spot, corresponding to the $-$ wave, appeared in the focal plane starting at $\omega = \omega_L$. Its brightness increased rapidly with ω, but the spot displacement leveled off (cf. Fig. 2). Both spots were observed together for frequencies 2.5541 eV $<\omega<$ 2.5575 eV. Our measurements showed that their frequencies and polarizations were identical. When the laser frequency ω increased further, the $+$ wave became too weak to be distinguished from the scattered background light. The maximum measurable distance between the $+$ and $-$ spots was $\approx 740\,\mu$m.

The spot displacement in the focal plane is uniquely related to the index of refraction of the corresponding wave. For small α, this relation is given by the formula[4]

$$\varphi=\alpha\left\{\frac{\operatorname{Re}[(n^2-\sin^2\theta)^{1/2}]}{\cos\theta}-1\right\}. \tag{1}$$

We substituted the experimentally measured displacement in Eq. (1) to calculate the refractive index (Fig 3) for the $+$ and $-$ waves [the imaginary part of n can be neglected in (1)].

It should be noted that more careful observations for $\omega<\omega_L$ revealed a third, much fainter spot whose deflection angle φ was twice that for the first spot corresponding to the

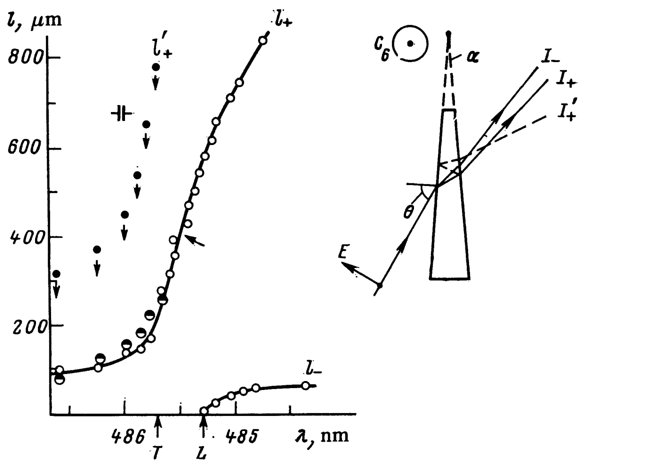

FIG. 2. Displacement of the spots in the focal plane F as a function of the wavelength of the light incident on the crystal. The dark circles ● show the displacement of the spot corresponding to threefold transmission of the $+$ wave through the wedge (cf. I'_+ in the diagram to the right). The half-open circles give the values of l'_+ divided by 3. T and L indicate the position of the transverse and longitudinal excitons.

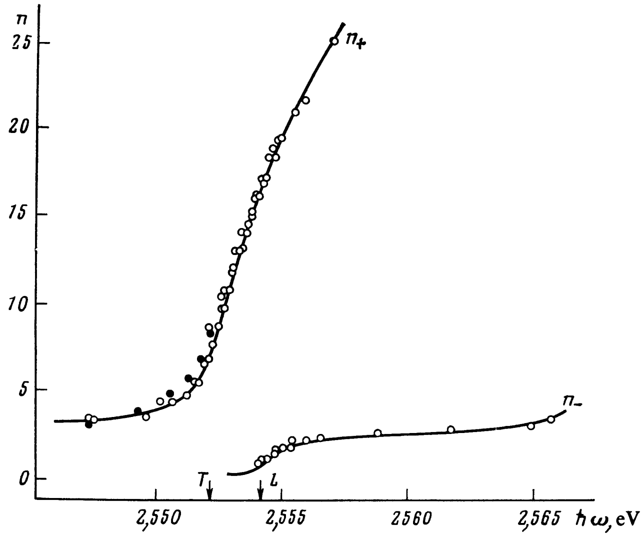

FIG. 3. Dispersion of the refractive indices n_+ and n_- near the $A_{n=1}$ exciton resonance in CdS. The theoretical curves were calculated by Pekar's formulas. The closed circles were found from observations of threefold transmission of the $+$ wave through the wedge.

$+$ wave. This weak spot was caused by threefold transmission of the $+$ wave through the wedge due to reflections from the sides (I'_+ in Fig. 2); it moved rapidly out of the viewing field and disappeared as ω increased. Figure 2 shows the spot displacement l in the focal plane F as a function of the wavelength. The displacements were measured from the point in the focal plane at which the transmitted laser light was focused (the spot could be observed only when the slits S were wide-open). The dark circles in Fig. 2 show the displacement of the spot corresponding to threefold transmission of the $+$ wave; if these displacements are divided by 3, they agree with the corresponding displacement for the singly transmitted light.

Figure 3 shows the measured refractive indices n_+ and n_- and the theoretical curves calculated by Pekar's formulas[1,5]:

$$n_\pm{}^2={}^1/_2(\mu+\varepsilon_0{}^*)\pm[{}^1/_4(\mu-\varepsilon_0{}^*)^2+b]^{1/2},$$

where

$$\mu=\mu'+i\mu'',\quad \mu'=(2Mc^2/\hbar\omega_0{}^2)(\omega-\omega_0),\quad \varepsilon_0{}^*=\varepsilon_0+\delta\varepsilon,$$
$$\mu''=(2Mc^2/\hbar\omega_0{}^2)\Gamma,\quad b=(2Mc^2/\hbar\omega_0{}^2)\varepsilon_0\Delta_{LT},$$

ε_0 is the phonon dielectric permittivity, $\delta\varepsilon$ is the correction to ε_0 from the higher-lying exciton resonances,[3] ω_0 is the resonance frequency for the transverse $A_{n=1}$ exciton, c is the speed of light in vacuum, M is the translational exciton mass, Δ_{LT} is longitudinal-transverse splitting, and Γ is the damping constant.

The agreement between theory and experiment is quite satisfactory. The calculations assumed the following parameter values: $\varepsilon_0 = 7.4$; $\omega_0 = 2.55205$ eV; $\Delta_{LT} = 2.1$ meV; $M = 0.8m_e$; $\Gamma = 0.02$ meV. The longitudinal exciton frequecy $\omega_L = 2.5541$ eV was deduced from the transmission spectra for the polarization $\mathbf{E}\|\mathbf{C}_6$.

We note that there is some discrepancy between the experimental and theoretical results—specifically, the dispersion curve recorded for n_+ has a small bump at $\hbar\omega = 2.5530$

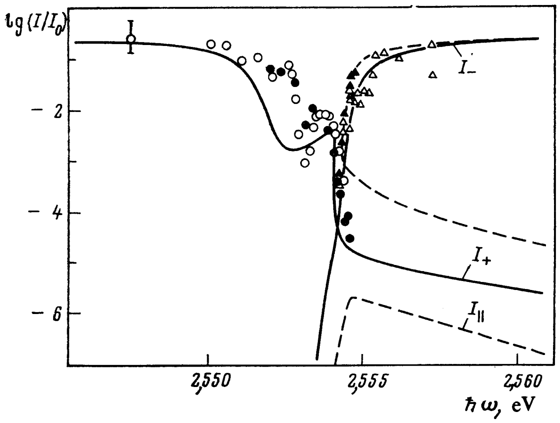

FIG. 4. Transmission of the $+$ and $-$ waves (circles and triangles) near the $A_{n=1}$ exciton resonance in CdS. The curves were calculated by the formulas in Ref. 4. The solid curve shows values calculated for two damping constants $\Gamma_1 = 0.08$ meV and $\Gamma_2 = 0.2$ meV; the dashed curves were calculated for $\Gamma_1 = \Gamma_2 = 0.08$ meV. The dark and light points correspond to two different series of measurements.

eV ($\lambda = 485.5$ nm) which is not present in the calculated curves. Although the height of the bump is comparable to the experimental error, the bump was faithfully reproduced in each series of measurements (cf. Fig. 2). A similar bump in n_+ was also noted in Ref. 6, where the interference of the additional waves was observed. It was attributed there to the absorption line of the triplet $A^{\perp}_{\Gamma_6}$ exciton.

Returning to the transmission spectra of the $+$ and $-$ waves, we note that because of difficulties in distinguishing the weak spot from the scattered background light, the transmissions could be measured simultaneously only in a subinterval of the wavelength interval for which the deflection angles could be measured and the transmissions observed. When the $+$ spot became very faint, its contribution to the transmitted signal became comparable to the contribution from the scattered light, so that the error in the intensity measurements increased abruptly; nevertheless, the location of the $+$ spot could still be determined accurately.

Figure 4 presents the data on the transmission of the $+$ and $-$ waves. The transmission of the $+$ wave drops as ω approaches ω_L. For the frequencies $\omega > \omega_L$ for which transmission of both the $+$ and $-$ waves can be observed, the $+$ wave transmission drops fastest, while the transmission of the $-$ wave rises abruptly. This behavior is predicted directly by the ALW theory.[5] The theoretical curves, calculated using Pekar's additional boundary conditions,[7] are shown in Fig. 4. The exciton damping constant Γ is the only adjustable parameter in these calculations—the remaining parameters were either determined in independent experiments or else were measured directly.

Figure 4 shows that the theoretical curve agrees best with the experimental curve for the $-$ wave. There is large scatter in the experimental points for the $+$ wave for $\omega \approx \omega_L$. It was shown in Ref. 8 that for these frequencies the experimental results for CdS (unlike those for CdSe) are hard to reproduce—the fluctuations vary randomly from one experiment to the next.

For $\omega > \omega_L$ the theory predicts a much slower decay with frequency than is found experimentally. Since there is much evidence that Γ is not actually constant but may increase with ω (see, e.g., Ref. 8), it seems natural to correct the theory by letting Γ take two values:

$$\Gamma = \begin{cases} \Gamma_1, & \omega < \omega_L \\ \Gamma_2, & \omega > \omega_L \end{cases}.$$

The solid curve in Fig. 4 shows the calculated results found for $\Gamma_1 = 0.08$ meV and $\Gamma_2 = 0.2$ meV. We see that this approximation considerably improves the agreement between theory and experiment.

The dashed curve at the bottom of Fig. 4 for $\omega > \omega_L$ shows $I_{\parallel}$, the theoretically calculated intensity of the wave emerging the wedge when the light in the wedge is longitudinally polarized. Even for $\Gamma = 0.08$ meV this intensity is too weak to be recorded experimentally.

Our results thus demonstrate convincingly that an additional light wave is present in the CdS crystal, and they can be used to calculate the optical constants and exciton parameters. The agreement between theory and experiment also suggests that the additional boundary conditions employed in the calculations are correct.

We thank I. N. Ural'tsev for kindly supplying the CdS crystal wedge.

[1] S. I. Pekar, Zh. Eksp. Theor. Fiz. **34**, 1176 (1958) [Sov. Phys. JETP **7**, 813 (1958)].

[2] I. Broser, R. Broser, E. Beckmann, and E. Birkicht, Sol. St. Commun. **39**, 11, 1209 (1981).

[3] M. V. Lebedev, M. I. Strashnikova, V. B. Timofeev, and V. V. Chernyĭ, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 366 (1984) [JETP Lett. **39**, 440 (1984)].

[4] A. A. Demidenko, S. I. Pekar, and B. E. Tsekvava, Fiz. Tverd. Tela (Leningrad) **27**, 741 (1985) [Sov. Phys. Solid State **27**, 457 (1985)].

[5] S. I. Pekar, Kristalloptika i Dobavochnye Svetovye Volny (Crystal Optics and Additional Light Waves), Naukova Dumka, Kiev (1982).

[6] V. A. Kiselev, B. S. Razbirin, and I. N. Uraltsev, Phys. St. Sol. **b72**, 161 (1975).

[7] S. I. Pekar, Zh. Eksp. Teor. Fiz. **33**, 1022 (1957) [Sov. Phys. JETP **6**, 785 (1957)].

[8] I. V. Makarenko, I. N. Uraltzev, and V. A. Kiselev, Phys. St. Sol. **b98**, 773 (1980).

Translated by A. Mason