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NONLINEAR FARADAY EFFECT IN CdS SEMICONDUCTOR IN AN
ULTRAHIGH MAGNETIC FIELD

V.V. Druzhinin, O.M. Tatsenko, A.I. Bykov, M.I. Dolotenko, N.P. Kolokol'chikov, Yu.B. Kudasov, V.V. Platonov, C.M. Fowler, B.L. Freeman, J.D. Goettee, J.C. King, W. Lewis, B.R. Marshall, B.J. Papatheofanis, P.J. Rodriguez, L.R. Veaser, and W.D. Zerwekh

A significant nonlinearity in the angle of rotation of the polarization plane was observed in CdS at wavelengths of 494-633 nm in the presence of high magnetic fields (0.5-5 MG). The onset of significant nonlinearity also depended on sample temperature. An absorption study with probe wavelength of ~ 494 nm revealed an increase in optical transmission associated with a splitting of the conduction band. Dispersion, field and temperature curves indicate a low conduction electron mass $m_c = 0.3 m_0$. A numerical calculation and interpretation of the observed effects was carried out using band theory.

The optical and magnetooptical properties of semiconducting crystals of CdS were studied, reviews of which are presented in [1,2].

This article describes joint American-Russian experiments to study the optical and magnetooptical properties of CdS in ultrahigh magnetic fields to ~ 7 MG.

The study of CdS semiconductor was carried out in two kinds of high field systems: the MC-1 generator at temperatures of 100 and 300 K at various wavelengths: 441, 495, 544 and 633 nm, and as is discussed later, in a strip generator, at temperatures of 6.5 K over a continuum of wavelengths. The CdS sample had two parallel, polished faces perpendicularly oriented to the C-axis which was, in turn, parallel to the magnetic field, a thickness of $h = 0.93$ mm, and a transmission spectrum with an absorption band to 520 nm (300 K) and 516 nm (100 K).

The optical and magnetooptical properties of CdS semiconductors have a host of interesting characteristics, among which the following can be identified:

- 1) a large refractive index $n = 2.4$ in the red spectral region and 3-4 in the region near the absorption edge (blue-green);
- 2) the manifestation of nonlinearity in the Faraday effect with megaGauss fields, which increases with shorter wavelengths and has an upward curving character. In comparison, the majority of other compounds which manifest nonlinearity, exhibit a downward curving slope;
- 3) an anomalously high positive Verdet coefficient $V = 0.5-2$

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min/cm G in the 500-630 nm spectral region;

4) a shift in the edge of the intrinsic absorption band under the effect of a magnetic field toward shorter wavelengths.

All these characteristics were experimentally observed in magnetic fields to 2 MG [1,2]. The results of the joint experiments corroborate the observed effects in magnetic fields to 7 MG. Thus, at probe wavelengths of 544 nm and $T = 100$ K, the nonlinearity of the Faraday effect starts to show up in magnetic fields of 1 MG, and at a wavelength of 633 nm and $T = 300$ K, in fields of 5 MG. Simultaneously with the Faraday effect, the transmission of light through a CdS sample was measured at a probe wavelength of 494 nm ($\Delta\lambda = \pm 6$ nm). With the zero field absorption edge at 510 nm, initially no light was transmitted. It was noted, however, that for a magnetic field of ~ 7.3 MG, a transmitted signal was observed through the CdS sample, which indicated a shift of the absorption band edge to the blue spectral region.

The above mentioned characteristics of the optical and magneto-optical properties of CdS have been experimentally observed for a relatively long time but have not, to date, been adequately explained.

This study proposes a microscopic-phenomenological theory which qualitatively and quantitatively explains these characteristics. The fundamental mechanism for interpreting the optical properties of CdS is based on the calculation of two contributions to the high frequency dielectric susceptibility which are due to a diamagnetic transition into the conduction band and a paramagnetic transition between the valence band and the bottom of the conduction band.

The general formula which describes the dispersion of the refractive index in the absence of a magnetic field has the form:

$$n = \sqrt{\frac{3 + 8\pi N\beta}{3 - 4\pi N\beta}} \quad (1)$$

where N is the molecular concentration of CdS per unit volume; β is the polarization tensor which is written in the form:

$$\beta = \sum_{m=1}^n \frac{2e^2 R_{KM} \omega_{KM}}{\hbar (\omega_{KM}^2 - \omega^2)} \quad (2)$$

Here R_{KM} is the matrix element between the ground ψ_K and excited ψ_M states; e is the electron charge; ω is the probe frequency and ω_{KM} is the transition frequency.

For the above-mentioned reasons, the summation in Eq. (2) is divided into two basic components having the transition frequencies:

ω_{01} - an indirect transition into the conduction band, and
 ω_{02} - an interband transition, that mainly determines the optical properties of CdS. The existence of these transitions verify and corroborate the structure of the energy band in CdS semiconductors, which is shown in Fig. 1 [3].

Fig. 1
 Structure of the energy bands in CdS semiconductors [3]. The frequency $\omega_{01} = 6.64 \times 10^{15}$ rad/sec (4.37 eV) corresponds to the transition $\Gamma_{15} \rightarrow X_1$; $\omega_{02} = 4.05 \times 10^{15}$ rad/sec (2.66 eV) to the transition $\Gamma_{15} \rightarrow X_3$. $T = 100$ K.

Thus the value for $4\pi N\beta$ in eqs. (1 and 2) can be written in the form:

$$4\pi N\beta = \frac{C_1}{\omega_{01}^2 - \omega^2} + \frac{C_2}{\omega_{02}^2 - \omega^2} \quad (3)$$

where C_1 and C_2 are the constants of Eq. (2) and are equal to ($i = 1, 2$).

$$\frac{4\pi N \beta 2e^2 R_{oi} \omega_{oi}}{\hbar}$$

Substituting the known values $N = N_A \rho / M = 2 \times 10^{22} \text{ cm}^{-3}$, $R_{oi} = 2.9 \times 10^{-8} \text{ cm}$ we find $C_1 = 61 \times 10^{30} \text{ sec}^{-2}$; $C_2 = 0.4 \times 10^{30} \text{ sec}^{-2}$. With these values from Eq. 3 we obtain the following results: far from the absorption edge, the contribution determined in Eq. (3) which corresponds to the dispersion curve for the refractive index (eqs. (1 and 2)) is dominated by the first component due to the large value of C_1 . In the region near ω_{02} (the edge of the intrinsic absorption band) the second component also becomes significant. Figure 2 shows the dispersion curve $n(\omega)$ calculated using Eq.s (1-3).

As is shown in Fig. 2, the calculated curves are in good agreement with the experimental values, which argues for the validity of the selected model.

The presence of an interband (paramagnetic) contribution to

Fig. 2
The calculated (dashed lines) dispersion $n(\omega)$. $\blacktriangle$ - experimental values [1], n_1 - diamagnetic, n_2 - paramagnetic components (first and second contributions in Eq. (2)).

the high frequency susceptibility explains not only the strong increase in n in the region near the intrinsic absorption edge ($\omega_{02} \sim 4 \times 10^{15}$ rad/sec), but also the characteristics of the Faraday effect, specifically the dispersion of the Verdet coefficient and nonlinearity of the Faraday effect.

As a result of the dispersion of the refractive index, it is possible to predict a Faraday effect composed of two basic contributions. This means that the Verdet coefficient for CdS has the following form:

$$V = V_d + V_n \quad (4)$$

Here, V_d is the diamagnetic (positive) component, resulting from the transition $\Gamma_{15} \rightarrow X_1$ transition with a frequency of $\omega_{01} \approx 6.64 \times 10^{15}$ rad/sec, which does not depend on the magnetic field, and V_n is the paramagnetic (negative) component, resulting from the interband transition $\Gamma_{15} \rightarrow \Gamma_1$ with frequency $\omega_{02} = \omega_{g0} = 4 \times 10^{15}$.

As known earlier from experimental data, the edge of the absorption band shifts in a magnetic field toward shorter wavelengths. It is possible to predict the frequency of this transition as it shifts in a magnetic field from:

$$\omega_g = \omega_{g0} + \omega_c/2 \quad (5)$$

where $\omega_c = \frac{eH}{m^*c}$ is the cyclotron frequency and m^* is the effective mass determined from the extent of the band splitting of the Landau levels.

The dispersion curves for the diamagnetic and paramagnetic

components differ. Based on classical predictions, they may be written in the form: $V_g = \text{const} \cdot F_n(\omega) \cdot f_1(\omega)$ where $F_n(\omega) = \frac{1}{n} \left(\frac{n^2+2}{3} \right)^2$ is a factor giving the functional dependence of V_g on the radiation frequency through the refractive index, and $f_1(\omega) = \omega_0 \cdot \omega^2 / (\omega_0^2 - \omega^2)^2$ is the characteristic function for the diamagnetic Faraday effect.

As for the paramagnetic component,

$$V_n = -\text{const}' \cdot F_n(\omega) \cdot f_2(\omega) \quad \text{where} \quad f_2(\omega) = \frac{\omega^2}{(\omega_0^2 - \omega^2)} \quad (6)$$

Thus, the Verdet coefficient clearly shows a nonlinear functional dependence which is shown in Fig. 3

Fig. 3.

Dispersion curve for the Verdet coefficient in CdS in weak magnetic fields. ▲ are experimental data [1], the dashed lines are calculated; V_g, V_n are the respective diamagnetic and paramagnetic components

From Fig. 3 it is apparent that the model used and experimental data agree quantitatively. The resulting paramagnetic component can be associated to a large degree with both the interband transition and exciton states [4]. While a definitive understanding of its characteristics requires additional data, it can still be said that the presence of a minus sign indicates significant splitting of the ground state.

In addition to the dispersion curve for the Verdet coefficient, we can show from Eq. (6) that there is also a functional dependence of the Verdet coefficient on magnetic field, specifically a functional dependence of the paramagnetic component. Let us consider this dependence, the resulting nonlinearity of the Faraday effect, its magnitude and shape. The Verdet coefficient increases with a growth in the magnetic field. While the dia-

magnetic component V_g does not depend on the magnetic field, the paramagnetic contribution V_n does, through the field dependency of the bandgap edge ω_g . From (6) we obtain this change ΔV :

$$\Delta V = \Delta V_n = A_n F_n(\omega) \frac{2\omega^2 \omega_{g0}}{(\omega_{g0}^2 - \omega^2)^2} \Delta \omega_g \quad (7)$$

where $\Delta \omega_g = \frac{eH}{2m^*m_0c}$ determines the shift of the lower Landau sublevel toward shorter wavelengths under the effect of a magnetic field.

From Eq. (7) it follows that ΔV is nonlinearly dependent on magnetic field, and when $\Delta V > 0$, this dependence curves upwards. The nonlinearity of the Faraday effect is due to an increase in the forbidden band $\Delta \omega_g$ in high magnetic fields and is associated with a decrease in $|V_n|$; which means that with an increase in probe radiation frequency, the Faraday effect increases and can be observed in increasingly weaker fields.

Figures 4 and 5 show experimental data and curves calculated from Eq. (7) and exhibit good agreement between the results.

A few additional calculations were carried out. According to the experimental results of [1,2] for a field of 1.9 MG, $\Delta \lambda_g = 63 \text{ \AA}$, or $\Delta \omega_g = 0.035 \text{ eV}$ ($0.536 \times 10^{15} \text{ rad/sec}$) which means that the effective mass m^* , expressed in electron mass units is equal to:

$$m^* = \left(\frac{2\Delta \omega_g m_0 c}{eH} \right)^{-1} = 0.312$$

Fig. 4.
Dependence of the angle of rotation of the polarization θ on magnetic field H for various wavelengths in semiconducting CdS [1,2]. The crystal was 1.15 mm thick. The dashed lines indicate linear dependence
• - calculated, Δ - experimental

Fig. 5.
Dependence of $\Theta = f(H)$ for CdS from the joint American-Russian experiments. Crystal thickness was 0.93 mm.

This result may be compared with the experimental values [2]:

$$m_p = (0.3 - 0.35) m_0, \quad m_e \approx 0.2 m_0$$

The growth in the Verdet coefficient ΔV was calculated from Eq. (11) based on the dependence of the Faraday effect on magnetic field for a probe wavelength of 517 nm in a magnetic field of 1.2 MG (Fig. 4). For $\Delta\omega_0 = 3.38 \times 10^{13}$ rad/sec, $H = 1.2$ MG, and $\omega = 3.64 \times 10^{15}$ rad/sec; $\Delta V = 0.1$ min/cm G, which is in good agreement with the experiment.

Thus, in an external ultrahigh magnetic field (with a strongly nonlinear Faraday effect), interband paramagnetic component arises which has a significant effect on the value of the refractive index, its dispersion, and on the value of the Verdet coefficient, its dispersion and its dependence on field. The value of the paramagnetic component, itself, depends on exciton states, the precise identification of which requires further experimentation.

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