MAGNETO-OPTICAL STUDIES OF ANOMALOUS ANISOTROPY OF THE 
Cd$_{1-x}$Mn$_x$Te CRYSTAL

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By using the magneto-optical and transmission-ellipsometric methods, giant anisotropy in both optical and magneto-optical properties was found for the single crystal Cd$_{1-x}$Mn$_x$Te (x=0.45). Two orthogonal axes, $E_\perp$ and $E_\parallel$, were in the (113) plane, and along the [471] and [211] directions, respectively. Both transmission-ellipsometric and magneto-optic Faraday rotation spectra located peaks at the same about 2-eV $E_{\text{MN}}$ position, that was in the gap between the conduction and valence bands. In discussion, $E_{\text{MN}}$ was attributed to the optical transition of Te $p \to$ Mn $d_5$, and several possible mechanisms for the anisotropy phenomena were suggested and studied. A general and analytical equation was given to calculate the anomalous anisotropy of the Faraday effect, showing good agreement with experimental results. The equation can be used to explain other magneto-optical anisotropic properties of the system.

KEYWORDS: Optics, magneto-optics, ellipsometry, Faraday effect, anisotropy

I. Introduction

Among the Mn-doped II-VI magnetic semiconductor groups, Cd$_{1-x}$Mn$_x$Te is probably the one that has been studied most extensively. It can be crystallized in a single-phase zinc-blende (ZB) structure up to a composition of x=0.45.[1] The free Mn atom has a 3d$^5$4s$^2$ configuration; hence, after doping the divalent Mn can replace Cd at the lattice sites. The behavior of the 3d$^5$ electrons of Mn$^{2+}$ with their spin parallel (up) by Hund’s rule plays a significant role, resulting in such a property as the giant Faraday rotation observed even at room temperature with a moderated magnetic field.[2] The pinning of the lowest excited energy level at ~ 2 eV above the $\Gamma_8$ valence band as x ≥ 0.4 is another interesting phenomenon[3,4], and is now attributed more likely to a Mn$^{2+}$ 3d$^5$:intraband transition of 6$A_1 \to 4T_4$ with a spin flip in the band.[5] This implies that the 3d$^5$ ground state is close to the $\Gamma_8$ maximum, which is also indicated by band structure calculations of Cd$_{1-x}$Mn$_x$Te.[6] Photoemission results,[7,8] however, located the 3d$^5$ band lying ~3.5 eV below $\Gamma_8$. The experiment results seem to be undisputable, leaving only the interpretation in controversy. So further effort should be made to understand the role of Mn$^{2+}$ better. It is usually thought that the Cd cations, which are arranged in an fcc sub-lattice of ZB structure, are randomly replaced by Mn$^{2+}$.[1] The Cd$_{1-x}$Mn$_x$Te crystal has a high-temperature paramagnetic phase;[9] therefore, at room temperature and zero field, the crystal should be macro-isotropic. Recently, however, as we studied the magneto-optical properties of Cd$_{1-x}$Mn$_x$Te with x=0.45, we observed a giant optical and magneto-optical anisotropy existed in the crystal. We described the phenomenon in this paper.

II. Experiments

Single crystal Cd$_{1-x}$Mn$_x$Te with x= 0.45, showing a uniform reddish color, was prepared by the modified Bridgman method without special heat treatment. The sample was cut to a thickness of about 1.8 mm from the ingot section, and was polished and etched on both sides. The crystal is however brittle and can be cleft along some orientations. The x-ray diffraction data indicated that the polished and the side-cleft planes have the {113} and {110} index, respectively, as shown in Fig. 1. The spectra of the dielectric function measured by ellipsometry were in agreement with other results[10]. The fundamental absorption gap $E_g$ is at about 2.125 eV which corresponds to the Mn composition of x=0.45 as was confirmed by the X-ray data.

A rotating analyzer type of Kerr apparatus[11] was used to measure absolutely the spectra of the doubled pole Faraday rotation $\theta_F$ at room temperature by coating an Al film on the back side of the sample. The incident angle was about 2&. The linear-polarization direction s of the incident beam was fixed and was perpendicular to the plane of incidence. We found that not only the hysteresis loop, which had zero coercivity, but also the Faraday spectra were strongly dependent on crystal orientation. At zero magnetic field, the pure cosine raw light signal was phase-shifted by a large value.
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Fig. 1. The orthogonal $E_\perp$ and $E_\parallel$ orientations, are in the (113) plane, and along the [4 7 1] and [21 1] direction, respectively. [113] is normal to the plane of the paper.

$\phi_o$ for some orientations. This would not be the situation for a sample in a paramagnetic state, for example a glass plate. This meant that a giant zero-field rotation existed in the sample. As the field $H$ was scanned from 0 to 10 kOe in the two opposite directions, $\phi_o$ did not vary linearly with $H$, and was dependent on both the photon energy $E$ and sample orientation.

To understand this phenomena, we then used the angle-variable scanning ellipsometer[12] to measure the polarization states of transmittance by setting a 90° incidence angle with the incident beam being normal to the sample surface. In the system configuration, the azimuthal angle of the polarizer $P_o$ (in the s direction) was fixed and perpendicular to the incident plane to eliminate the residue polarization effect of the light source. The azimuthal angles $P$ and $A$ are those of the the polarizer and analyzer, respectively, and were initially zero with respect to the s axis. The polarizer and analyzer were synchronously rotated at different rate of $A=p=\omega_0\frac{\tau}{l}$, controlled by two micro-stepping motors having 10000 steps per revolution. If the sample was transparent and optically isotropic, inserting the sample would give the same result. If the sample is not optically isotropic, however, linearly polarized light will emerge from the sample in an elliptically polarized state. This is the case with Cd$_{1-x}$Mn$_x$Te ($x=0.45$) as can be seen in Fig. 2 where the results for the sample and air are compared. The sample has two special orientations, $E_\perp$ and $E_\parallel$, lying in the (113) plane and perpendicular to each other, as indicated in Fig. 1. The results show clearly that Cd$_{1-x}$Mn$_x$Te ($x=0.45$) is optically anisotropic.

Next, we scanned the photon energy in the 1.5-2.3 eV range with a 0.01-eV interval to determine $\Psi$ and $\Delta$ by setting $E_\perp$ and $E_\parallel$ be along the s axis, respectively. The spectra of $\Psi$ and $\Delta$ are shown in Fig. 3. The two curves for $\Psi_\perp$ and $\Psi_\parallel$ are nearly mirror-reflected by the line of $\Psi=45^\circ$. Since $\cos\Delta$ is an even function, there is a sign uncertainty in $\Delta$, so we give the $\Delta$ spectra without sign in the figure to show that the magnitudes of $\Delta_\perp$ and $\Delta_\parallel$ are consistent. The large values of $\Delta$ ($> 100^\circ$ in zero order) implies that the crystal has different dispersion features, i.e., there are two refractive indexes, $n_\perp$ and $n_\parallel$ corresponding to the $E_\perp$ and $E_\parallel$ orientation, respectively. The sharp peaks for both $\Psi$ and $\Delta$ spectra are located exactly at 2.07 eV.

Fig. 2. The raw data were measured by a transmittance ellipsometer at 2 eV for air, and the sample as $E_\perp$'s and rotating 45° from the $E_\parallel$ direction, respectively, and showed clearly the anisotropy.

Once the orientations of the crystal were determined, we measured the Faraday data again. We found that when either the $E_\perp$ or $E_\parallel$ orientation was parallel to the s direction, the zero-field phase-shift $\phi_o$ was nearly zero, and $\theta_F_\perp$ and $\theta_F_\parallel$ were small. When the sample was rotated about 45°, $\phi_o$ and $\theta_F_{45^\circ}$ were both maximum. $\theta_F_{45^\circ}$ was about 10 times larger than $\theta_F_{E_\perp}$. The
spectra of $\theta_F^{45^\circ}$ measured under different magnetic fields were shown in Fig. 4. The hysteresis loops of $\theta_F^{\perp}$, $\theta_F^H$ and $\theta_F^{45^\circ}$ were anti-symmetric and were not exactly linear to the magnetic field $H$ as shown in top insert of Fig. 4. In other crystal orientations, the loops were not anti-symmetric to $H$. Comparing the 2.07-eV peaks of $\Psi$ and $\Delta$ at zero field in Fig. 3, the peaks of the $\theta_F$ spectra were located around 2 eV, and were red-shifted with increasing field. We attribute this structure, say $E_{Mn}$, to the same origin in top insert of Fig. 4. In other crystal orientations, the loops were not.

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The two normal modes of light propagating along the $z$ direction is given: [19]

$$ R_+ = \begin{bmatrix} 0 & \rho_0 & \sin \alpha \\ i \rho_0 & 0 & -i \cos \alpha \\ \rho_0 & i (\delta_0 + \Delta_0) & \cos \alpha \end{bmatrix} \begin{bmatrix} \sin \alpha \\ -i \cos \alpha \\ \cos \alpha \end{bmatrix} ,$$

and their refractive index $n_{\pm}$ are

$$ n_{\pm}^2 = \frac{\epsilon_{11} + \epsilon_{33}}{2} \pm \Delta_0. $$
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where

$$\delta_0 = \frac{\epsilon_{11} - \epsilon_{22}}{2}, \quad \rho_0 = \epsilon_{12},$$

$$\Delta_0 = \left(\delta_0^2 + \rho_0^2\right)^{1/2}, \quad \tan \delta_0 = \frac{\delta_0 + \Delta_0}{\rho_0}.$$  (4)

Therefore, in terms of Eqs. (1)−(4), we found that for a general case the experimentally measured Faraday rotation $\psi$ for a magneto-optical anisotropy system will be different from ordinary one, and can be given:

$$\tan 2\psi = \frac{\sin 20 \cos \delta - \cos 20 \sin 2\alpha \sin \delta}{\cos 20(\cos 2\alpha + \sin 2\alpha \cos \delta) + \sin 20 \sin 2\alpha \sin \delta},$$  (5)

where, $\theta$ is the azimuthal angle of the linearly-polarized incident light, $\alpha$ and $\delta$ are two parameters related to the optical and magneto-optical anisotropy of the material structure. For an isotropic system, $\alpha = 45^\circ$ and $\delta = 2\theta_F$, where $\theta_F$ is the true Faraday rotation angle defined in the literature. By Eq. (5), we have calculated the Faraday rotation by fitting the two parameters and obtained the results in good agreement with experimental data which was shown in Fig. 5, and will be discussed in more detail elsewhere.

In conclusion, by using the magneto-optical and transmission ellipsometric methods, the giant optical and magneto-optical anisotropy of single crystal Cd$_{1-x}$Mn$_x$Te ($x=0.45$) were shown. Both ellipsometric and magneto-optic Faraday rotation spectra located the peaks at the same $E_{Ma}$ position, that was clearly in the gap between the conduction and valence bands and was attributed to the optical transition of Te p $\rightarrow$ Mn d$_1$. A general and analytical equation was given to explain other magneto-optical anisotropic properties of the system. A better understanding of the anisotropy of Cd$_{1-x}$Mn$_x$Te may reveal further how Mn$^{2+}$ influences the properties of the crystal.

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References