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Masaaki Imamura* | Daisuke Tashima | Jiro Kitagawa | Hironori Asada

Abstract—The II-VI based magnetic semiconductors with a direct and wide optical bandgap are expected to show high potential for optical applications utilizing short wavelength laser diodes (LDs), such as 532-nm green and 475-nm blue LDs. We have confirmed that the Faraday rotation \( \theta_F \) in the ZnMnTe and ZnMnSe films deposited on quartz glass (QG) and sapphire (SA) substrates by using molecular beam epitaxy (MBE) is large near the absorption edge. This paper reports the magneto-optical properties of ZnMnTe and ZnMnSe films synthesized on the QG and SA substrates, and shows the result of a direct Faraday rotation observation successfully made for the ZnMnTe films under 1.28-kHz alternating magnetic fields. The optical absorption characteristics of the ZnMnTe films grown on the SA substrates by MBE are discussed by comparing them with the optical absorption properties and photoluminescence spectra of the II-VI ZnTe parent single crystals.

Index Terms—Magnetic semiconductors, magneto-optical effects, wide bandgap, ZnMnTe and ZnMnSe films.

1. Introduction

The II-VI semiconductors ZnTe crystals which do not have transition metal ions, such as Mn, have actively been studied to detect terahertz electric fields\(^1\)\(^\text{[1-3]}\). Another area of intensive studies on II-VI semiconductors is the field of quantum engineering which includes heterojunctions\(^6\)\(^\text{[4,5]}\), quantum wells (2D structure)\(^7\)\(^\text{[6,7]}\), quantum wires (1D), and quantum dots (0D)\(^8\)\(^\text{[8,9]}\). On the other hand, the II-VI based magnetic semiconductors of the \( \text{A}^{1-x}_{-1} \cdot \text{Mn}_x \cdot \text{B}^x \) composition with a direct and wide optical bandgap\(^9\)\(^\text{[9,10]}\) show the promise for magneto-optical applications utilizing short wavelength laser diodes (LDs), such as a 532-nm green or 475-nm blue beam. The wavelengths of LDs used for high-density optical recordings have become shorter. The required optical setups need an isolator that works effectively for shortwave LDs to eliminate back reflections of the laser beam. In addition to the optical isolators, the development of magneto-optical sensors that work on a short optical wavelength has been expected\(^12\)\(^\text{[12,13]}\). The II-VI magnetic semiconductors \( \text{Zn}_{1-x} \cdot \text{Mn}_x \cdot \text{Te} \) and \( \text{Zn}_{1-x} \cdot \text{Mn}_x \cdot \text{Se} \) have the absorption edge at 428 nm...
to 544 nm and 428 nm to 458 nm, respectively. When increasing the Mn concentration $x$, the shift of the absorption edge to the higher energy gap for these materials is small compared with the prototype II-VI magnetic semiconductor Cd$_{1-x}$Mn$_x$Te. It will be discussed in subsection 3.1. The Faraday rotation angle $\theta_F$ for the ZnMnTe films deposited on quartz glass (QG) and sapphire (SA) substrates is large near the absorption edge[15],[16]. Zn$_{1-x}$Mn$_x$Te forms a ternary alloy of a zinc-blende structure with $x$ up to 0.86, and Zn$_{1-x}$Mn$_x$Se exhibits a zinc-blende structure for $x<0.30$ and a wurtzite structure for 0.30<$x$<0.55[17]. The band structure when replacing the atoms of the group II element by Mn in the direct-gap zinc-blende "parent" ZnTe or ZnSe semiconductor is a zinc-blende open gap near the $\Gamma$ point as shown in Fig. 1. The presence of localized magnetic ions in these semiconductors leads to an exchange interaction between the sp band electrons and the d electrons associated with Mn$^{++}$, resulting in the Zeeman splitting. The spin splitting of the top of the valence band ($\Gamma_8$) and the bottom of the conduction band ($\Gamma_6$) in a magnetic field is schematically shown in the right side of Fig. 1. This spin-dependent phenomenon leads to the Faraday effect due to the difference in the optical absorption or reflection for the left and right circular polarized light caused by the Zeeman splitting under the magnetic field.

The specific values of the Faraday rotation in the room temperature region have not been shown for the ZnMnTe and ZnMnSe films synthesized on the QG and SA substrates by molecular beam epitaxy (MBE) so far. This fact motivated us to carry out the materials research on II-VI magnetic semiconductors. The presented paper reports the magneto-optical characteristics of those films, and describes the results of the Faraday rotations for the ZnMnTe films subject to alternating magnetic fields from a practical viewpoint of their applications.

2. Experimental Procedure

2.1. Film Preparation and Measuring Equipment

Magnetic semiconductor films were synthesized on the transparent QG and SA ((0001) orientation C-cut $\alpha$-Al$_2$O$_3$) substrates heated to 300 °C by using MBE. Deposition was carried out at an average rate of 1.0 Å/s to 1.5 Å/s for 6 h to 8 h using a 5N ZnTe effusion cell heated to 465 °C to 475 °C and a 4N MnTe cell at 1035 °C to 1040 °C. Using this system, 2-mm-thick to 3-mm-thick ZnMnTe films with the Mn mole-fraction $x$ from 0.18 to 0.27 were prepared on the 0.5-mm-thick QG and SA substrates. The ZnMnSe films were prepared similarly using a 5N ZnSe effusion cell heated to 640 °C to 650 °C and a 4N MnSe cell at 820 °C to 850 °C, and their magneto-optical properties were compared with those of ZnMnTe films. An X-ray diffractometer using Cu-$K_\alpha$ radiation and reflection high energy electron diffraction (RHEED) were used to observe crystallinity. A (111) diffraction spectrum of weak intensity was observed by the X-ray diffractometer for the ZnMnTe and ZnMnSe films deposited on the QG.
substrates. Fig. 2 (a) shows a weak diffraction spectrum of (111) observed for the Zn$_{0.75}$Mn$_{0.25}$Te film deposited on the QG substrate. Although the films are considered to have a certain amount of crystallinity, RHEED that monitors the crystal growth on the surface of films during deposition showed a ring pattern which gives an indication of the polycrystalline film growth. The films were assumed to be polycrystalline grown perpendicularly to the surface of QG substrates. Fig. 2 (b) shows the diffraction spectra observed in the Zn$_{0.73}$Mn$_{0.27}$Te film grown on the SA substrate. As shown in Fig. 2 (b), the (220) spectrum of strong intensity is exhibited at $2\theta=41.9^\circ$, although the quite smaller (111) diffraction peak is also observed at $2\theta=25.3^\circ$. Moreover, the streaked pattern of RHEED was observed, indicating the single-crystal growth. The (220) epitaxial growth of the ZnMnTe film on SA is considered to be due to a specific (2022) face of the hexagonal (0001) C-cut $\alpha$-Al$_2$O$_3$ crystal observed at $2\theta=42.65^\circ$.

Fig. 3 represents the crystal structure of zinc-blend. Mn is substituted for the atoms of the group II element. The group VI element, Te or Se, is arranged in the center of a tetrahedrally bonded structure. The <111> and <220> directions of the zinc-blend crystal are indicated in Fig. 3.

When the concentration of Mn$^{++}$ is small ($x<0.01$), the Mn$^{++}$ spins have been regarded as isolated, i.e., independent from another$^{[17]}$. In that case, the magnetization $M$ is described by the Brillouin function and, this situation leads to the Curie-Weiss law in dependence on temperature. For magnetic semiconductors of arbitrary $x$, the magnetization $M$ is not expressed by the standard Brillouin function because of the Mn-Mn interaction. In this case, $M$ has been considered to be expressed by the Curie-Weiss law in the form:

$$M = \frac{C}{(T - \Theta)} H$$

where $C$ is the Curie constant of the paramagnetic phase, $T$ denotes the temperature, $H$ is the magnetic field, and $\Theta$ is the Curie temperature. Fig. 4 shows the magnetic properties of the ZnMnTe films observed by using a vibrating sample magnetometer.
Fig. 4 shows the magnetization curves for the Zn
0.73Mn
0.27Te film measured at –173 °C (100 K) and the room temperature (RT: 300 K) in the field range of ±20 kOe. Fig. 4 (b) is an M-T curve measured for the Zn
0.73Mn
0.27Te film grown on the SA substrate in relation to the temperature in the range of –195 °C (77 K) to RT. When applying the Curie-Weiss law to the M-T curve of Fig. 4 (b), the Curie temperature Θ was calculated at about −207 °C (66 K). The ZnMnTe film is considered to be paramagnetic at RT.

Atomic concentrations were determined as mole fractions through an energy dispersion spectral analysis. The thickness of the films was measured by using a Talystep profilometer. The Faraday rotation angle θF at RT was observed in the wavelength range of 460 nm to 700 nm by using a spectrophotometer.

2.2. AC Faraday Signal Measuring Equipment

We made direct observations of the Faraday rotation for the ZnMnTe films subject to alternating magnetic fields. Fig. 5 shows the equipment used for the measurement. It has a NdFeB ring magnet (with an outer diameter of 40 mm) driven by a servomotor at a 6000-rpm maximum rotation speed. The magnet has 32 poles along the periphery of the ring. Fig. 6 shows the locations of the ring magnet, a film, and the Hall sensor used to obtain a reference signal. Fig. 7 plots the magnetic flux distribution of the ring magnet. This equipment can produce an AC
The Faraday rotation of the film was measured at a point located 2 mm away from the outer periphery of the ring magnet, where the magnetic field is 620 Gauss (62 mT). A 532-nm, 3-mW green LD was used as the light source and an avalanche photodiode as the detector. After passing through a bandpass filter, the signal from the detector was measured by a digital oscilloscope. At 532 nm, the films exhibit a negative Faraday rotation (i.e., a counterclockwise rotation of the polarized light). On the other hand, the bare QG substrates exhibit a positive Faraday rotation. Therefore, the net signal was reduced by the substrates.

3. Results and Discussion

3.1. Bandgap Energy and Absorption Characteristics

$A_{II}^{\text{II}}Mn_{x}B_{VI}^{\text{VI}}$ alloys (where $A_{II}$ and $B_{VI}$ are group II and group VI elements, respectively) are direct-gap semiconductors whose band extrema occur at the $\Gamma$ point originating from the $s$ and $p$ orbitals of the constituent atoms (referred to as $sp$ bands)\cite{18,19}. A linear interpolation between $A_{II}^{\text{II}}B_{VI}^{\text{VI}}$ and $Mn_{x}B_{VI}^{\text{VI}}$ compounds gives a good first-order approximation of the dependence of the bandgap energy $E_g$ on $x$\cite{16,17}. The results upon replacing the group II element by Mn in Cd$_{1-x}$Mn$_x$Te, Zn$_{1-x}$Mn$_x$Te, and Zn$_{1-x}$Mn$_x$Se are shown in Fig. 8. As Mn is substituted for Cd or Zn in the CdTe, ZnTe, or ZnSe lattices, the energy gap increases. The energy gaps of the parent $A_{II}^{\text{II}}B_{VI}^{\text{VI}}$ and Mn$_x$B$_{VI}^{\text{VI}}$ binary compounds at RT are 1.53 eV, 2.28 eV, 2.71 eV, 2.90 eV, and 2.90 eV for CdTe, ZnTe, ZnSe, MnTe, and MnSe, respectively. Fig. 8 also shows the photon energy of the red, green, and blue laser beams. Whereas the red laser is useful for the CdMnTe films, the green laser is expected to be effective for the ZnMnTe films. Values obtained from the ZnTe-MnTe and ZnSe-MnSe interpolations are compared with those obtained from the $(\alpha h\nu)^2-E_g$ plot in Table 1, where $\alpha$ is the absorption coefficient of the film, $h$ is the Planck constant, and $\nu$ is the frequency of light used for the spectrophotometric measurement of $\alpha$. Fig. 9 shows the transmittance of the ZnMnTe and ZnMnSe films used in calculating the $(\alpha h\nu)^2-E_g$ plot values. The optical absorption edge differs by about 60 nm for the selenide films compared with the telluride films. The variation $\Delta E_g$ in Zn$_{1-x}$Mn$_x$Te is indicated for $x$=0.2 in Fig. 9. The absorption edge is shifted to higher energy (blue shift) by 0.11 eV. Whereas the difference in the film thickness was clearly reflected to the transmittance of the ZnMnSe films, the differences in the film thickness and types of substrates (QG and SA) were not clear in the transmittance data observed in the ZnMnTe films.

3.2. Faraday Rotation for ZnMnTe and ZnMnSe Films

The ZnMnTe and ZnMnSe films of the zinc-blend open gap, which exhibit a Faraday transition by the Zeeman...
Table 1: Values of bandgap energy for the ZnMnTe and ZnMnSe films on the QG and AS substrates

<table>
<thead>
<tr>
<th>Film</th>
<th>$E_g$ (eV)</th>
<th>$(a\nu h)^2 - E_g$ plot</th>
<th>Thickness (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zn$<em>{0.33}$Mn$</em>{0.67}$Se$^*$</td>
<td>2.74</td>
<td>2.69</td>
<td>1.40</td>
</tr>
<tr>
<td>Zn$<em>{0.33}$Mn$</em>{0.67}$Se$^*$</td>
<td>2.76</td>
<td>2.64</td>
<td>2.23</td>
</tr>
<tr>
<td>Zn$<em>{0.15}$Mn$</em>{0.85}$Te$^*$</td>
<td>2.38</td>
<td>2.38</td>
<td>2.81</td>
</tr>
<tr>
<td>Zn$<em>{0.15}$Mn$</em>{0.85}$Te$^*$</td>
<td>2.39</td>
<td>2.39</td>
<td>2.93</td>
</tr>
<tr>
<td>Zn$<em>{0.25}$Mn$</em>{0.75}$Te$^*$</td>
<td>2.42</td>
<td>2.42</td>
<td>2.36</td>
</tr>
<tr>
<td>Zn$<em>{0.25}$Mn$</em>{0.75}$Te$^*$</td>
<td>2.40</td>
<td>2.34</td>
<td>2.61</td>
</tr>
<tr>
<td>Zn$<em>{0.25}$Mn$</em>{0.75}$Te$^*$</td>
<td>2.43</td>
<td>2.38</td>
<td>2.36</td>
</tr>
</tbody>
</table>

\[ \theta_F = K \chi H \left( \frac{\hbar^2 \omega^2}{(E_g - \hbar^2 \omega^2)^{3/2}} \right) I \]  

(2)

where $K$ is a constant, $\chi$ is the magnetic susceptibility, $E_g$ is the bandgap energy, $\hbar \omega$ is the photon energy, and $I$ is the film thickness. The constant $K$ is written in the form:

\[ K = \left( \sqrt{F_0 / 2hc} \right) \left[ \left( \beta - \alpha \right) / g \mu_B \right] \]  

(3)

where $F_0$ is the oscillator strength and $\beta - \alpha$ is the $sp$-$d$ exchange constant. Equation (2) shows that $\theta_F$ is large ($\rightarrow \pm \infty$) near the optical absorption edge.

Fig. 10 shows the variation of the Faraday angle with the wavelength measured at RT in the Zn$_{1-x}$Mn$_x$Te and Zn$_{1-x}$Mn$_x$Se films deposited on the QG and SA substrates. A negative peak value of the Faraday rotation angle $\theta_F$ at RT occurs near 550 nm for the Zn$_{1-x}$Mn$_x$Te films that have an absorption edge at 520 nm to 530 nm, and near 450 nm for the Zn$_{1-x}$Mn$_x$Se films that have an absorption edge at 450 nm to 460 nm. The
Faraday rotation spectra of the Zn$_{1-x}$Mn$_x$Te films are not noticeably changed when the Mn concentration is altered. The rotation angle $\theta_F$ measured for the Zn$_{1-x}$Mn$_x$Te films in the wavelength range of 550 nm to 660 nm also exhibits a negative value. The substrates exhibited a small positive value of rotation. $\theta_F$ of the SA substrate is a fixed value of about $+0.02$ °/cm-G for wavelengths and that of the QG substrate is a fixed value of about $+0.03$ °/cm-G. The Faraday spectra in Fig. 10 have had the substrate spectra subtracted off. Whereas the difference in the Mn concentration was clearly reflected to $\theta_F$ of the ZnMnSe films, the differences in the Mn concentration and types of substrates (QG and SA) were not clear in the $\theta_F$ data observed in the ZnMnTe films. The values of the Faraday rotation angle $\theta_F$ depend on the thickness of films as well as the Mn concentration. The results suggest that the Zn$_{1-x}$Mn$_x$Te and Zn$_{1-x}$Mn$_x$Se films can be useful for green and blue LDs, respectively.

3.3. Green Laser Application of the ZnMnTe Film at RT

Fig. 11 plots the Faraday effect signal measured for the Zn$_{0.82}$Mn$_{0.18}$Te film deposited on QG using the ring magnet and 532-nm, 3-mW green LD. The upper curve graphs the Faraday rotation of the film and the lower one shows a reference signal of the fields measured when a Hall element is placed near the ring magnet. The rotating ring magnet produces an alternating field of 620-Gauss amplitude at a frequency of 1.28 kHz. The angle between the polarizer and analyzer axes was approximately 10°. As seen in Fig. 11, an undistorted Faraday rotation signal of 18 mV P-P has been observed for the ZnMnTe film. The signal is attenuated by about 25% due to the opposing Faraday rotation of the 0.5-mm-thick substrates. Similar Faraday rotation signals were observed for the Zn$_{0.80}$Mn$_{0.20}$Te and Zn$_{0.75}$Mn$_{0.25}$Te films. Using the present films and equipment, the signal to noise ratio (SNR) was 40 dB. The advantage of an optical field sensor is its anti-noise properties, since the sensor does not use electrical wires.

4. Conclusion

Magnetization in the II-VI paramagnetic semiconductor films stems from spin rotations, not from domain wall motions, therefore the films respond linearly to magnetic fields. It is expected that the films will have good response at high frequencies as well. In this study, we showed that the wide-gap ZnMnTe films work efficiently for 532-nm
green LDs. Although we have not confirmed the operation of the ZnMnSe films, we believe that they will be useful in green and blue laser applications.

Two groups of films prepared on the QG substrates and SA ones are entirely different in crystallinity. The films on QG are basically polycrystal films deposited on the amorphous glass substrate, and those on SA are epitaxial single crystal films grown on the hexagonal crystal substrate. However, a value difference in \( \theta_N \) between the ZnMnTe films on QG and the ZnMnTe films on SA could not clearly be confirmed. The result is considered to show that the magneto-optical effect is not practically influenced by the crystallinity of the magnetic films.

The Faraday rotation near the absorption edge and optical transparency are in a trade-off relation. The transmittance of films decreased gradually with shortening the wavelength of light from 700 nm to the optical absorption edge as shown in Fig. 9, and the decrease of transparency observed in the wavelength range of 550 nm to 700 nm for the ZnMnTe films is felt somewhat large. We evaluated the absorption characteristics of ZnTe films grown on the SA and ZnTe single crystal substrates by MBE. Those films also exhibited similar absorption characteristics to those of the ZnMnTe films at the same wavelength range of 550 nm to 700 nm. The multiple reflections of the thin ZnMnTe/ZnTe films may have concern in the decrease of transparency. On the other hand, the bulk ZnTe single crystal plate obtained from the Bridgeman crystal growth keeps high and constant transparency from 800 nm to the absorption edge as shown in Fig. A1 in the appendix\(^{[20]}\). The optical absorption is still a subject for further study. From a practical point of view, we are interested in an unstrained ZnMnTe single crystal that is grown by using the Bridgeman method.

Appendix

Transparent Characteristics of Bulk ZnTe Crystals at RT

Whereas the transmittance of the ZnMnTe films grown on the SA substrates by MBE decreased gradually with shortening the wavelength of light from 700 nm to the optical absorption edge as shown in Fig. 9, the undoped ZnTe (100)/(111) and P-doped (111) single crystals with a shape of 10.0 mm in length, 10.0 mm in width, and 0.5 mm in thickness kept a high and constant transmittance in that wavelength region as shown in Fig. A1. The transmittance of the O-doped ZnTe (111) single crystal is somewhat lowered near the optical absorption edge. Undoped ZnTe single crystals exhibited the photoluminescence spectrum of very low intensity at 558 nm (green) brought by a direct-gap transition at RT, and PL of very high intensity at the wavelength of 565 nm at RT was observed for the P-doped ZnTe single crystal. PL of the P-doped ZnTe has been considered to be due to a charge transition from the conduction band to the acceptor level formed by the impurity P atoms, and that transition produces PL of very high intensity\(^{[20]}\). The observed value 0.04 eV on \( \Delta E_g \) indicated in Fig. A1 is considered to show the energy difference between the energy of the acceptor level and that for the top of the valence band. The O-doped ZnTe single crystal exhibits the high intensity of PL at 680 nm (red) at RT as shown in Fig. A2. The group VI element Te is partially replaced with the impurity VI element O, which has the same electron-arrangement as Te, and PL has been considered to be due to luminescence arising from the energy of the excited O atoms bound by an isoelectric trap\(^{[20]}\). Fig. A3 shows the optical transparency of the ZnMnTe and ZnTe films grown on the SA and ZnTe single crystal substrates. In the optical wavelength range of 500 nm to 800 nm, the transparent properties of the ZnMnTe and ZnTe films on SA were almost the same, and the ZnTe films grown on the ZnTe single crystal substrates also exhibited lower transparency.

![Fig. A1. Optical transparency of the ZnTe single crystals measured at RT.](image-url)
Although the RHEED observation exhibited the (111) growth of the ZnTe film grown on the ZnTe substrate as shown in Fig. A4, the film looked metallic a little and was not stoichiometric in the composition of Zn and Te. Therefore, the decrease of transparency in this case (the ZnTe film grown on the ZnTe single crystal substrate) is considered to be due to an influx of excessive Zn atoms, which produces free electrons, into the film from the ZnTe substrate. Fig. A5 shows the optical transparency of the O-doped single crystal slab samples: Unpolished (unstrained) and polished (strained) to a mirror plane by using 1-μm diamond slurry. After polishing, the transmittance of the strained ZnTe single crystal drastically decreased, and the PL intensity decreased greatly as well. The decrease of the transmittance and PL intensity mostly recovered after annealing at 300 °C for 24 h in a vacuum. Two causes of the internal distortion in the films deposited on the SA crystal substrates are considered as follows: 1) A lattice distortion brought by the thermal distortion in film preparation and 2) a lattice mismatch between the crystal lattice of the film and that of substrate brought through the epitaxial growth.

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**Fig. A2. PL intensity of the unstrained and strained O-doped (111) single crystal slab samples. PL is observed at RT. The high intensity of PL observed at 680 nm (red) has been considered to be due to isoelectric trap[20].**

**Fig. A3. Optical transparency of the ZnMnTe and ZnTe films grown on SA and ZnTe single crystal substrates. Data was obtained at RT.**

**Fig. A4. RHEED pattern: (111) growth of the ZnTe film on the ZnTe single crystal (111) substrate.**

**Fig. A5. Optical transparency observed at RT for the O-doped (111) single crystal slab sample (10.0 mm×10.0 mm×0.5 mm): Unpolished (unstrained) and polished (strained).**
References


Masaaki Imamura was born in Ureshino in 1945. He received the B.E., M.E., and D.E. degrees from Kyushu University, Fukuoka in 1969, 1973, and 1979, respectively. Since 1985, he has been with the Department of Electrical Engineering, Fukuoka Institute of Technology (FIT), Fukuoka from 1985 to 1987 as an associate professor and since 1988 as a professor. He is an emeritus professor of FIT. His interests include the magneto-optics for II-VI diluted magnetic semiconductors (DMSs) such as ZnMnTe or ZnMnCoTe films synthesized using molecular beam epitaxy (MBE) and their magneto-optical applications to sensors.

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