Giant Faraday rotation spectra of $Zn_{1-x}Mn_xSe$ observed in high magnetic fields up to 150 T

T. Yasuhira, K. Uchida, Y. H. Matsuda, and N. Miura
Institute for Solid States Physics, University of Tokyo, Roppongi, Minato-ku, Tokyo, 106-8666, Japan

A. Twardowski
Institute of Experimental Physics, Warsaw University, Hoza 69, 00681 Warsaw, Poland
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We have observed interesting features in the energy and field dependencies of Faraday rotation spectra in $Zn_{1-x}Mn_xSe$ under high magnetic fields up to 150 T. Continuous magneto-optical spectra were measured on a two-dimensional plane as a function of magnetic field and photon energy. As the rotation angle is obtained as continuous lines on the plane, we can detect any small change of the Faraday rotation, such as the stepwise small increase corresponding to the crossover of the magnetic energy levels of Mn ion pairs. In a sample with a relatively low Mn concentration ($x=0.015$), we found that the sign of the field coefficient of the Faraday rotation is reversed at some field due to the competition between the paramagnetic component due to the magnetization of Mn ions and the diamagnetic component due to the interband transition in the matrix crystal. We also found that the diamagnetic part in high fields shows a peculiar energy dependence, taking a minimum at around 2.4 eV. This result suggests that the diamagnetic part is determined not only by the interband transition but also by the *d-d* transition in Mn ions. The magnetic-field dependence of the diamagnetic part showed a kinklike behavior at around $B=80$ T.

I. INTRODUCTION

Diluted magnetic semiconductors (DMS's) of II-VI compounds exhibit many fascinating properties,[1–3] such as giant Faraday rotation.[4,5] Faraday rotation (FR) in DMS alloys is in general very large due to the magnetization of the Mn^{2+} ions. Giant FR is of importance not only for device applications but also for investigating fundamental properties of DMS's.

FR in DMS's is a convenient means to study the magnetization of Mn ions optically because the FR angle is predominantly determined by the magnetization of Mn ions. In DMS's with a relatively low concentration of the magnetic component, giant FR is described by a Brillouin function like a curve corresponding to the Mn magnetization, and it shows a saturation at high magnetic fields. The saturation corresponds to a complete alignment of the spins of the isolated Mn^{2+} ions in strong fields. As the Mn content is increased, the Mn pairs and larger clusters formed in the crystal make a significant contribution to the magnetization, and the effect also shows up in the FR. For example, in high magnetic fields, additional structures appear due to the alignment of the moment of spin pairs of the neighboring Mn ions, and successive steplike changes are observed corresponding to the crossover of the magnetic energy levels of the Mn spin pairs.[6,7] From such steps we can estimate the antiferromagnetic coupling constant between the nearest-neighbor Mn ion pairs J_{NN}. The magnetization steps were studied in many DMS's. Regarding $Zn_{1-x}Mn_xSe$, Foner *et al.* studied the steps by magnetization,[6] but to the best of our knowledge there has been no report of the determination of J_{NN} in $Zn_{1-x}Mn_xSe$ by means of FR. In more highly doped samples, the effect of larger clusters reduces the FR due to the antiferromagnetic interaction and the saturation of the magnetization or FR takes place only in very high magnetic fields.[6,8] In a previous paper,[9] we determined J_{NN} in $Zn_{1-x}Mn_xSe$ from the Faraday rotation.

Besides the magnetic component, there are contributions from different other mechanisms in the FR. It is well known that the virtual interband transition gives rise to FR in nonmagnetic semiconductors.[10–13] Such a mechanism gives a FR in the opposite direction to the magnetic component. We can assign the magnetic component as a paramagnetic part and the interband component as a diamagnetic component. The diamagnetic term gives just a small correction to the FR in comparison to the paramagnetic term, but after the saturation of Mn magnetization in high magnetic fields, it should tend to give a significant contribution. There have been several investigations concerning how the competition between the paramagnetic component arising from the Mn magnetization and the diamagnetic part develops.[14–16] In the low-field range, the FR spectra were measured over a wide range of x values as a function of temperature spanning the paramagnetic and the magnetically ordered phases.[1,17] The spectral photon energy dependence was measured, but the field range was only up to a few T. Experiments with FR in DMS's have also been done at high magnetic fields over 100 T.[8,18] These experiments were performed by using monochromic light from a laser, so that the data taken through these experiments were only oscillatory signals with repeating bright and dark fringes. In the measurement using laser lines, it is not so easy to determine the sense of the rotation or a small change of the rotation.

Another interesting problem is the spectrum arising from the *d-d* transition in Mn^{2+}. There are some experimental reports about the intra-Mn^{2+} transition in DMS's. The magnetoreflectance spectra for $Zn_{1-x}Mn_xTe$ at low temperatures revealed an enormous Zeeman splitting of free excitons associated with the interband Faraday rotation.[1,19] However, there has been no indication of the Zeeman splitting associ-

ated with the transitions within Mn^{2+} ions. This means that the intra-Mn^{2+} transition should have little influence on the Faraday rotation. On the other hand, the spectra of the Faraday rotation and ellipticity in $Zn_{1-x}Mn_xSe$ clearly showed peaks which can be regarded as transitions within Mn^{2+} ions.[20] It is an interesting question whether there is any effect of the intra-Mn^{2+} transition in the Faraday rotation. These can be studied in high magnetic fields, where the effect of the large Brillouin-function-like part is saturated.

In the present study, we measured both the magnetic-field dependence and the photon energy dependence of the FR, exploiting a type of streak spectrometer comprising a charge-coupled-device (CCD) camera or an image-converter camera in high magnetic fields up to 150 T.[21] As the continuous spectra are available on a two-dimensional plane, a number of features of FR can be studied for DMS's.

II. EXPERIMENTAL PROCEDURE

The samples of $Zn_{1-x}Mn_xSe$ used in the experiments were single crystals, grown by the Bridgeman method. The Mn concentration of the samples were $x=0.015$ (sample No. 1), $x=0.05$ (sample No. 2), and $x=0.13$ (sample No. 3). The samples were cut to a thickness of about 0.5 mm and optically polished. The samples were put between two linear polarizers set at 45° with respect to each other. The light was led by optical fibers to and from the sample, which was mounted at the center of the magnet. Pulsed high magnetic fields up to 45 T were generated by a nondestructive wire-wound pulse magnet (pulse width ~10 ms), and higher magnetic fields up to 150 T (pulse width ~7 μs) were generated by the single-turn coil methods.[22] In the former case, the Faraday rotation spectra were measured over the wide photon energy range by using a CCD with a 512×512 detector segments matrix. The time-resolved spectra were obtained by a streak-mode of the CCD with sequential shift of the spectral signals synchronized with the pulse magnetic field. By this measuring system, we can obtain continuous FR spectra at low temperatures in one pulse. In a field produced by the single-turn coil technique (~150 T), time-resolved two-dimensional spectra were obtained by using a streak spectrometer comprising an image-converter camera and a CCD camera.

III. RESULTS AND DISCUSSION

Figure 1(a) demonstrates the raw data of the CCD image for the Faraday rotation spectra of the sample with $x=0.015$ up to 41.0 T at $T=4.2$ K. The vertical axis of the image corresponds to the time, which is synchronized with a pulsed magnetic field. The horizontal axis corresponds to the photon energy. Each dark and bright stripe in the figure is caused by a 90° rotation of the linearly polarized light. Replotting the image as a function of energy and magnetic field, we obtain an image as shown in Fig. 1(b). From such images we obtain the magnetic-field dependence of the FR at various photon energies.

Figure 2 shows another example of the streak spectra of the Faraday rotation in a sample with $x=0.13$, at $T=1.6$ K. We can see the stepwise increase of the Faraday rotation, corresponding to the magnetization steps due to the magnetization of the Mn ion pairs. From such steps, the nearest-neighbor antiferromagnetic exchange constant was obtained as $J_{NN}=-13.1\pm0.3$ K.[9]

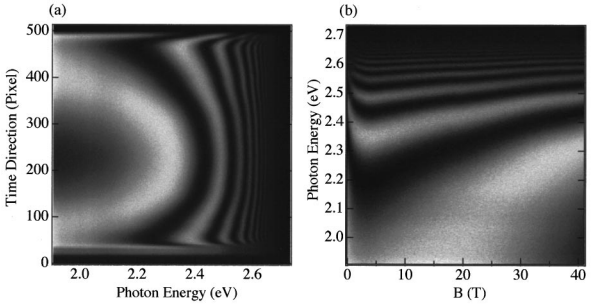

FIG. 1. Faraday rotation spectra of $Zn_{0.985}Mn_{0.015}Se$ at 4.2 K (CCD image). (a) The vertical axis of the image corresponds to time, which is synchronized with a pulsed magnetic field. The horizontal axis corresponds to the photon energy. (b) The replotted image of (a) as a function of energy and magnetic field.

In both Fig. 1(b) and Fig. 2, we can see that the Faraday rotation shows a rapid rise in the low-field region corresponding to the magnetization of the Mn ions. Figure 1(b) shows that there is a sharp turnaround of the FR stripes at about 7 T. Scanning the FR spectra along the magnetic-field direction at a constant photon energy, we notice the line encounters the same continuous stripe twice. This indicates that the field coefficient of the FR (differential Verde constant) is reversed at some field around 7 T at $T=4.2$ K.

In order to study more details of the FR spectra in the high-field range, the measurement was extended up to 150 T using the single-turn coil technique[22] and a streak spectrometer with an image-converter camera. Figure 3 shows the data of the FR in a sample with $x=0.015$, at $T\sim8$ K. We can see a striking anomaly at around 2.4 eV in Fig. 3. The Faraday rotation angle of various photon energies up to 150 T at $T\sim8$ K is plotted in Fig. 4 as a function of magnetic field. The rotation angle at every photon energy increased up to 7 T, as is observed in the low-field measurement. At 7 T, the field coefficient of the FR is reversed, showing a turnaround. The field dependence of the rotation angle before the turnaround is a Brillouin-function-like rapid increase due to the paramagnetic contribution from the Mn^{2+} magnetization. After the turnaround, it is almost a linearly decreasing function

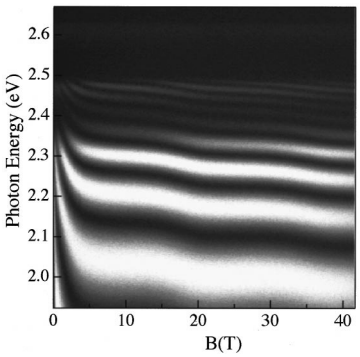

FIG. 2. Faraday rotation spectra of $Zn_{0.87}Mn_{0.13}Se$ at 1.6 K (CCD image). The horizontal axis corresponds to the magnetic fields, and the vertical axis to photon energy. Two points with wavy stripes correspond to the magnetization step.

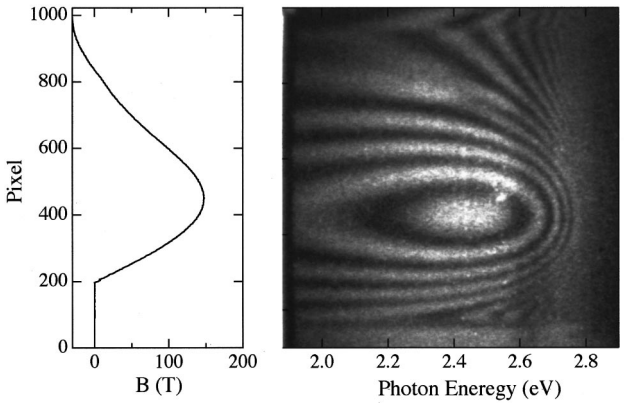

FIG. 3. Faraday rotation spectra of $Zn_{0.985}Mn_{0.015}Se$ at 7 K (CCD image). The horizontal axis corresponds to the photon energy, and the vertical axis to time which is synchronized with a pulsed magnetic field.

of magnetic field. For $B>7$ T, the diamagnetic contribution becomes prominent after the magnetization saturation and the sign of the FR gradient changes.

The FR is represented by two terms,

$$\theta_F=\theta_0(\hbar\omega)\mathcal{B}_{5/2}(B,T)+\chi(\hbar\omega)B, \tag{1}$$

where the first and the second terms represent the paramagnetic contribution of the Mn ions and the diamagnetic contribution of the interband transition, respectively. $\mathcal{B}_{5/2}(B,T)$ is the Brillouin function for magnetic field B and temperature T. θ_0 and χ are fitting parameters which are dependent on the photon energy. The small phenomenological antiferromagnetic interaction of isolated Mn^{2+} ions with distant neighbors[23,24] is neglected for simplicity. As the signs of the two terms are opposite from each other, the direction of FR can be reversed when the first term is saturated in high magnetic fields. It should be noted that $Cd_{1-x}Mn_xTe$ did not show such a reversal of the field coefficient of the FR, and the reversal is a unique feature in $Zn_{1-x}Mn_xSe$ due to the fact that the two terms in Eq. (1) compete with the same order of magnitude.

Figure 4 shows a comparison between the experimental data of the field dependence of the FR at different photon energies with the calculated result using Eq. (1) with θ_0 and χ as fitting parameters. We can see a reasonably good agreement between experiment and calculation. In Fig. 4, we can

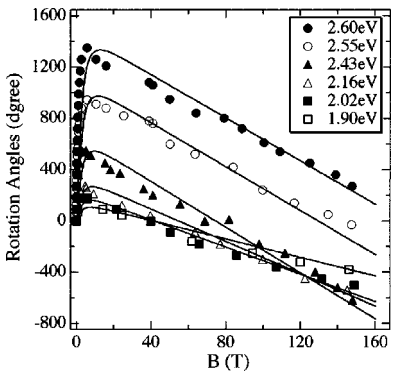

FIG. 4. Faraday rotation angles of $Zn_{0.985}Mn_{0.015}Se$. This graph is plotted taking the reverse rotation at 7 T into consideration.

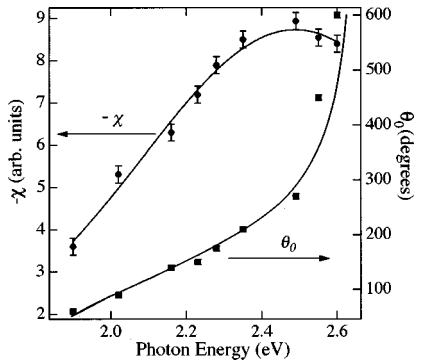

FIG. 5. Photon energy dependence of the fitting parameters, θ_0 and $-\chi$. The solid lines are guides for the eye.

see that the gradient of the linearly decreasing part of the FR (χ) above $B>7$ T has a large photon energy dependence. Furthermore, even the value of the FR angle itself changes the sign in high fields above 50 T. The zero cross of the FR occurs at lower fields as the photon energy is decreased. Such a behavior of χ can be investigated only by a measurement over a wide field range as in the present experiment. The fitting parameters θ_0 and χ are shown as functions the photon energy $\hbar\omega$ in Fig. 5. θ_0 increases as the photon energy is increased toward the energy gap. This behavior of θ_0 is reasonable if we recall the expression of the giant Faraday rotation.[4]

$$\theta_F=\left(\frac{\sqrt{F_0}d}{2\hbar c}\frac{\beta-\alpha}{g_M\mu_B}M\right)\frac{1}{E_0}\frac{y^2}{(1-y^2)^{3/2}},$$

$$y=\frac{E}{E_0}, \tag{2}$$

where F_0 is a constant involving the oscillator strength of the excitonic transition, α and β are the exchange integrals for the conduction- and valence-band electrons with Mn^{2+} ions, M is the magnetization per unit volume, g_M is the Landé g factor of the Mn^{2+} spins which is about 2, E_0 is a single oscillator energy, E is the photon energy, and μ_B is a Bohr magneton.

The value of the coefficient of the linear term $-\chi$ is also increased with increasing photon energy below $\hbar\omega \sim 2.4$ eV, but it has a maximum at around 2.4 eV. The existence of the maximum of the linear term is actually the reason for the extremum point in the FR image in Fig. 3. The diamagnetic component should have increased as the photon energy increases below the band gap because it is predominantly determined by the interband transition of the mother crystal ZnSe.[11–13] This anomaly in the energy dependence of $|\chi|$ at around 2.4 eV suggests some other contribution to the FR. In the experiment of the piezomodulated reflectivity spectra of $Zn_{1-x}Mn_xSe$,[19] a structure was observed in the vicinity of 2.3 eV, and the structure was attributed to an electronic transition within Mn^{2+} ions [the ${}^6A_1({}^6S) \rightarrow {}^4T_2({}^4G)$ transition]. The energy of the anomaly in the present experiment (2.4 eV) is very close to this Mn^{2+} d-d transition. The origin of such a unique structure appearing in the Faraday rotation spectra might originate from the ${}^6A_1({}^6S) \rightarrow {}^4T_2({}^4G)$ transition.

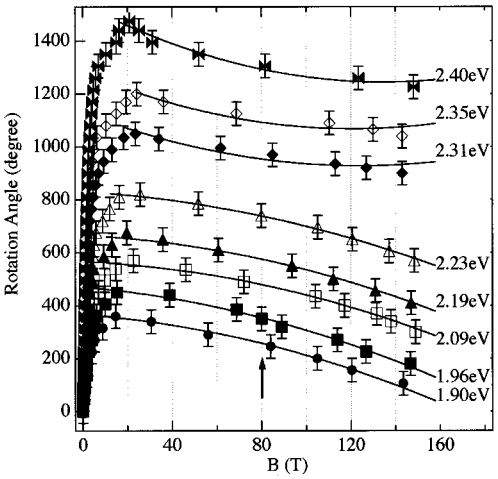

FIG. 6. Faraday rotation angles of $Zn_{0.95}Mn_{0.05}Se$ at 8 K at various photon energies up to 150 T. This graph is plotted taking the reverse rotation at 20 T into consideration. The solid lines are guides for the eye. Depending on the photon energy, the line shows an upturn or a downturn at around 80 T, as shown by an arrow.

Another type of anomaly was found in the field dependence of the FR for a sample with $x=0.05$. In Fig. 6 we plot the FR angle in this sample at various photon energies up to 150 T at ~8 K. The turnaround of the FR is also noticed in this sample at 20 ± 0.5 T. A remarkable feature is that the field dependence of the Faraday rotation shows a distinct upturn above 80 T for an energy higher than 2.3 eV, and a downturn at the same field for the lower energy. These features were also confirmed by measuring the Faraday rotation with monochromatic radiation from lasers at wavelengths of 514.5 nm (2.4 eV) and 632 nm (1.96 eV). The origin of the anomalous field dependence is not clear at the moment, but, as mentioned above, this energy is very close to the energy of intra-Mn^{2+} transition. This effect may be also related to the *d-d* transition. Further study is necessary for clarifying these anomalies near 2.4 eV.

IV. CONCLUSION

We have observed various features in the giant Faraday rotation in $Zn_{1-x}Mn_xSe$, by measuring continuous spectra recorded on a two-dimensional plane as a function of both magnetic field and photon energy in high magnetic fields up to 150 T. The FR shows a clear turnaround at some field as a function of magnetic field due the competition between the paramagnetic term arising from the Mn magnetization and the diamagnetic term arising from the interband transition. The field coefficient of the latter shows a clear anomaly at a photon energy around 2.4 eV. The field dependence of the FR shows an upturn and a downturn at around 80 T, and the turning direction is swapped at around 2.3 eV. These energies are very close to the Mn^{2+} *d-d* transition which was assigned as $^6A_1(^6S)\rightarrow{}^4T_2(^4G)$. We can deduce that the Mn^{2+} *d-d* transition may be responsible for these features observed in the Faraday rotation in $Zn_{1-x}Mn_xSe$.

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