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1998 J. Phys.: Condens. Matter 10 11611

(http://iopscience.iop.org/0953-8984/10/49/056)

J. Phys.: Condens. Matter **10** (1998) 11611–11615. Printed in the UK PII: S0953-8984(98)96572-9

Anomalous photon energy and field dependencies of the Faraday rotation in ZnMnSe under high magnetic fields

T Yasuhira†, K Uchida†, Y H Matsuda†, N Miura† and A Twardowski‡

† Institute for Solid State Physics, University of Tokyo, 7-22-1 Roppongi, Minatoku, Tokyo 106-8666, Japan

‡ Institute of Experimental Physics, Warsaw University, Hoża 69, 00-681 Warsaw, Poland

Received 4 June 1998

Abstract. Pulsed-field measurements of the Faraday rotation (FR) spectra over a wide photon energy range for $Zn_{1-x}Mn_xSe$ ($x = 0.015$) were made up to 150 T at low temperatures by means of a streak spectrometer. We observed a reversal of the direction of the FR with increasing field. Dividing the Verdet constant into a paramagnetic term in the low-field range and a diamagnetic term in the high-field part, we found that the latter exhibits anomalies at around 2.4 eV.

1. Introduction

The introduction of magnetic ions in semiconductors results in a fascinating class of semiconducting alloys—known as diluted magnetic semiconductors (DMSs). The most popular group of DMSs are the II–VI compounds in which a fraction x of the cations are Mn^{2+}, e.g., $Cd_{1-x}Mn_xTe$. Such alloys as $A^{II}_{1-x}Mn_xB^{VI}$, in contrast to $A^{II}B^{VI}$ semiconductors, exhibit many striking magneto-optical properties [1, 2] such as giant Faraday rotation [3, 4]. Their special magneto-optical behaviours are recognized as arising from the exchange interaction between the d electrons of Mn and the sp band electrons.

The FR in DMS alloys is very large in general, with the sign opposite to that for nonmagnetic II–VI compounds. There were several studies which investigated how the sign reversal develops by measuring the Faraday effect over a wide range of x-values as a function of temperature spanning the paramagnetic and the magnetically ordered phase [2]. These experiments were done over a wide range of photon energy up to a magnetic field of a few teslas. On the other hand, experiments on the Faraday effect for DMSs in much higher magnetic fields, over 100 T, were also performed. These experiments were realized by using monochromatic light from a laser as a light source [5, 6]. The data taken from experiments using lasers, however, provide information only at a single photon energy, so it is difficult to obtain detailed information on the FR of the spectra.

In earlier work on such samples as $Cd_{1-x}Mn_xTe$ [4] and $Zn_{1-x}Mn_xTe$ [3], a strong interband FR has been reported. The magnetoreflectance spectra for $Zn_{1-x}Mn_xTe$ [2, 8] at low temperatures showed an enormous degree of Zeeman splitting of the free excitons associated with the interband FR, but there were no indications of a Zeeman splitting associated with the transitions within Mn^{2+} ions. This means that intra-Mn^{2+} transitions should have little influence on the FR. However, the spectra of the FR and ellipticity in $Zn_{1-x}Mn_xSe$ clearly showed peaks, which can be regarded as indicating the transitions within Mn^{2+} [7].

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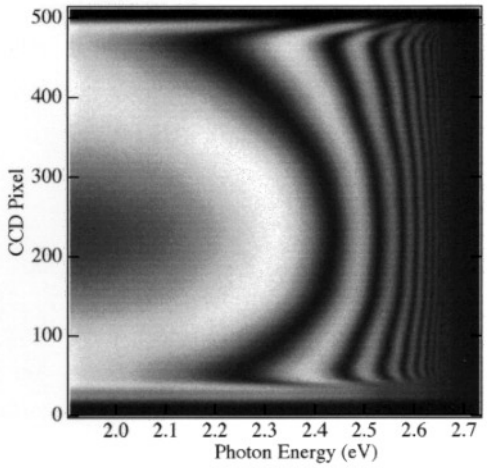
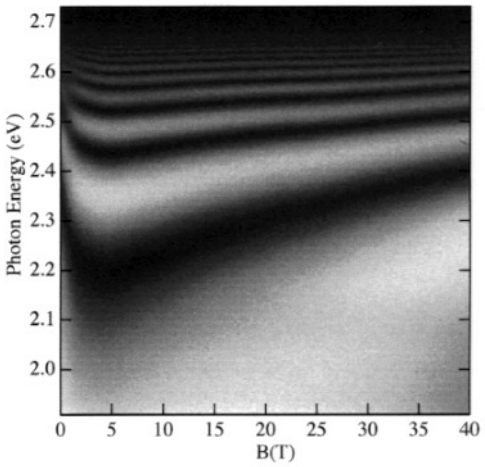
In this paper we report FR spectra up to 150 T over a wide photon energy region. We have observed clear anomalies at around the photon energy of 2.4 eV. This energy is very close to the Mn^{2+} d–d transition which was assigned as $^6A_1(^6S) \rightarrow {}^4T_2(^4G)$. Therefore, we concluded that this unique feature is a signature of the contribution of the intra-Mn^{2+} transitions to the FR in $Zn_{1-x}Mn_xSe$.



(a)



(b)

Figure 1. The FR spectra of $Zn_{0.985}Mn_{0.015}Se$ at 1.6 K. (a) The CCD image of the FR up to 41 T. (b) The transformed image derived from (a) with the magnetic field on the horizontal axis and the photon energy on the vertical axis.

2. Experiment

The samples of $Zn_{1-x}Mn_xSe$ with $x = 0.015$ used in the experiments were single crystals, grown by the Bridgman method. The Mn concentration was determined by atomic

absorption. Experiments were performed on polished platelets whose thicknesses were chosen so as to give at least a few full rotations. The light was transmitted through the sample normal to the face of the platelets and the field direction (the Faraday configuration). The samples were put between two linear polarizers set at 45° with respect to each other. Pulsed high magnetic fields up to 42 T were generated by a nondestructive pulse magnet (pulse width $\sim$ 10 ms), whereas higher magnetic fields up to 150 T (pulse width $\sim$ 7 μs) were generated by the single-turn coil methods. In the former case, the FR spectra were measured over a wide photon energy range by using a charge-coupled device (CCD) with a 512 $\times$ 512 detector array combined with a monochromator and a 150-groove grating. The time-resolved spectra were obtained from a streak mode of the CCD with the sequential shift of the spectral signals synchronized with the pulsed magnetic field. Using this measuring system, we can take continuous FR spectra at temperatures up to 1.6 K in one pulse. In the latter field, time-resolved two-dimensional spectra were obtained by using a streak spectrometer comprising an image-converter camera and a CCD camera. In the image-converter camera, the image of the spectra was streaked in the perpendicular direction, synchronously with the magnetic field, and thus the entire magnetic field dependence of the spectra is obtained in one sweep of the pulse field. The image data from the CCD camera were analysed digitally with a computer. A Xe flash lamp was used as a light source and the optical fibre system was employed to transmit the light signal.

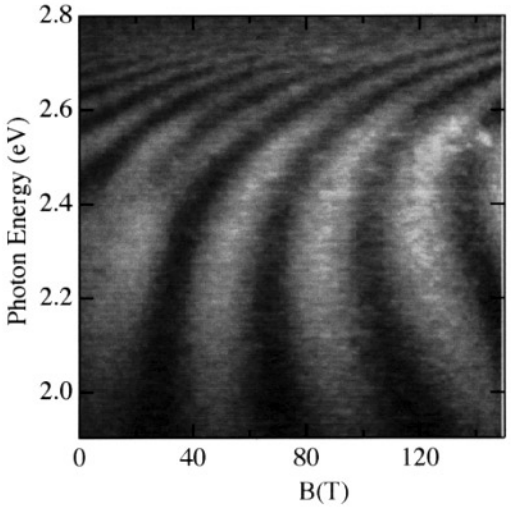


Figure 2. The Faraday rotation spectra of $Zn_{0.985}Mn_{0.015}Se$ up to 150 T obtained by the streak spectrometer.

3. Results and discussion

Figure 1(a) demonstrates the resulting FR spectra up to 41.0 T. The vertical axis of the image corresponds to the time, which is synchronized with a pulsed magnetic field; for the 0th and 512th pixels the magnetic field is low and at around the 250th pixel it is at its maximum. The horizontal axis corresponds to the photon energy. Each striped pattern, repeating dark and bright areas, is caused by 90° rotation of the incident polarized light. From the image, we obtain the energy dependence of the FR in high magnetic field. Remarkably, we notice that

the continuous striped patterns turn sharply to higher energy. (b) shows the transformed image derived from (a). The horizontal axis corresponds to the magnetic field and the vertical axis to the photon energy. The sharp change of the CCD image can be found at about 5 T.

In figure 1(b), which shows a replotting of (a) with the magnetic field on the horizontal axis, it is clear that the direction of the FR changes sign at around 5 T. In addition, it is clear that there is a large dispersion in the FR. In order to study more detail of the FR spectra, the measurement was extended up to 150 T using the single-turn coil technique and an image-converter camera. The streak spectra up to 150 T are demonstrated in figure 2. The image of the vertical axis corresponds to the time of the synchronized pulsed magnetic field up to 150 T, and the horizontal axis to the photon energy. A unique structure was observed. $Cd_{1-x}Mn_xTe$, one of the most typical DMSs, showed no such feature. The Faraday effect of this material is expected to be mainly caused by the interband transition. We can see a striking anomaly at around 2.4 eV as a centre of the half-rounded shape of the stripes in figure 2.

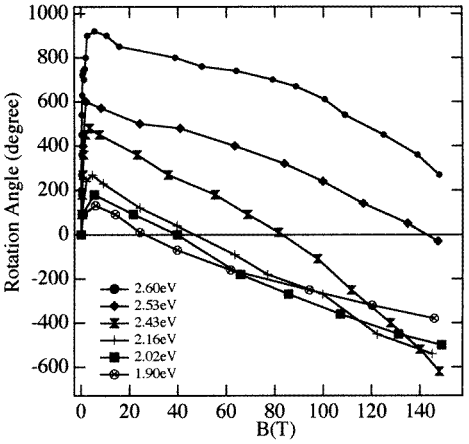


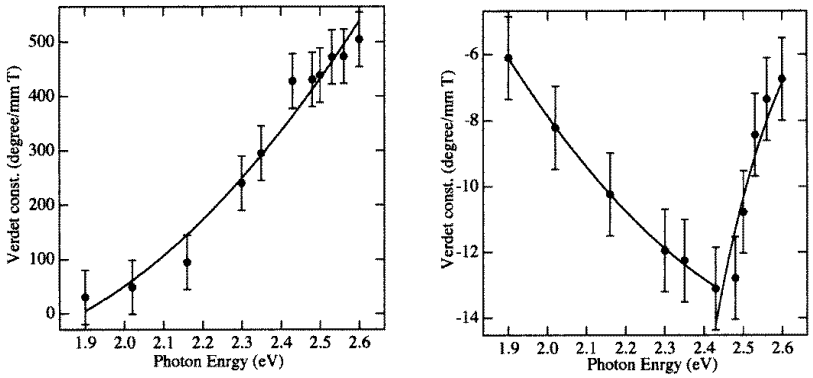
Figure 3. The Faraday rotation angle of $Zn_{0.985}Mn_{0.015}Se$. This graph is plotted taking the reverse rotation at 5 T into consideration.

The FR angles for various photon energies up to 150 T are plotted in figure 3. The rotation angles for every photon energy increased until 5 T was reached, i.e. in the low-field measurement. When the reverse rotation occurred, the rotation angle began to decrease. The field dependence of the rotation angles before the reversal of the FR is a Brillouin-function-like rapid increase, but after the reversal it was almost linear. Qualitatively, the FR of a DMS can be expressed as the sum of a Brillouin-function-like paramagnetic part and a diamagnetic part. The contribution of the latter part is considered to be almost proportional to the magnetic field and opposite in sign to the former. For $Zn_{1-x}Mn_xSe$ with $x = 0.015$, at $B < 5$ T the Brillouin-function-like part is larger than the linear part, but at $B \geqslant 5$ T the influence of the linear part overcomes the Brillouin-function-like part and the sign of the FR angle changes. We approximate the field dependences of the FR angles in the low-field limit $B < 5$ T and for $B \geqslant 5$ T separately (as functions of B), and from the equation [3]

$$\theta_F = VBl$$

we calculated the Verdet constant V; figures 4(a) and 4(b) show V for the two regions. The Verdet constant for $B < 5$ gets larger as the photon energy increases. However, for

$B \geqslant 5$, V has a minimum at around 2.4 eV. In the experiment in which the piezomodulated reflectivity spectra of $Zn_{1-x}Mn_xSe$ [8] were obtained, a structure attributed to an electronic transition of Mn^{2+} was reported in the vicinity of 2.3 eV. This special energy 2.4 eV is very close to this Mn^{2+} d–d transition, so the origin of such a unique structure appearing in the FR spectra can be considered to be the $^6A_1(^6S) \rightarrow {}^4T_2(^4G)$ transition. This peculiar feature suggested that the Mn^{2+} transitions play an important role in the FR in $Zn_{1-x}Mn_xSe$. Further research on this is under way in our laboratory.



(a) (b)

Figure 4. The Verdet constant of $Zn_{0.985}Mn_{0.015}Se$. (a) $B < 5$ T. (b) $B \geqslant 5$ T.

In summary, we measured the FR spectra of $Zn_{1-x}Mn_xSe$ ($x = 0.015$) up to 150 T over a wide photon energy range, and found that the direction of the FR was changed by the external magnetic field when it exceeded 5 T. Furthermore, a peculiar feature appeared at around 2.4 eV; this suggests that intra-Mn^{2+} transitions contribute to the Faraday effect in $Zn_{1-x}Mn_xSe$.

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