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Giant Zeeman splitting in the magneto-reflectance spectra of a diluted magnetic semiconductor (Zn,Cr)Te

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1. Introduction

Diluted magnetic semiconductors (DMSs), in which a part of non-magnetic cations in II-VI or III-V semiconductors are substituted by magnetic elements, have long been attracting attention due to its peculiar magnetic, transport and optical properties. Recently, they have been studied as possible candidates of ferromagnetic semiconductors, which are desired for the application in spintronics. One of the most characteristic features of DMSs is the strong exchange interaction between the delocalized band electrons (*s*- or *p*-like orbitals) and the localized electrons (*d*-like orbital) of magnetic ions. This interaction, called *sp-d* exchange interaction, causes peculiar magneto-optical effect such as giant Zeeman splitting[1]. Among various materials of DMSs, (Zn,Cr)Te draws interest due to an experimental observation of room-temperature ferromagnetism[2]. In addition, the intrinsic nature of ferromagnetism was confirmed from the consistency between the magnetic-field dependencies of macroscopic magnetization measured by SQUID and the magnetic circular dichroism (MCD) near the band-gap of the host ZnTe[2]. It is considered to be essential for understanding of the intrinsic properties of DMSs and of the mechanism of ferromagnetism to estimate the magnitude of the exchange interaction. Previously, the exchange constant of (Zn,Cr)Te was derived from the magneto-reflectivity measurement on bulk crystals[3]. However, the reported value was too large compared to other Cr-based II-VI DMSs[3,4], and there might be some uncertainties which resulted from a too small value of the Cr content in measured samples. On the other hand, in our pervious attempt to estimate the Zeeman splitting by measuring the magneto-reflectance of (Zn,Cr)Te thin films, the obtained value of the exchange constant was too small[5], where we might underestimate the splitting energy of a thin (Zn,Cr)Te layer affected by non-magnetic ZnTe layer underneath it. In the present study, we have performed the magneto-reflectance measurement of (Zn,Cr)Te thin films with different thicknesses and have attempted to extract the intrinsic magneto-optical property of (Zn,Cr)Te.

2. Experimental methods

The growth of $Zn_{1-x}Cr_xTe$ thin films was performed by molecular beam epitaxy (MBE) equipped with solid sources of Zn, Te and Cr. In order to avoid the deterioration of optical quality due to strain, a ZnTe (001) monocrystalline piece was used as a substrate and a ZnTe buffer layer (~200nm) and successively a $Zn_{1-x}Cr_xTe$ layer was grown on it. The Cr content x in the $Zn_{1-x}Cr_xTe$ layer was limited to less than x ~0.01. In this study, we prepared a series of $Zn_{1-x}Cr_xTe$ films with varied thicknesses d in the range of 0.4~1.4μm. The Cr content x in the $Zn_{1-x}Cr_xTe$ layer was estimated using electron probe microanalyzer (EPMA). The magnetization of the films was measured using superconducting quantum interference device (SQUID) magnetometer with a magnetic field applied perpendicular to the film plane. We measured reflectance spectra using a high-pressure Xe lamp, a monochromator of a focal length of 25 cm (a diffraction grating of 2400 grooves/mm), and a charge coupled device (CCD) camera. A magnetic field up to 7 T was applied for the samples immersed in superfluid liquid helium and the magneto-reflectance spectra in the Faraday configuration (light wave vector parallel to magnetic field) were measured for the two components σ^+ and σ^- of the circular polarization. The splitting energy was determined by subtracting the exciton energies observed in the reflectance spectra for the both polarizations.

3. Experimental results and discussions

Figure 1 shows a typical example of the magneto-reflectance spectra of a $Zn_{1-x}Cr_xTe$ film. The Cr content of this sample is x = 0.0026 and the thickness of the $Zn_{1-x}Cr_xTe$ layer is d = 1.4μm. A "dip" structure at around 2.388eV in the reflectance spectra at zero field, close to the band-gap energy of ZnTe (2.39eV at T = 4.2K), is assigned as a signal of the free exciton of the band-edge. At a magnetic field of 7 T, the position of this "dip" structure was shifted to a higher energy for the both polarizations but that for the σ^+ circular polarization was located at a higher energy compared to that for the σ^- polarization. Figure 2 plots the excitonic energy derived from the "dip" positions in the reflectance spectra as a function of magnetic field for the respective circular polarizations. As shown in the figure, the excitonic energy for the both polarizations was blue-shifted with the increase of magnetic field, but the shift for the σ^+ circular polarization is always larger than that for the σ^- polarization. The splitting energy between the σ^+ and σ^- polarizations is ΔE = 0.68 meV at the maximal field of 7 T. The order of the exciton energies for the σ^+ and σ^- polarizations, which is the opposite to that in Mn-doped II-VI DMSs, suggests a ferromagnetic *p-d* exchange interaction in $Zn_{1-x}Cr_xTe$.

In the measurement of the energy splitting for

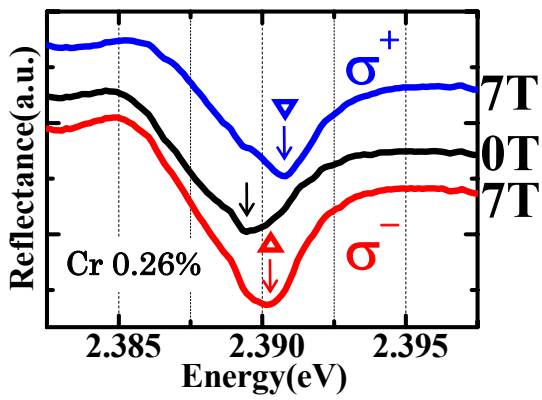

Fig. 1 Magneto-reflectance spectra of $Zn_{1-x}Cr_xTe$ (x = 0.0026, thickness 1.4μm). The spectra were taken for the σ^+ and σ^- circular polarizations in the Faraday configuration at 2K.

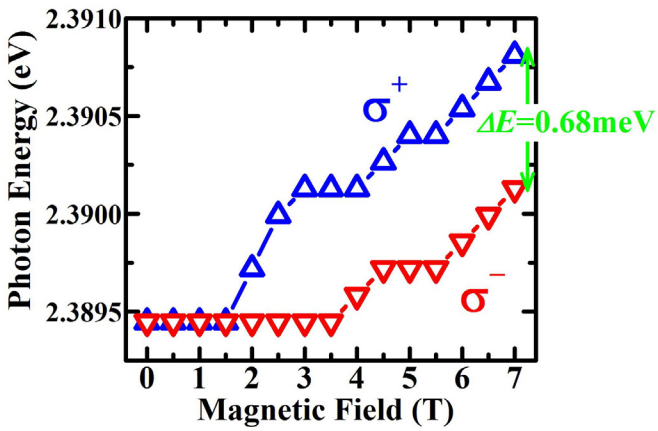

Fig. 2 Magnetic-field dependence of the excitonic energies for the respective circular polarizations for the same sample as shown in Fig. 1.

$Zn_{1-x}Cr_xTe$ films having different thicknesses and almost the same Cr composition around x = 0.0026, we found the following tendency; with the increase of thickness d of the $Zn_{1-x}Cr_xTe$ layer, the splitting energy at 7 T increases until d ~ 1.0 μm, and then is saturated around d ~ 1.0 μm. This dependence of the splitting energy on the thickness d suggests that the reflectance spectra from a thin $Zn_{1-x}Cr_xTe$ layer are affected by the ZnTe layer which was deposited beneath the $Zn_{1-x}Cr_xTe$ layer. The splitting energy saturated for d > 1.0 μm is considered not to be affected from the ZnTe underlayer and that value could be regarded to reflect the intrinsic property of $Zn_{1-x}Cr_xTe$.

In the magneto-reflectance spectra shown in Fig. 1, only two components of the excitonic transition were observed for the both polarizations, instead of four components corresponding to the spin splitting of J = 3/2 states of the heavy-hole and light-hole subbands. If the signal observed in the magneto-reflectance spectra is assumed due to the heavy-hole subband, the giant Zeeman splitting due to the *sp-d* exchange interaction is expressed as

$$\Delta E = N_0(\alpha - \beta)\, x \langle S_z \rangle ,$$

where N_0 is the number of cation sites in the unit volume, α, β are the constants of the *s-d* and *p-d* exchange interaction, g is the g-factor determined from the model calculations[6], $\langle S_z \rangle$ is the average alignment of localized moments of the magnetic element. In the mean-field approximation, the Zeeman splitting ΔE is proportional to the macroscopic magnetization of localized moment $M = N_0\, g\, \mu_B x \langle S_z \rangle$, where μ_B is the Bohr magneton. Therefore, the exchange constant $N_0(\alpha - \beta)$ can be derived by comparing the observed splitting energy in the magneto-reflectance spectra and the macroscopic magnetization measured by SQUID. From the observed splitting in Fig. 2, $N_0(\alpha - \beta)$ = 0.354 eV is derived for the Cr content of x = 0.0026.

4. Summary

We have measured the Zeeman splitting energy in the magneto-reflectance spectra of $Zn_{1-x}Cr_xTe$ thin films grown by MBE. From the measurement on $Zn_{1-x}Cr_xTe$ layers with different thicknesses d, the intrinsic magneto-optical properties of $Zn_{1-x}Cr_xTe$ could be confirmed for samples with d > 1.0 μm. From the comparison of the splitting energy in the magneto-reflectance spectra and the macroscopic magnetization measured by SQUID, the constant of the *sp-d* exchange interaction $N_0(\alpha - \beta)$ = 0.354 eV was derived for the Cr content of x = 0.0026.

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