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Magnetic and Magneto-Optical Properties of a Novel Ferromagnetic Semiconductor $CdGeP_2$:Mn

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Magnetic and magneto-optical properties were measured at room temperature in the Mn-diffused surface layer of a $CdGeP_2$ single crystal. The magnetization curve measured using VSM clearly shows a ferromagnetic hysteresis loop with Hc=0.4 kOe superposed on the diamagnetic component. The ferromagnetic Curie temperature was determined as 320 K from the magnetization vs. temperature curve. The magneto-optical Kerr effect was measured between 1.2 and 4 eV. The peak Kerr ellipticity of 0.14 deg appeared around 1.75 eV.

Key words: magnetic semiconductor, $CdGeP_2$:Mn system, chalcopyrite structure, room temperature ferromagnetism, magneto-optical Kerr spectrum

1. Introduction

Spin-electronics have been proposed to utilize spin-related properties of electrons as well as charge-related ones to extend functionality of electronic devices. For this purpose magnetic semiconductors attract attention since they posses both magnetic and semiconducting properties.

A lot of Mn-containing diluted magnetic semiconductor compounds are being studied at present and all the materials are based on either II-VI or III-V binary compounds.[1, 2] However, the highest Curie temperature obtained to date is about 110 K in $Ga_{1-x}Mn_x$ As. For practical application room temperature ferromagnetism has been strongly required.

Here we pay attention to II-IV-V_2 ternary semiconductors. The II-IV-V_2 compounds were first investigated by Goryunova[3] as a group of crystal-chemical analogs of III-V compounds. A series of compounds from this family attracts attention by their high nonlinear optical characteristics ($ZnGeP_2$, $CdGeAs_2$), polarization optical and emission properties, strong photo-response ($CdSnP_2$, $CdGeP_2$, $CdSiAs_2$), and also by wide possibilities in heteroepitaxy of solid solutions with binary compounds (GaAs, InP).[4] Since the II-IV-V_2 family contains the group II element as a constituent element, it can more easily accept the divalent Mn atom than the group III-V compounds. In addition, the electronic properties are quite similar to those of III-V compounds, which is promising for future device applications. Nevertheless no investigation have been done to date on the ternary system of II-IV-V_2 with considerable content of a transition element and their possible magnetic properties.

Among the II-IV-V_2 family we selected $CdGeP_2$ in the present study, since Mn is known to occupy the Cd site easily in CdTe. The $CdGeP_2$ crystallizes in tetragonal chalcopyrite structure with lattice constants of a=0.5741 nm and c=1.0775 nm, and u-parameter of 0.282.[3] The energy gap is 1.72 eV (300K) and 1.83 eV (≤80K).[5] Both n and p conductivity types were reported, with the mobility 1500 cm^2/Vs and 90 cm^2/Vs, respectively.[5]

Recently we have succeeded in observing the room temperature ferromagnetism in the $CdGeP_2$:Mn magnetic semiconductor system.[6]

This paper describes preparation, magnetic and magneto-optical properties of the grown material in the $CdGeP_2$:Mn quaternary system.

2. Experimental

As a host material we used a single crystal of $CdGeP_2$, which has been grown at the Ioffe Institute by directional crystallization. The flat-parallel crystal plate showed nearly rectangular shape with size of 3×5 mm^2.

Thin Mn layer of about 30 nm in thickness was deposited on the $CdGeP_2$ crystal in a MBE chamber using a Knudsen cell for evaporation of Mn source, followed by the thermal treatment at about 500°C for 30 minutes. Details of preparation techniques are described elsewhere. The control of the surface quality for the starting substrate and modified surface at every stage of a technological trail was monitored using high-energy electron diffraction (RHEED) technique.[7]

The microstructure on the surface and cleavage of a structure was studied with EDX and FE-SEM techniques using the Hitachi S-4500 scanning electron microscope with the instrument EMAX-577OW and Si crystal detector of Horiba Ltd. model S782XI with the spectral resolution of 144eV. X-ray diffraction (XRD) measurements were carried out on instruments Rigaku RAD-IIC and Rigaku RAD-B with InP crystal monochromator.

The magnetization was measured using a vibrating sample magnetometer (VSM) with a temperature controlling attachment. The surface morphology as well as the magnetic states of the surface was observed using a Seiko Instruments Type SI-3800 AFM/MFM scanning probe micrometer.

Spectra of the polar magneto-optical Kerr rotation and ellipticity were measured between 1.2 and 4 eV at 300 K by the polarization modulation technique using PEM (photoelastic modulator).

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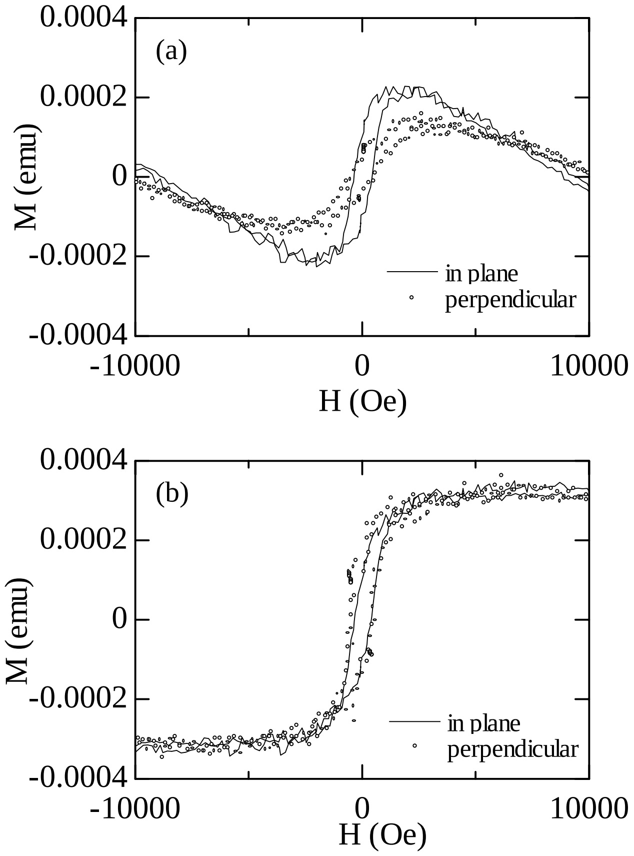

Fig.1 Magnetization curves of Mn-duffused $CdGeP_2$ layer. (a) before and (b) after correction for diamagnetic part and demagnetization field.

3. Results and discussion

3.1 Characterization of crystalline properties

Crystal structure was characterized by XRD. In the conventional θ-2θ chart, we find two sets of diffraction pattern, both of which can be indexed to chalcopyrite phase with different lattice constants. Careful investigation was carried out to confirm absence of materials with other crystal phases. No trace of any binary compounds was observed except for very small amount of GeP. Under careful XRD condition avoiding the influence of the substrate, the XRD pattern by the Mn-diffused surface layer was separately obtained. Reduction of the lattice constant of 0.8% was observed.

The EDX observation on the surface revealed that Mn / Cd ratio is as large as 0.53. This means 1/3 of Cd has been substituted by Mn atom. The Mn/Cd ratio decreased rapidly toward the depth direction and the value becomes only 0.009 at 2.5μm.

3.2 Magnetization measurements

Figs. 1(a) shows raw data of the magnetic hysteresis curves of the $CdGeP_2$:Mn system measured at room temperature (298 K). Straight lines are for in-plane magnetization, and dots for perpendicular magnetization.

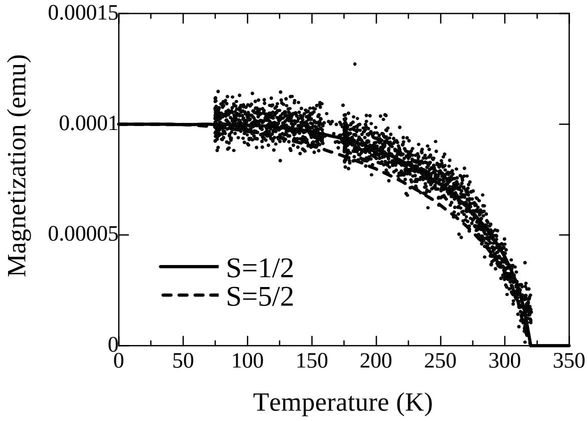

Fig. 2 Temperature dependence of magnetization in the Mn-diffused $CdGeP_2$ layer.

The curves are clearly composed of diamagnetic and ferromagnetic components. The former may be attributed to the host substrate and the latter to the new magnetic semiconductor layer. Applying suitable corrections for diamagnetism and demagnetization field, ferromagnetic hysteresis curves are obtained as shown in Fig. 1(b). The ferromagnetic component shows a well-defined hysteresis loop with the saturation field Hs about 3 kOe and coercivity Hc of about 0.4 kOe. The saturation magnetization at room temperature was 3.5×10^{-4} emu. Assuming that the deposited Mn of 30 nm in thickness on the 3×5 mm^2 surface area was completely incorporated into the host semiconductor, the magnetization per atomic unit was evaluated as 0.956×10^{-20} emu/atom, from which the gS value was determined as 1.03 μ_B.

Fig. 2 presents a plot of the remanent magnetization as a function of temperature. The curve was simulated by the molecular field theory. The best fit was obtained using the Brillouin function with S=1/2. The Curie temperature was estimated to be 320 K.

3.3 AFM and MFM measurements

Figs. 3(a) and 3(b) are AFM and MFM images, respectively, for the surface of the Mn-diffused crystal measured at room temperature. In the AFM image, a texture consisting of small crystallites is observed. The averaged size of the crystallites is 100 nm in diameter. On the other hand, stripe and circular domain patterns are observed in the MFM image. The width of the stripe domain is about 1 μm, far larger than the size of the fine texture observed in the AFM image. This suggests the magnetization is not coming from individual fine texture but is uniformly distributing over the entire surface of the crystal.

3.4 Magneto-optical measurements

Polar magneto-optical Kerr hysteresis loop of the Mn-diffused layer measured at room temperature was quite similar to the perpendicular magnetization curves shown in Fig. 1. Since Kerr loop measure the magnetization of the surface, similarity of hysteresis loop between VSM

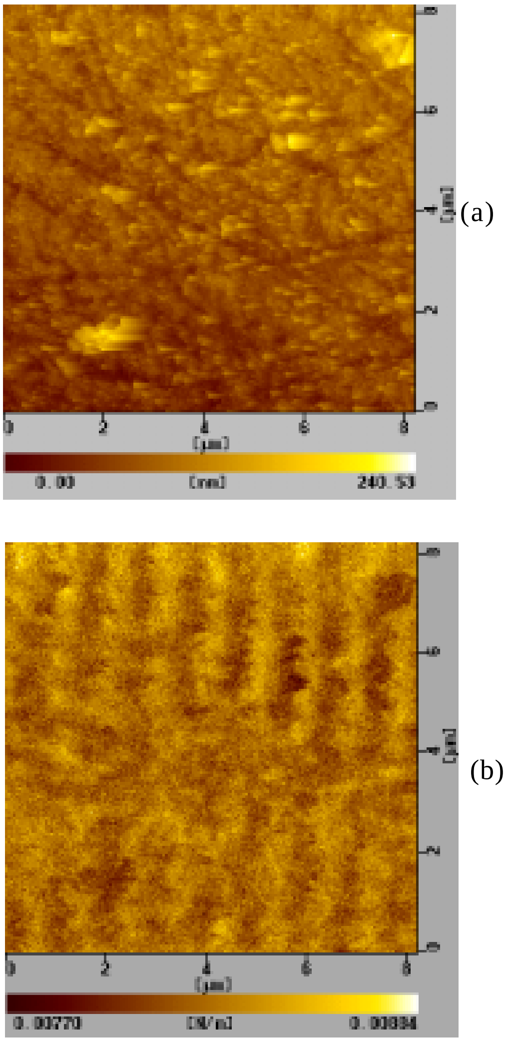

Fig. 3 (a)AFM image and (b) MFM image of Mn-diffused layer.

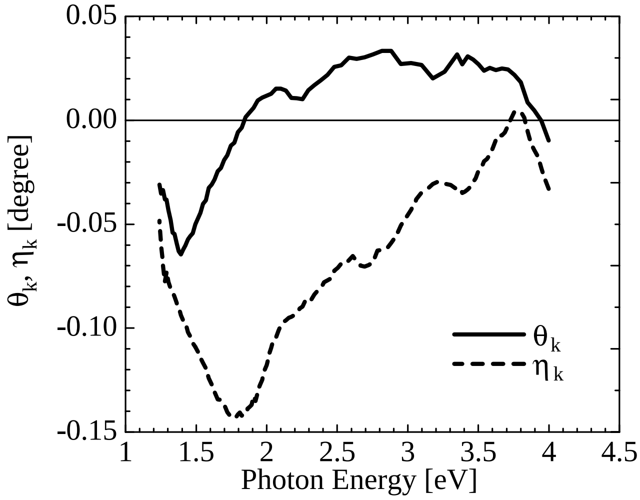

Fig. 4 Spectra of magneto-optical Kerr rotation (straight line) and Kerr ellipticity (dashed line) of the Mn-diffused layer measured at room temperature.

and Kerr measurements suggests uniformity of magnetic properties in the Mn-diffused layer. However, the magneto-optical signal suffers much noise probably due to large fluctuation of magnetic moment near *T*c.

Spectra of polar magneto-optical Kerr rotation and ellipticity measured at room temperature are shown in Fig. 4. The Kerr rotation is relatively small and is subjected to a negative peak at 1.4 eV, a zero-crossing behavior at 1.8 eV and a few peaks between 2 and 4 eV. The peak Kerr rotation value was as small as 0.065 deg.

On the other hand, Kerr ellipticity has a distinct negative peak at 1.75 eV, approaches gradually to zero towards higher energies up to 3.8 eV, where a small positive peak appears.

The energy at which the ellipticity shows a maximum coincides with the value of the energy gap of the host crystal. Relation between the complex Kerr rotation Φ_K and the complex Faraday rotation per unit length Φ_F/l can be expressed as follows: $\Phi_F / \ell = -i(\pi / \lambda)(1-\varepsilon)\Phi_K$.

Since diagonal permeability ε can be expressed as $\varepsilon = n^2$ for photon energy just below the band gap, we can assume $\varepsilon = 12.1$ using the value of $n = 3.48$ near the band edge. From $\Phi_K = -i\eta_K = -i0.12$ deg at λ=800 nm, Φ_F/l is estimated as 5.2×10^4 deg/cm. This value is comparable to the Faraday rotation of Bi-substituted magnetic garnet.

Therefore, this material has a potential as the Faraday-rotation element for optical isolators, although the care should be taken to avoid the effect of the birefringence in this material, which is characteristic of the tetragonal chalcopyrite crystal structure.

4. Conclusion

Magnetic and magneto-optical characterization was carried out in the novel magnetic semiconductor $CdGeP_2$:Mn. The Curie temperature of this material was determined as 320 K and showed a well-defined hysteresis loop at room temperature. Magneto-optical Kerr ellipticity showed a peak value of 0.14 deg around 1.8 eV, from which Faraday rotation as large as 50,000 deg/cm was estimated.

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