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Novel Quaternary Dilute Magnetic Semiconductor (Ga,Mn)(Bi,As): Magnetic and Magneto-Transport Investigations

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Abstract Magnetic and magneto-transport properties of thin layers of the (Ga,Mn)(Bi,As) quaternary dilute magnetic semiconductor grown by the low-temperature molecular-beam epitaxy technique on GaAs substrates have been investigated. Ferromagnetic Curie temperature and magneto-crystalline anisotropy of the layers have been examined by using magneto-optical Kerr effect magnetometry and low-temperature magneto-transport measurements. Post-growth annealing treatment of the layers has been shown to enhance the hole concentration and Curie temperature in the layers. A significant increase in the magnitude of magneto-transport effects caused by incorporation of a small amount of Bi into the (Ga,Mn)As layers, revealed in the planar Hall effect (PHE) measurements, is interpreted as a result of enhanced spin-orbit coupling in the (Ga,Mn)(Bi,As) layers. A two-state behaviour of the planar Hall resistance at zero magnetic field provides its usefulness for applications in non-volatile memory devices.

Keywords Magnetic semiconductors · Thin films · Raman spectroscopy · Magneto-optical Kerr effect · Magneto-resistance · Planar Hall effect

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1 Introduction

GaAs layers containing a few percent of Mn atoms became a model material among dilute magnetic semiconductors [1, 2]. When intentionally undoped, they are of p-type, where Mn atoms, substituting Ga atoms in GaAs crystal lattice, supply both mobile holes and magnetic moments. Below certain temperature (Curie temperature), the layers become ferromagnetic due to the hole-mediated ordering of Mn spins. Homogeneous layers of (Ga,Mn)As containing up to above 10 % of Mn atoms can be grown by a low-temperature (150–250 °C) molecular-beam epitaxy (LT-MBE) [3, 4]. Their magnetic properties, such as the Curie temperature and magnetic anisotropy, are sensitive to hole concentration and the growth-induced strain which allows tuning of those properties by growth and post-growth treatment conditions. Moreover, appropriate nanostructurization of thin (Ga,Mn)As layers offers the prospect of taking advantage of the magnetic domain walls in novel spintronic devices. In this context, our recent studies on several types of nanostructures patterned from ferromagnetic (Ga,Mn)As layers pointed to their utility for spintronic applications [5–7].

On the other hand, the replacement of a small fraction of As atoms by much heavier Bi atoms in GaAs results in a rapid decrease in its bandgap energy [8, 9] and in a strong enhancement of the spin-orbit coupling that is accompanied by a giant separation of the spin-split-off hole band in Ga(Bi,As) [10]. The large spin-orbit coupling could be advantageous for spintronic materials as it strongly affects their magneto-transport properties.

In order to explore this issue, we have investigated an impact of Bi incorporation into (Ga,Mn)As layers on their structural, magnetic and magneto-transport properties.

Recently, homogeneous layers with a high structural perfection of the (Ga,Mn)(Bi,As) quaternary dilute magnetic semiconductor, containing up to 6 % Mn and 1 % Bi, have been grown with LT-MBE under compressive misfit strain, on GaAs substrate [11, 12], as well as under tensile misfit strain, on (In,Ga)As buffer layer [13]. Their magnetic properties were similar to those of the ternary (Ga,Mn)As layers [14]. The easy axes of magnetization in the layers grown under compressive and tensile misfit strain were in plane and out of plane of the layer, respectively [13]. Their longitudinal magneto-resistance and the planar Hall resistance were significantly enhanced as a result of Bi incorporation [15]. In the present paper, we report on magnetic and magneto-transport properties of thin epitaxial layers of the (Ga,Mn)(Bi,As) quaternary compound grown under compressive misfit strain, proving their usefulness for possible spintronic applications.

2 Experimental

We investigated (Ga,Mn)(Bi,As) layers grown on a semi-insulating GaAs (001) substrate by an LT-MBE technique at a temperature of 230 °C. The layers were 10 nm thick and contained 6 % Mn and 1 % Bi. Reference (Ga,Mn)As layers were also grown under the same conditions. Such grown layers were subjected to the annealing in air at the temperature of 180 °C during 50 h. The post-growth annealing at temperatures below the growth temperature substantially improves magnetic and electrical properties of the layers of both of (Ga,Mn)As [16] and (Ga,Mn)(Bi,As) [11], as a result of out-diffusion of Mn interstitials. Incorporation of bismuth into the (Ga,Mn)As layers as well as the annealing-induced out-diffusion of Mn interstitials have been confirmed by the results of lattice parameter determination in the layers by means of high-resolution X-ray diffraction measurements [12].

Hole densities in the *p*-type (Ga,Mn)As and (Ga,Mn)(Bi,As) layers were estimated from Raman spectroscopy. The micro-Raman measurements were performed at room temperature using a confocal Raman microscope and a 532-nm semiconductor laser as an excitation source. Magnetic properties of the layers were studied using magneto-optical Kerr effect (MOKE) magnetometry. The MOKE experiments were performed in longitudinal geometry using He-Ne laser as a source of linearly polarized light. The laser spot was about 0.5 mm in diameter, and the angle of light incidence on the sample was about 30°. We used a standard lock-in technique with a photo-elastic modulator operating at 50 kHz and a Si-diode detector. Magnetic hysteresis loops were measured in the temperature range 6−150 K, in magnetic fields up to 2 kOe oriented in the plane of the layers [13]. Magneto-transport measurements were carried out on Hall bars aligned along the [1̄10] crystallographic direction, having 100 μm width and 200 μm distance between the voltage contacts. The Hall bars were prepared by means of electron-beam lithography patterning and chemical etching. Both the longitudinal resistance, R_{xx}, and the planar Hall resistance, R_{xy}, were measured simultaneously using a low-frequency (18 Hz) lock-in technique. The measurements were performed for various orientations of the in-plane magnetic field (H) at liquid helium temperature.

3 Results and Discussion

We have estimated the low-temperature hole concentrations in the layers from the micro-Raman spectra recorded from the (001) surface in the backscattering configuration [11, 17]. They were 8.5×10^{19} and 7×10^{19} cm^{-3} in the as-grown (Ga,Mn)As and (Ga,Mn)(Bi,As) layers, respectively, and increased after the annealing to 1.6×10^{20} and 1×10^{20} cm^{-3}, respectively. These values are in a quite good agreement with ones obtained from the Hall effect measured at room temperature.

We have recorded the magnetization hysteresis loops for the (Ga,Mn)As and (Ga,Mn)(Bi,As) layers by the MOKE measured as a function of magnetic field. Nearly rectangular loops, obtained for the both materials, evidence for the in-plane magnetization. A detailed analysis of the obtained hysteresis loops indicated easy magnetization axes along the in-plane $\langle 100 \rangle$ directions and hard axes along two magnetically non-equivalent in-plane $\langle 110 \rangle$ directions at low temperatures. The [1̄10] direction was magnetically easier than the perpendicular [110] one. That rather complicated magneto-crystalline anisotropy is a characteristic of (Ga,Mn)As layers grown under a compressive misfit strain [6, 14]. From temperature dependence of the amplitudes of hysteresis loops, shown in Fig. 1, we have determined the ferromagnetic Curie temperatures to be about 80 and 63 K for the as-grown (Ga,Mn)As and (Ga,Mn)(Bi,As) layers, respectively, which increased to the values of 87 and 82 K, respectively, as a result of the annealing.

Electrical resistance of ferromagnetic materials depends on the angle (θ) between the directions of current flow and the magnetization vector, resulting in anisotropic magneto-resistance (AMR). In a single-domain sample, the AMR components are [18]

$$R_{xx} = R_{\perp} + (R_{\parallel} - R_{\perp}) \cos^2 \theta \qquad (1)$$

$$R_{xy} = 1/2(R_{\parallel} - R_{\perp}) \sin 2\theta \qquad (2)$$

where $R_{\perp}$ and $R_{\parallel}$ are the resistances for the in-plane magnetization vector oriented perpendicular and parallel to the
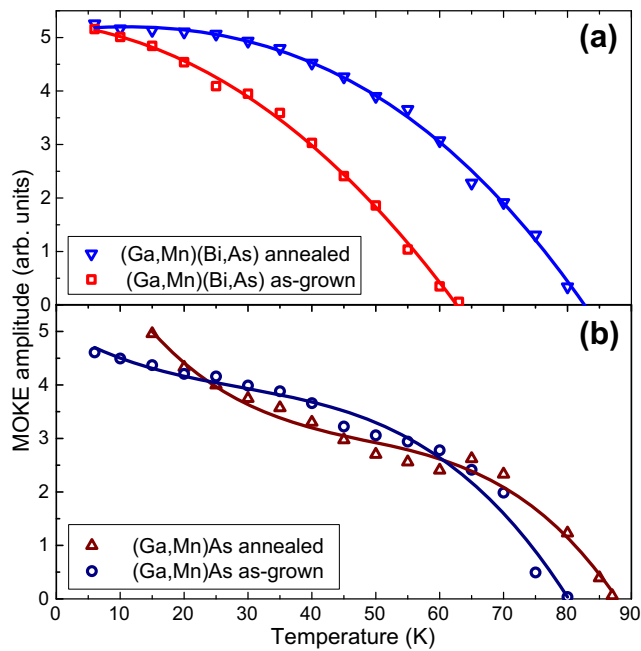
Fig. 1 Temperature dependences of the amplitudes of magnetization hysteresis loops recorded at various temperatures with MOKE magnetometry under an in-plane magnetic field along the [100] crystallographic direction (points) for both the as-grown and annealed (Ga,Mn)(Bi,As) (**a**) and (Ga,Mn)As (**b**) layers. The *solid lines* act as a guide to the eye

Fig. 2 Relative longitudinal resistance (R_{xx}) for the Hall bars of (Ga,Mn)(Bi,As) and (Ga,Mn)As layers measured at the temperature of 4.2 K, while sweeping an in-plane magnetic field, parallel to the current, in opposite directions, as indicated by *arrows*. The curves have been vertically offset for clarity

current, respectively. Equation (1) describes the longitudinal MR, while (2) the transverse resistance or the so-called planar Hall effect (PHE) [19]. PHE manifests itself as a spontaneous transverse voltage that develops, owing to the spin-orbit interaction, in response to longitudinal current flow under an in-plane magnetic field or even in the absence of a magnetic field. In ferromagnetic (Ga,Mn)As, in contrary to metallic ferromagnets, $(R_\parallel - R_\perp) < 0$ [20]. The same also holds for (Ga,Mn)(Bi,As), as shown in our previous paper [15]. Importantly, the magnitude of PHE in (Ga,Mn)As layers is up to 4 orders of magnitude greater than that previously found in metallic ferromagnets, which is called the giant PHE [20]. A very large magnitude of PHE observed in (Ga,Mn)As results primarily from the combined effects of a strong spin-orbit interaction in the valence band of the crystal with zinc blende structure and the large spin polarization of holes in (Ga,Mn)As.

The normalized longitudinal MR for the Hall bars of the annealed (Ga,Mn)As and (Ga,Mn)(Bi,As) layers, measured under magnetic field parallel to the current, is presented in Fig. 2. The up-and-down magnetic field sweep in the range of ± 1 kOe results in non-monotonic behaviour at low fields, which follows (1), superimposed on isotropically negative MR extended to higher fields. Negative MR is a common property of ferromagnetic metals resulting from the reduction of spin-disorder scattering of charge carriers due to the
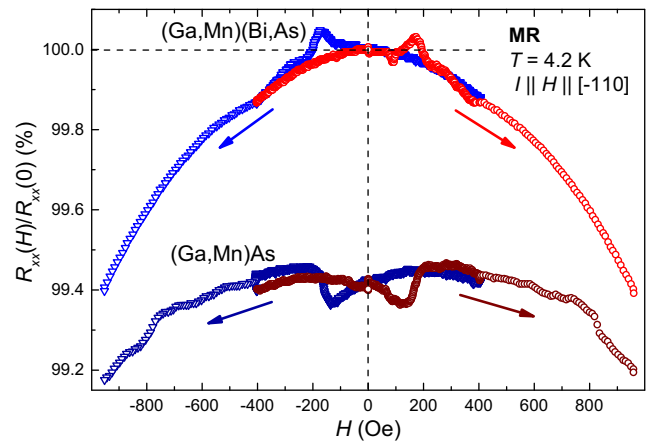
alignment of magnetic ion spins in an external magnetic field. Additionally, in (Ga,Mn)As, at the lowest temperatures, when the spins are fully ferromagnetically ordered, the negative MR can result mainly from the magnetic field-induced suppression of quantum interference contribution to the resistivity caused by the effect of weak localization [21]. A distinctly larger magnitude of the negative MR observed for the (Ga,Mn)(Bi,As) layer, shown in Fig. 2, likely results from stronger spin-orbit coupling caused by Bi incorporation into the (Ga,Mn)As crystal lattice.

Figure 3 presents the planar Hall resistance for both layers measured under magnetic field perpendicular to the current. While sweeping the magnetic field in the range of ± 1 kOe, R_{xy} displays the double hysteresis loop with
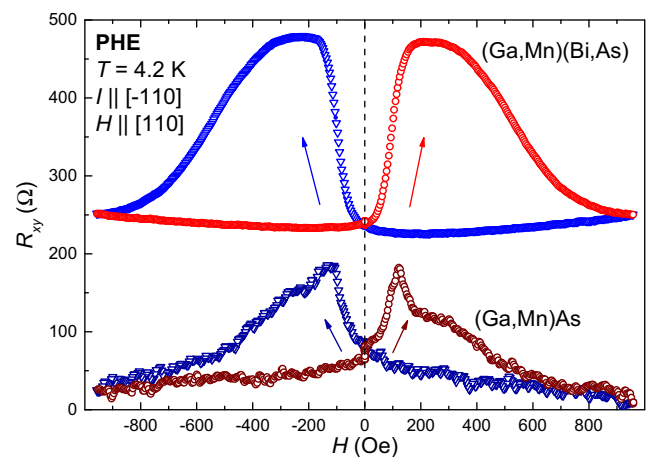

Fig. 3 Planar Hall resistance (R_{xy}) for the Hall bars of (Ga,Mn)(Bi,As) and (Ga,Mn)As layers measured at the temperature of 4.2 K, while sweeping an in-plane magnetic field, perpendicular to the current, in opposite directions, as indicated by *arrows*. The curves have been vertically offset for clarity
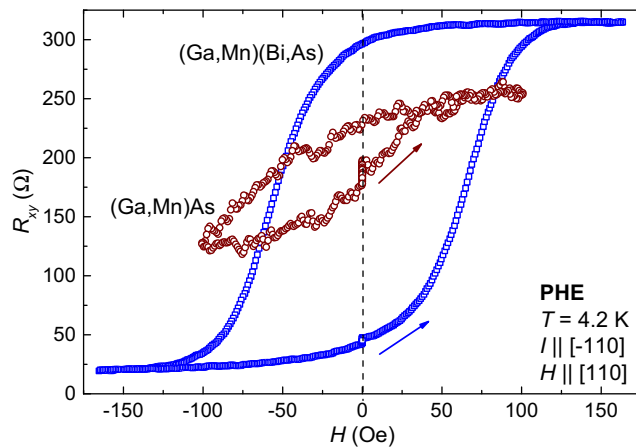
Fig. 4 The same as in Fig. 3 but for a narrower range of sweeping magnetic field

recurrence points at magnetic fields where the magnetization vector in the layer is forced to be directed along the external magnetic field. The planar Hall resistance changes between two extremum values, which, according to (2), are obtained at the angles (θ) of 45°, 135°, 225°, and 315°, corresponding to the orientations of the magnetization vector along the in-plane $\langle 100 \rangle$ crystallographic directions. The up-and-down magnetic field sweep in the whole range causes a rotation of the magnetization vector by 360° between all the four in-plane $\langle 100 \rangle$ directions that represent equivalent easy axes of magnetization defined by cubic magneto-crystalline anisotropy.

In turn, while sweeping a magnetic field in a narrow range of ±200 Oe, R_{xy} displays a single hysteresis loop analogous to the usual magnetization hysteresis loop, as shown in Fig. 4. Here, the magnetic field scan results in 90° rotations of the magnetization vector between two in-plane $\langle 100 \rangle$ directions and the results are consistent with the expected single-domain structure of the Hall bar. This planar Hall resistance can assume one of the two stable values at zero magnetic field, depending on the previously applied field, which can be exploited in non-volatile memory elements. All the results presented in Figs. 2, 3, and 4 demonstrate much larger magnitudes of both the negative longitudinal MR and the PHE resistances in the (Ga,Mn)(Bi,As) layer as compared with that of the (Ga,Mn)As one. This is a manifestation of a large enhancement of the spin-orbit interaction upon the addition of Bi into the GaAs layers.

4 Conclusions

Homogeneous layers of the (Ga,Mn)(Bi,As) quaternary dilute magnetic semiconductor have been grown by the low-temperature MBE technique on GaAs substrate. A small amount of Bi incorporated into (Ga,Mn)As significantly enhances the strength of spin-orbit coupling in this material, which manifests itself in its magneto-transport properties as an increase in both the magnitude of negative MR and the PHE resistance. This makes the novel dilute magnetic semiconductor advantageous for spintronic applications. Moreover, we have demonstrated that the PHE can be very useful in the study of magnetic properties of ferromagnetic semiconductors and their structures. The PHE can be also used as a basis for a new type of a non-volatile memory elements, in which a bit of information is written magnetically and read electrically.

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