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Magnetic and magnetotransport properties of new III-V diluted magnetic semiconductors: GaMnAs

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We have studied magnetic and magnetotransport properties of novel III-V diluted magnetic semiconductors, $(Ga_{1-x}Mn_x)As$. The GaMnAs thin films were grown on GaAs(001) substrates by low temperature molecular beam epitaxy. We present magnetoresistance, extraordinary Hall effect, and $M-H$ characteristics of two $(Ga_{1-x}Mn_x)As$ samples having different Mn content x. [S0021-8979(97)41208-2]

I. INTRODUCTION

Molecular beam epitaxy (MBE) has made it possible to grow a variety of new heterostructures and alloy semiconductors having novel properties. Among them, diluted magnetic semiconductors (DMSs) generated much interest, since they can combine the properties of both semiconductors and magnetic materials. Traditionally, II-VI based DMSs were extensively studied,[1] because some magnetic ions (Mn^{2+}, Fe^{2+}) are easily incorporated into II-VI compounds by substituting group II atoms. On the other hand, the study of III-V based DMSs was very limited, and detailed investigations were done only on InMnAs.[2,3] Very recently, we have successfully grown a new type of III-V DMS, GaMnAs,[4,5] based on GaAs which is the most widely used III-V semiconductor in high speed electronics and optoelectronics. One of the advantages of III-V based materials over II-VI is that one can study transport properties, because doping control is far more difficult in II-VI than in III-V. This article presents the magnetic and magnetotransport properties of GaMnAs thin films grown by MBE on GaAs.

II. EPITAXIAL GROWTH AND STRUCTURAL PROPERTIES

MBE growth of $(Ga_{1-x}Mn_x)As$ thin films on semi-insulating (001) GaAs substrates was performed with our ULVAC MBC-508 system using solid sources of Ga, As, and Mn.[4] Although the equilibrium solubility of Mn atoms in GaAs is quite low ($\sim 10^{19}$ cm^{-3}), $Ga_{1-x}Mn_xAs$ films with x up to 0.078 were grown at 200–300 °C under As rich conditions. This low temperature MBE (LT-MBE), which realizes strong nonequilibrium growth conditions, allows a large amount of Mn atoms to be incorporated into the host GaAs lattice far above its solubility. Mn content x was determined from the ratio of the Mn flux to the Ga flux and corrected by electron probe micro analyzer (EPMA) measurements after the growth. We obtained homogeneous GaMnAs ternary alloys when the Mn content x and growth temperature T_g were relatively low ($x \leqslant 0.078$, $T_g \leqslant 300$ °C). In contrast, when the Mn content x and growth temperature were relatively high, MnAs clusters were formed in GaAs. X-ray diffraction measurements revealed that the crystal structure of the homogeneous GaMnAs films was zinc-blende and the lattice constants were slightly larger than that ($a_0 = 0.56533$ nm) of GaAs. Considering the compressive strain in the GaMnAs thin films on GaAs, the intrinsic lattice constants a of cubic $Ga_{1-x}Mn_xAs$ follow the Vegard's law; $a/a_0 = 1 + cx$ ($c = 4.04 \times 10^{-2}$). This indicates that the GaMnAs films are very uniform alloys.

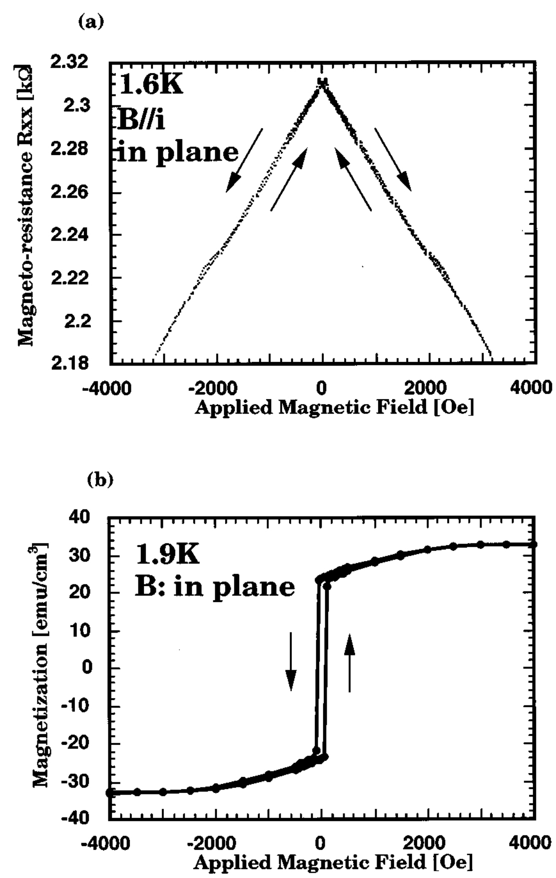

FIG. 1. (a) Magnetoresistance ($R_{xx} - H$) at 1.6 K, and (b) magnetization ($M - H$) at 1.9 K of $Ga_{1-x}Mn_xAs$ ($x = 0.074$) when the magnetic field is applied in-plane (parallel to the current).

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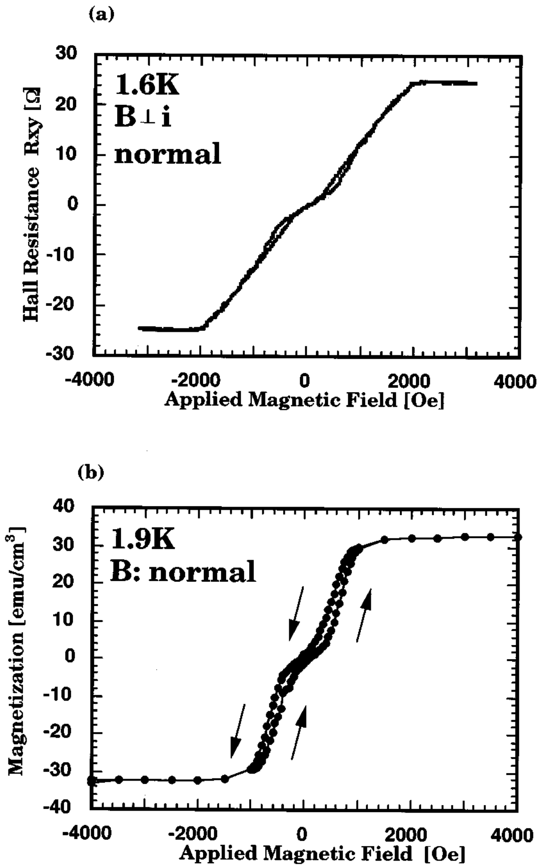

FIG. 2. (a) Hall resistance ($R_{xy}-H$) at 1.6 K, and (b) magnetization ($M-H$) at 1.9 K of $Ga_{1-x}Mn_xAs$ ($x=0.074$) when the magnetic field is applied perpendicular to the plane.

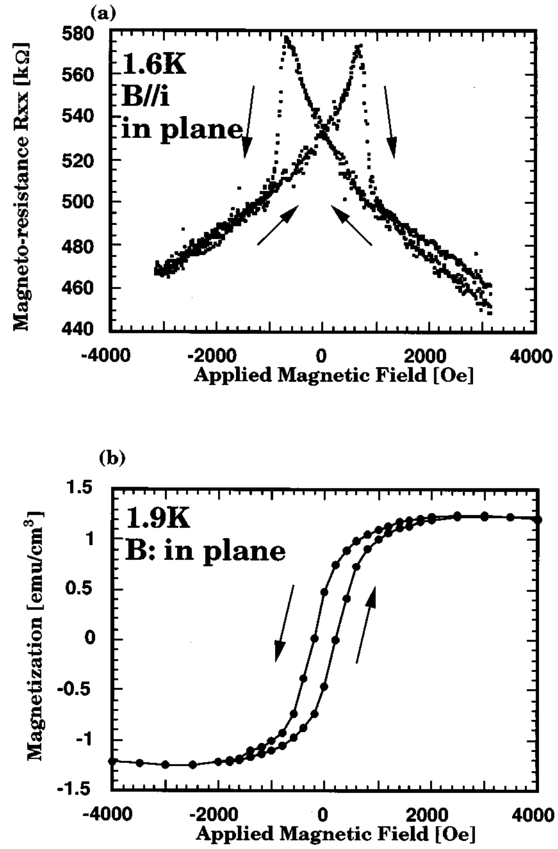

FIG. 3. (a) Magnetoresistance ($R_{xx}-H$) at 1.6 K, and (b) magnetization ($M-H$) at 1.9 K of $Ga_{1-x}Mn_xAs$ ($x=0.005$) when the magnetic field is applied in-plane (parallel to the current).

III. MAGNETIC AND MAGNETOTRANSPORT PROPERTIES

All the GaMnAs ($x=0.005$–0.078) samples we have grown showed p-type conduction with the hole concentration of 3.8×10^{17} cm^{-3}–2.58×10^{20} cm^{-3}, and no ferromagnetic behavior was observed at room temperature.

Here, we investigated low temperature magnetization and magnetotransport properties of two homogeneous $Ga_{1-x}Mn_xAs$ thin films with different Mn composition, $x=0.074$ and $x=0.005$. Transport measurements were performed on photolithograthically patterned Hall bars with a width and a length of 200 μm and 1.3 mm. Magnetization measurements were done using a superconducting quantum interference device (SQUID).

A. GaMnAs ($x=0.074$)

First, we show the results of the homogeneous GaMnAs ($x=0.074$) film with a thickness of 1.4 μm. The hole concentration of this sample at room temperature was 2.58×10^{20} cm^{-3}, and the mobility was 7.24 cm^2/V s. Figure 1(a) shows the magnetoresistance R_{xx} measured at 1.6 K, when the magnetic field was applied in-plane parallel to the current. As shown in this figure, negative magnetoresistance with hysteretic behavior was observed, suggesting the presence of ferromagnetic ordering at low temperature. Two peaks are observed at low field ($\sim\pm50$ Oe) and the hysteretic loop is closed entirely at about 2500 Oe. It is interesting to compare with the results of magnetization ($M-H$) measurements using SQUID when the magnetic field was applied in-plane at 2.0 K, as shown in Fig. 1(b). A square hysteresis was recorded at low field, with the coercive field H_c of 56 Oe, and remnant magnetization M_r of 23.7 emu/cm^3. Magnetization was saturated at 2500$\sim$3000 Oe. In the range of $H<\sim3000$ Oe, the $M-H$ curve corresponds to the magnetoresistance curve very well. The two peaks in the $R_{xx}-H$ curve correspond to the reversal of magnetization at $\sim$50 Oe, and the completion of the hysteresis in the $R_{xx}-H$ corresponds to the saturation field at $\sim$2500 Oe. At even higher field (3000 Oe$\leqslant H \leqslant$4 T), we observed negative magnetoresistance in $R_{xx}-H$ characteristics while magnetization is saturated. We also measured the temperature dependence of remnant magnetization, and estimated the Curie temperature of this sample to be 55–60 K.

Next, we performed Hall measurements of this sample at low temperature (1.6 K), as shown in Fig. 2(a). The Hall resistance of ferromagnetic materials generally satisfies the following equation.

$$R_{xy}=(R_0/d)B+(R_s/d)M,$$

where R_0 is the ordinary Hall constant, R_s the extraordinary Hall constant, and d the sample thickness. Here the second term is the extraordinary Hall effect, which is caused by anisotropic scattering between the carriers and local mag-

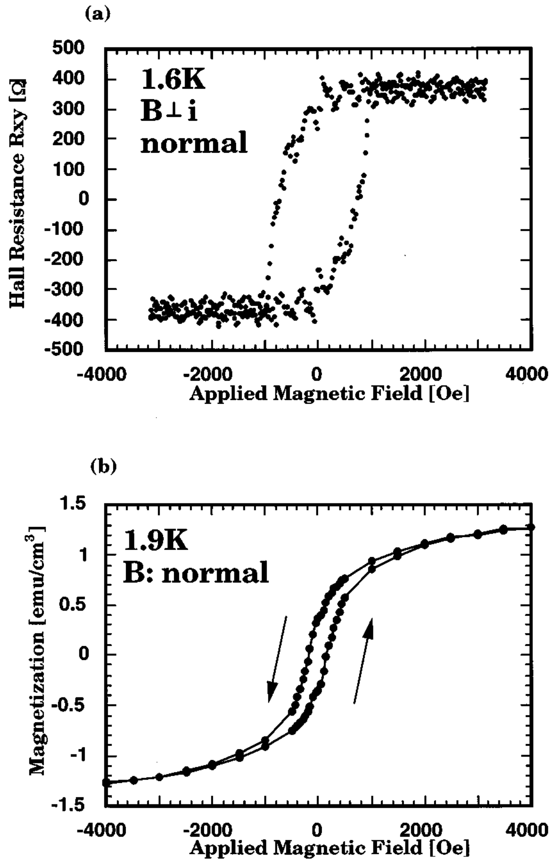

FIG. 4. (a) Hall resistance ($R_{xy}-H$) at 1.6 K, and (b) magnetization ($M-H$) at 1.9 K of $Ga_{1-x}Mn_xAs$ ($x=0.005$) when the magnetic field is applied perpendicular to the plane.

netic moments of Mn atoms. Since the carrier (hole) concentration is quite high in our sample, the first term is negligible compared with the second term. Therefore, the Hall resistance R_{xy} is proportional to the perpendicular component of magnetization of the sample. Figure 2(b) shows a $M-H$ curve measured by SQUID at 1.9 K when the field is applied perpendicular to the film plane. The $M-H$ and $R_{xy}-H$ characteristics are similar, only a small hysteresis is seen and the magnetic field to fully saturate the sample is ∼2000 Oe. This indicates that the easy axis of magnetization of this sample lies in the plane.

B. GaMnAs ($x=0.005$)

Figures 3(a) and 3(b) show the magnetoresistance and magnetization, respectively, of the homogenous GaMnAs ($x=0.005$) film with a thickness of 1.0 μm when the magnetic field was applied in-plane parallel to the current. The hole concentration of this sample was 1.93×10^{19} cm^{-3} and the mobility was 10.6 cm^2/V s at room temperature. This sample also shows ferromagnetic ordering at low temperature. However, the magnetization of this sample ($M_r=0.7$ emu/cm^3, $M_s=1.3$ emu/cm^3) and its Curie temperature T_c, which was estimated to be about 10 K, are much lower than those of the previous GaMnAs sample ($x=0.074$).

We also measured the Hall resistance R_{xy} of this sample at 1.6 K [Fig. 4(a)], and magnetization at 1.9 K [Fig. 4(b)], with the magnetic field applied perpendicular to the plane. Both graphs show clear hysteresis, and, especially the $R_{xy}-H$ curve, have a relatively larger Hc of ∼600 Oe, indicating that this sample has some perpendicular component of magnetization. The difference in the coercivity Hc between the $R_{xy}-H$ [Fig. 4(a)] and the $M-H$ [Fig. 4(b)] is due to the slight difference in the measurement temperature. We found that the value of Hc increases with decreasing temperature in this range (around 2 K). Comparing the $M-H$ curves of Fig. 3(b) and Fig. 4(b) with different directions of the applied field, the $M-H$ characteristics look similar to each other, indicating the lack of magnetic anisotropy.

IV. SUMMARY

We have studied magnetic and magnetotransport properties of new GaAs based diluted magnetic semiconductors (Ga,Mn)As grown by low temperature MBE. All the GaMnAs samples showed p-type conduction, exhibiting ferromagnetic ordering at low temperature. The sample with $x=0.074$ shows relatively intense magnetization ($M_r=23.7$ emu/cm^3) and higher Curie temperature (T_c∼60 K), having well aligned in plane magnetic anisotropy. In contrast, the GaMnAs sample with $x=0.005$ shows much weaker magnetization ($M_r=0.7$ emu/cm^3) and lower Curie temperature (T_c∼10 K) with the presence of perpendicular magnetization.

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