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Peculiar Magnetic Properties at Low Temperature of MnAs Thin Films Grown on GaAs(001) Substrate

Yongsu Kwak, Jinyoung Maeng, and Jonghyun Song*

Department of Physics, Chungnam National University, Daejeon 34134, Republic of Korea

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We carried out investigation on the magnetic properties of MnAs thin film grown on GaAs(001) substrate by using molecular-beam epitaxy. The magnetic Curie temperature was ~340 K which was higher than that of the bulk. Magnetization (*M*) versus magnetic field (*H*) showed strong two-fold symmetric anisotropy. At low temperature of 2 K, the *M-H* curve and temperature dependence of magnetization exhibit peculiar behavior that has not been observed before. It should be noted that these unique results were only observed when a magnetic field was applied. We also observed a weak-localization-like magnetoresistance (MR) at 2 K. As an origin of the observed peculiar magnetic properties and MR, we suggest diamagnetic domains which are phase transited from ferromagnetism at low temperature.

Keywords : MnAs, thin film, ferromagnetism, *M-H* curve, weak-localization

1. Introduction

Spintronics devices that utilize the spin degrees of freedom of charge carriers are attracting attention in the hope of overcoming the limitations of conventional semiconductor-based electronics [1]. For such spintronics devices, the spin of carrier should be sufficiently maintained to the 'spin-polarized' state during the process in the devices before relaxation to the randomly directed state [2]. Injection of spin carriers directly into semiconductor materials with a high efficiency is one way to realize such spin-polarized current state in the host semiconductor materials [3-7]. This method is advantageous due to the simplicity that arises from just adhesion of adjacent ferromagnetic layer as a spin-injector. As a candidate of such ferromagnetic spin aligner material, MnAs is promising due to its high magnetic Curie temperature (T_c) of 313~318 K (in the bulk state) [8, 9]. Particularly for the GaAs and InAs hosts, MnAs has an another advantage because it has a common element of As that can increases the efficiency of spin-injection.

MnAs thin films when grown on GaAs(001) or Si(001) substrates are classified to type-A and type-B depending on their crystal orientation. For the type-A, the hexagonal pillar is lying on the substrate surface. That is, the MnAs[0001] axis is parallel to the in-plane direction of the film surface whereas it is angled in type-B [10]. The MnAs thin films of type-B are classified again into type-I and type-II depending on the domain structure: the type-I consists of a single-domain while the type-II has dual domains that rotate 90 degrees from each other. The type of crystal structure of MnAs thin film is determined by growth conditions such as the growth temperature during growth or the way of ejection of As flux before growth [10]. The magnetic characteristics are known to strongly depend on the crystal structure and orientation [11]. In this study, we report on the peculiar behavior of magnetization at low temperature, which has not been reported so far. We also discuss the correlation between observed magnetic properties and electrical-transport, which is believed to provide useful information for MnAs thin film device applications.

2. Experimental

MnAs thin film with a thickness of ~21 nm was grown on "epi-ready" insulating non-doped GaAs(001) substrate using a standard solid-source molecular-beam epitaxy (MBE), equipped with a homemade As cracker cell. The oxide-free GaAs substrate was prepared by etching in BHF solution before introducing in the growth chamber.

*Corresponding author: Tel: +82-42-821-5455
Fax: +82-42-822-8011, e-mail: songjonghyun@cnu.ac.kr

During the growth, the surface structure and thickness were monitored by reflection high-energy electron diffraction (RHEED). Before the growth, the substrate was heated to 700 °C to drive off any surface oxide, followed by the lowering to target growth temperature. Following the growth, crystal structure was characterized *ex situ* by using four-circle x-ray diffractometer. Magnetic properties of the sample were measured using a Magnetic Property Measurement System (MPMS) with SQUID magnetometer. Magnetic field dependent resistivity measurements were performed in Hall-bar patterned configuration.

3. Results and Discussion

We grew MnAs thin film on oxide free GaAs(001) substrate. The growth temperature was kept to 350 °C during the growth. To figure out the correlation between the crystal structure and magnetic property, we carried out structural characterization by x-ray diffraction (XRD) measurements. Figure 1(a) shows a θ-2θ scan for the scattering vector directed along the surface normal out-of-plane direction. The peak at $2\theta = 31.9°$ is from the MnAs($\bar{1}101$) reflection, while the additional small peaks at $2\theta = 32.2°$ and $2\theta = 31.5°$ are interpreted to be due to the total thickness of the film; thickness fringes. The total thickness, extracted from these thickness fringes is ~31 nm, which is similar to that estimated from the RHEED intensity and thickness monitor measurements during the growth. The appearance of the peak at $2\theta = 31.9°$ indicates that the MnAs thin film studied here is type-B with a crystal orientation relation of MnAs[$\bar{1}\bar{1}20$]// GaAs[$\bar{1}10$] and MnAs[$1\bar{1}02$]//GaAs[110] as sketched in Fig. 1(a) [10]. We also measured θ-2θ scan in the wide range of 2θ from 20° to 40° and confirmed the synthesis of only type-B MnAs thin film without any secondary phase. Figure 1(b) shows a θ-rocking curve for the MnAs($\bar{1}101$) reflection. The observed full width half maximum (FWHM) of ~0.15 degree shows a highly qualified crystal structure of MnAs thin film, even at a relatively thin thickness of ~31 nm.

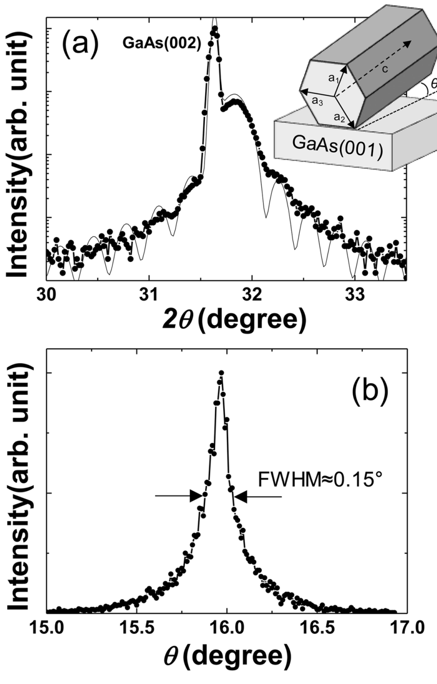

Fig. 1. Structural characterization by XRD of type-B MnAs thin film grown on GaAs(001) substrate. (a) θ-2θ scans for the reflection from the MnAs($10\bar{1}1$) planes. (b) θ-rocking curve for the reflection from the MnAs($1\bar{1}01$) planes.

Figure 2 shows the measured magnetic properties of

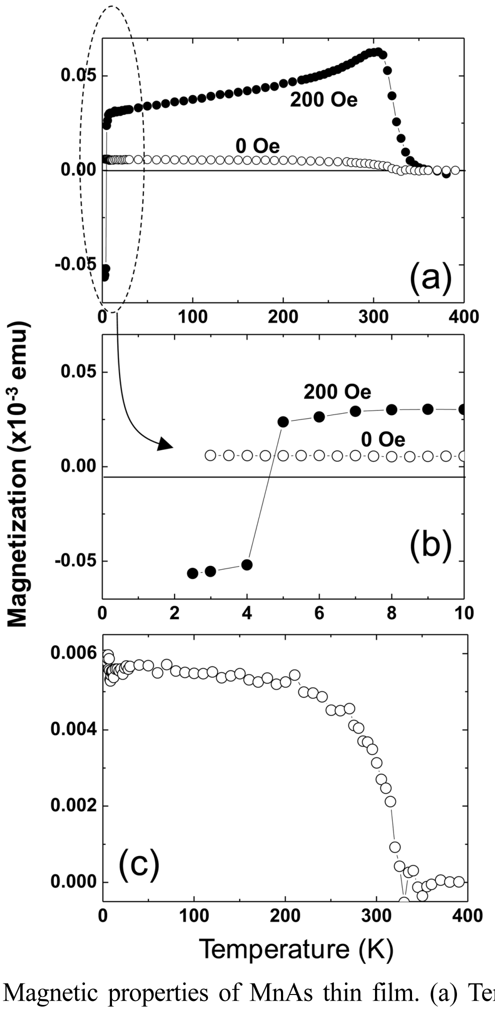

Fig. 2. Magnetic properties of MnAs thin film. (a) Temperature dependences of magnetization measured in zero-cooled-field mode when the magnetic field of 200 Oe was applied and also no magnetic field was applied. The magnetic field was applied along the GaAs[$\bar{1}10$]) direction. (b) Zoomed plot of (a) in a low temperature range. (c) The zoomed temperature dependence of magnetization without an applied magnetic field which is plotted in (a).

MnAs thin film studied here. As shown in Fig. 2(a), the zero-field-cooled temperature dependence of magnetization with an applied magnetic field of 200 Oe along the GaAs[$\bar{1}$10] direction exhibits a magnetic Curie temperature of ~340 K, the higher value compared to that of the bulk (~318 K). In particular, as shown in Fig. 2(b), a unique behavior that has never been observed was measured in the low temperature range: the magnetization at below $T < 5$ K was drastically switched to negative with decreasing the temperature. However, the remnant magnetization measured without applying a magnetic field did not show a such sudden change in magnitude as plotted in Figs. 2(b) and 2(c) in the same temperature range. These observations imply a possibility of partly reversed ferromagnetic state in the MnAs thin film.

To investigate and get more insight on the magnetic properties, we measured magnetic field dependence of magnetization. Figure 3 displays magnetization (***M***) versus magnetic field (***H***) (***M-H***) curves measured at different temperatures and configurations. As shown in Fig. 3(a), all ***M-H*** curves exhibit strong anisotropy: when a magnetic field was applied along the GaAs[$\bar{1}$10] direction, the magnetization was easily saturated at ***H*** ~ 0.1T whereas it was saturated at ***H*** ~ 1.0T when the field was directed along the GaAs[110]. These measurements determine the magnetic easy and hard axes to the GaAs[$\bar{1}$10](MnAs[$\bar{1}\bar{1}$20]) and GaAs[110](MnAs[1$\bar{1}$02]), respectively. This strong magnetic anisotropy was observed at the entire temperature range below the Curie temperature as shown in Figs. 3(b) and 3(d). Similar shaped ***M-H*** curves were observed over the temperature range of $T \geq 10$ K, but attention should be paid to the peculiar response of magnetization revealed at low temperature of 2 K, as plotted in Figs. 3(b) and 3(d): unlike the ***M-H*** curves at T=10 K, the rapidly suppressed magnetization near 0.05T recovers and increases as the magnetic field decreases to negative, eventually forming a zigzag shape as shown in Fig. 3(c). A similar zigzag shape was also observed when the magnetic field was applied along the GaAs[110] direction. Considering the magnetization response on the

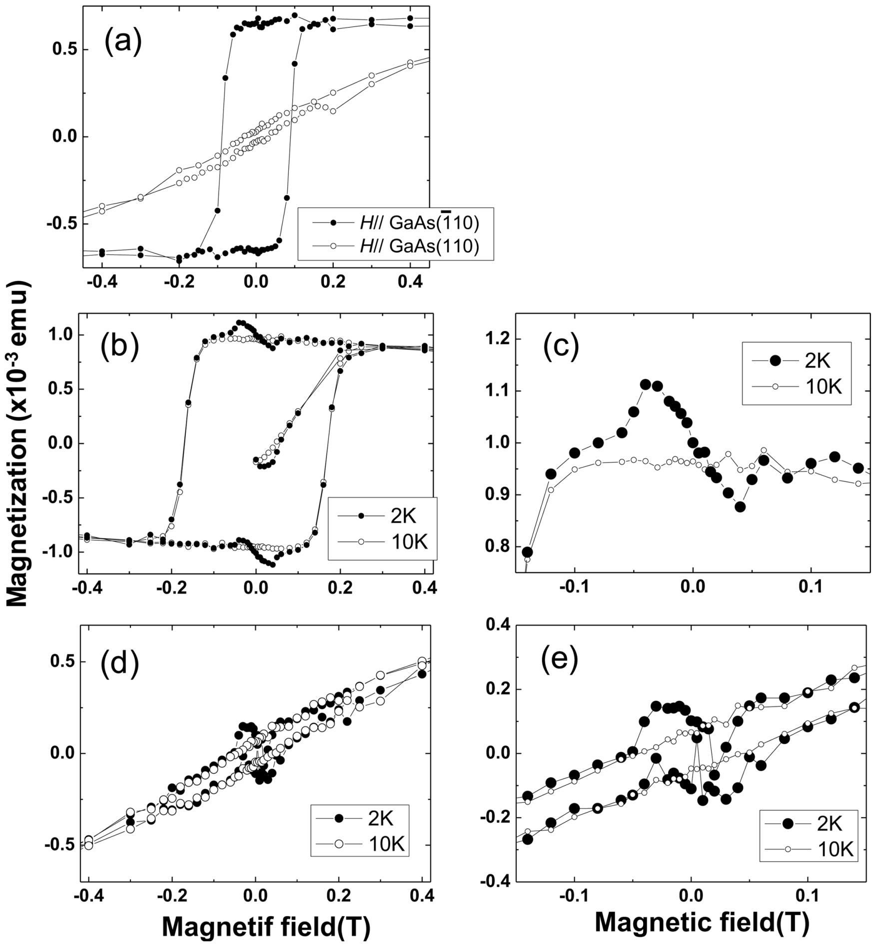

Fig. 3. ***M-H*** curves of MnAs/GaAs(001) thin film measured at (a) T = 300 K, and (b), (d) T = 10 K, 2 K. The external applied magnetic field was applied along the (b) GaAs[$\bar{1}$10] and (d) GaAs[110] directions. (c) and (e) are zoomed plots of (b) and (d), respectively.

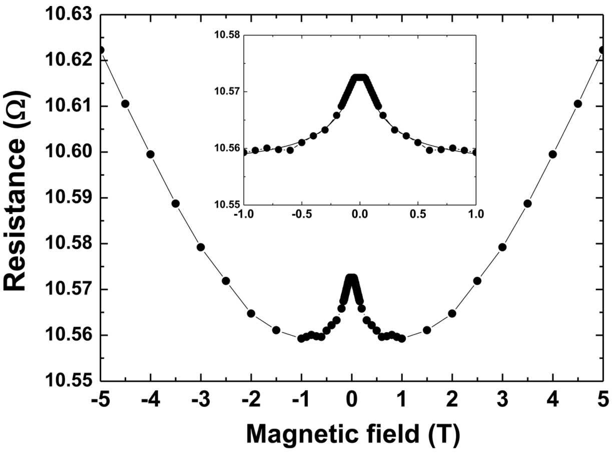

Fig. 4. Magnetoresistance of MnAs/GaAs(001) thin film measured at 2 K. The applied magnetic field was along the surface normal out-of-plane direction. The Hall resistance was removed from the original data. Inset shows the fitted data with weak-localization by using the parameter $B_{SO} = 0$, $B_\varphi = 30$ and $B_e =$ 50 mT where B_{SO} is the spin-orbit characteristic field, B_φ is the phase coherence characteristic field, and B_e is the elastic characteristic field in Hikami-Larkin-Nagaoka equation.

magnetic field observed near 0T, these behaviors can be interpreted due to the formation of diamagnetic domains randomly embedded in the ferromagnetic MnAs thin film. These diamagnetic domains appear to return to ferromagnetism when the applied magnetic field exceeds 0.05T since the ***M-H*** curve closely follows the trace measured at 10 K in the magnetic field range of $|\boldsymbol{H}| > 0.05$T. Although it is not possible to figure out the exact composition of the diamagnetic domains in the current, we propose meta-stable type-B MnAs particles, recalling the fact that the measured magnetization returns to ferromagnetism when the applied magnetic field exceeds 0.05T. These observations are consistent to the temperature dependence of magnetization discussed in Fig. 2. The fact to note is that the behavior of such magnetization fluctuations is only observed when a magnetic field is applied as shown in Figs. 2(b) and 2(c). This fact supports meta-stable diamagnetic domain formation scenario.

The magnetoresistance (MR) with an applied magnetic field along the out-of-plane to the sample surface is plotted in Fig. 4. As shown, the MR behavior, which may be due to weak localization, was exhibited at low temperature of 2 K: in the low magnetic field range, the resistance decreases as the magnetic field increases [12-14]. In high magnetic fields, the resistance increases rapidly and it is believed to has reached the classical limit. This weak-localization-like MR behavior at low temperature is differentiated to the typically observed negative MR at high temperature not shown here. As we know, weak-localization is a quantum interference effect, which occurs in the randomly disordered system. Interpreting the diamagnetic domains formed at low temperatures into a randomly disordered system, the peculiar behavior of MR is consistent with the discussions on the temperature dependence of magnetization and ***M-H*** curves in Figs. 2 and 3.

4. Conclusions

We grew MnAs thin film on GaAs(001) substrate and studied its magnetic and electrical-transport properties in detail. The MnAs thin film studied here shows a type-B crystal orientation with a highly qualified crystallinity. The Curie temperature was ~340 K, which was higher than that of the bulk. Magnetization shows two-fold symmetry with the easy(hard)-axis of magnetization along the GaAs[$\bar{1}$10](GaAs[110]). The temperature dependence of magnetization was observed to decrease suddenly to negative below $T < 5$ K. In addition, we observed peculiar fluctuations of magnetization in the ***M-H*** curves measured at low temperature of 2 K. For these anomalous magnetic response to magnetic fields, we suggest the formation of diamagnetic domains. The MR observed at 2 K is consistent with this scenario.

Acknowledgment

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