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HTML ABSTRACT + LINKS

Existence and atomic arrangement of microtwins in hexagonal MnAs ferromagnetic epilayers grown on GaAs (100) substrates with monolayer InAs Buffers

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Selected-area electron-diffraction pattern (SADP) and high-resolution transmission electron microscopy (HRTEM) measurements were carried out to investigate the existence and the atomic arrangement of microtwins in hexagonal MnAs ferromagnetic epilayers grown on GaAs (100) substrates with monolayer InAs buffer layers by using molecular beam epitaxy. The magnetization curve as a function of the magnetic field at 5 K showed that the MnAs thin films were ferromagnetic, and the magnetization curve as a function of the temperature revealed that the ferromagnetic transition temperature was as high as 325 K. The SADP and HRTEM results showed that an epitaxial relationship was formed among the hexagonal MnAs layer, the InAs layer, and the zincblende GaAs substrate. Microtwins existed between the MnAs grain boundaries on top of the InAs layers. Based on the SADP and HRTEM results, we present a possible atomic arrangement of the microtwins for the MnAs-InAs-GaAs heterostructure. © *2005 American Institute of Physics*.
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Hybrid heterostructures, which combine ferromagnets with semiconductors, have become particularly attractive because they allow investigations of the fundamental physical properties of such structures[1–3] and because they have potential applications in spintronic devices.[4–6] Among the many ferromagnet/semiconductor heterostructures, epitaxial MnAs films grown on GaAs substrates are the promising candidates for next-generation spin-injection devices.[7–13] Epitaxial MnAs thin films grown on GaAs substrates, which integrate magnetic and semiconductor properties, have been attractive because of the current interest in both investigations of fundamental physical properties[15] and promising applications in spintronics, such as spin injection[16] and magnetologic devices.[17] Furthermore, since the Curie temperature (T_c) of the MnAs epitaxial thin film is above room temperature,[18] spintronic devices fabricated utilizing MnAs-GaAs heterostructures can be operated at room temperature. Even though some studies have addressed the formation and the magnetic properties of hexagonal MnAs thin films directly grown on GaAs substrates,[7–14] the formation and the microstructural properties of hexagonal MnAs thin films grown on monolayer InAs buffer layers have not yet been reported because of the delicate problems encountered in the growth process.

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This letter reports an observation of existence of the microwins in hexagonal MnAs thin films grown on GaAs (100) substrates with GaAs and InAs buffer layers by using molecular beam epitaxy (MBE). Superconducting quantum interference device (SQUID) measurements were performed to characterize the magnetic properties of the MnAs thin films. Selected-area electron-diffraction pattern (SADP) and high-resolution transmission electron microscopy (HRTEM) measurements were carried out to investigate the atomic and the twinned structures in the MnAs thin films grown on GaAs substrates with monolayer (ML) InAs layers. A possible twinning structure for the hexagonal MnAs/zincblende GaAs heterostructure is presented on the basis of the HRTEM and the SADP results.

The MnAs thin films used in this work were grown on semi-insulating GaAs (100)-oriented substrates by using a MBE system. As soon as the chemical cleaning process was finished, the GaAs substrates were mounted onto a molybdenum susceptor. At a substrate temperature of 580 °C, 70 nm GaAs buffer layers were grown on GaAs substrates, and at a substrate temperature of 200 °C, 100 nm MnAs thin films were grown on 6.0 ML thick InAs buffer layers that had been grown at a substrate temperature of 400 °C on GaAs buffer layers. The insertion of the ML InAs buffer layer between the MnAs and the GaAs layers might lead to a release of the stress in the MnAs epilayer. Reflection high-energy electron

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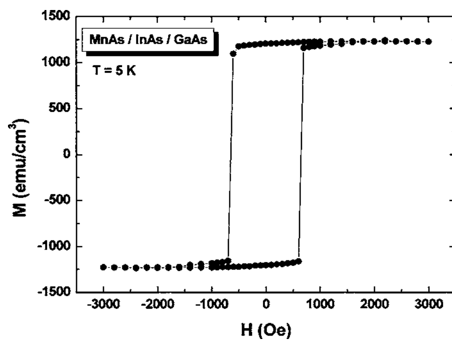
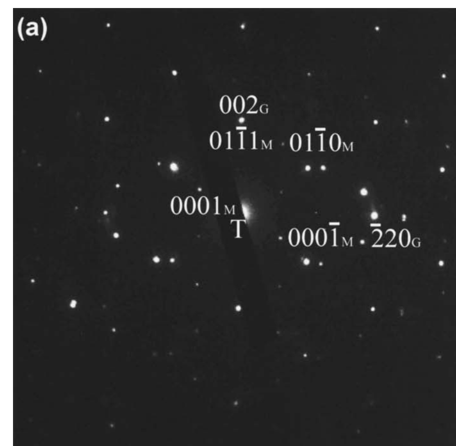


FIG. 1. Magnetization curve as a function of the magnetic field at 5 K for a hexagonal MnAs thin film grown on a GaAs (100) substrate with InAs and GaAs buffer layers. The direction of the magnetic field is parallel to the sample surface.

diffraction patterns were used to monitor the surface reconstruction during growth.

The x-ray diffraction curve indicated that the MnAs thin films grown on GaAs (100) substrates with InAs and GaAs buffer layers had hexagonal single-crystal structures. Figure 1 shows a magnetization curve as a function of the magnetic field at 5 K for the hexagonal α MnAs thin films grown on InAs buffer layers. This curve indicates that the MnAs thin films are ferromagnetic. When an external magnetic field was applied in the in-plane direction of the hexagonal α MnAs thin film, a sharp hysteresis loop appeared in the curve, as shown in Fig. 1. A ferromagnetic single phase had been formed, and the ordered direction of the spin was parallel to the sample surface. The remanent magnetization along the in-plane direction of the hexagonal α MnAs thin film, determined from Fig. 1, was 1206.9 emu/cm^3. Since the magnetization properties of hexagonal α MnAs thin films grown on GaAs (100) substrates with InAs and GaAs buffer layers are anisotropic, the films' easy axis is the GaAs [110] direction.[17] The magnetization curve as a function of the temperature for MnAs thin films grown on GaAs (100) substrates indicates that the ferromagnetic properties of the MnAs thin films are maintained until 325 K. Therefore, the T_c value of MnAs thin films is 325 K.

Figure 2(a) shows a SADP of the MnAs-InAs-GaAs heterostructure, and Fig. 2(b) depicts a schematic diagram of the diffraction spots corresponding to the SADP of the heterostructure. The SADP indicates that the crystal structure of the MnAs thin film grown on the InAs buffer is hexagonal. Since the thickness of the InAs layer is very thin, the electron diffraction spots related to the InAs layer do not appear in the SADP. The electron diffraction spots denoted by $hkil_M$ and hkl_G correspond to the MnAs and the GaAs layer indices, respectively. The incident beam directions of the MnAs epilayer and the GaAs (100) buffer layer are $[2\bar{1}\bar{1}0]$ and [110], respectively. The empty and the filled circles represent the diffraction spots corresponding to MnAs grains and are indicative of a twin relationship. When the MnAs epilayer is grown on a InAs buffer layer with a thickness of more than 3 ML, a twinned MnAs epilayer is formed, and the growth direction of the MnAs epilayer is $[01\bar{1}1]$. A hexagonal α MnAs epilayer can be generally grown along the $[1\bar{1}00]$ direction on a (001) GaAs substrate and is referred to as a type-A epilayer. Any variation of the template changes the epitaxial growth directions of the MnAs epilayer to $[\bar{1}101]$



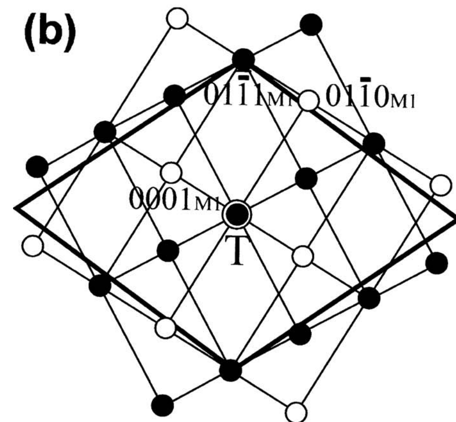


FIG. 2. (a) Selected area electron diffraction pattern of the MnAs-InAs-GaAs heterostructure and (b) schematic diagram of diffraction spots corresponding to the selected area electron diffraction pattern of the MnAs-InAs-GaAs heterostructure.

and $[\bar{1}102]$; this is referred to as a type-B epilayer.[19] Therefore, the MnAs thin film grown on the InAs buffer layer in this study is a type B MnAs. The angle between the {0001} diffraction spots is approximately 60°. Even though the MnAs thin film directly grown on the GaAs (100) substrate can be a type-A or a type-B epilayer, since the MnAs thin film grown on the InAs buffer layer is a type B MnAs, the use of the InAs buffer layer for the hexagonal MnAs ferromagnetic epilayer is necessary for obtaining the type B MnAs.

A HRTEM image of the MnAs-InAs-GaAs heterostructure is presented in Fig. 3. Figure 3 directly shows the crystal structures on both sides of, and near, the MnAs-InAs-GaAs heterointerface, and the HRTEM image shows that a MnAs-InAs-GaAs heterostructure is formed. The thicknesses of the InAs grains are between 15 and 30 nm, and the epitaxial MnAs thin film is grown on the InAs grains. The angle be-

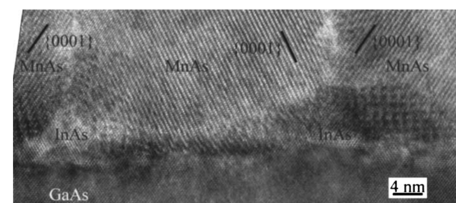


FIG. 3. High-resolution transmission electron microscopy image of the MnAs-InAs-GaAs heterostructure.

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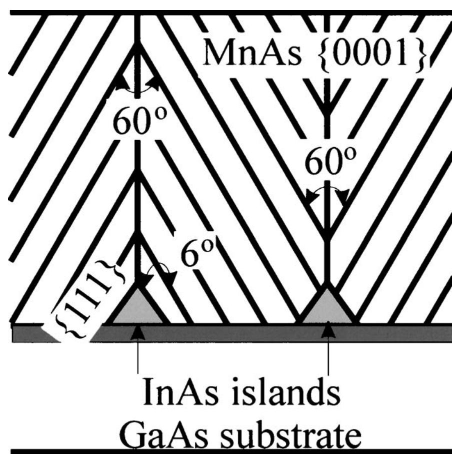

FIG. 4. Schematic diagram of the microtwin structure of an hexagonal MnAs epilayer grown on a GaAs (100) substrate.

tween the InAs {111} and the GaAs (001) directions is approximately 54°, and the angle between the InAs {111} and MnAs (0001) directions is about 6°. Two MnAs grains close together on top of the InAs islands cause twinning due to the formation of the grain boundary. The interface contrast between the MnAs grains is slightly smeared due to the lattice mismatch. The InAs layer does not exist continuously between the islands. The composition plane is related to the $(01\bar{1}\bar{3})$ plane, which is in reasonable agreement with the result of the SADP measurements shown in Fig. 2. The angle between two {0001} MnAs grains is approximately 60°, which is also consistent with the SADP results. Therefore, the twins are generated due to a relaxation of the lattice mismatch when the MnAs epitaxial layer is grown on an InAs layer.

When hexagonal MnAs layers are grown on (100) GaAs substrates or (100) InAs buffer layers, the lattice mismatch becomes different with growth directions. When the hexagonal MnAs is grown along the $(01\bar{1}1)$ plane, the lattice mismatches between the $\{01\bar{1}\bar{3}\}$ MnAs and the {110} GaAs planes and between {110} InAs and {110} GaAs planes, determined from the bulk values,[9] are approximately 22% and 31%, respectively. The lattice mismatch between the MnAs and the InAs is lager than that between the MnAs and the GaAs. However, while the InAs islands with the {111} planes are formed, the lattice mismatch between the {0001} MnAs and the {111} InAs planes is about 15%. Therefore, the MnAs twinning planes are formed on top of InAs islands, the MnAs {0001} plane, which is almost parallel to the InAs {111} direction, might reduce the strain energy due to the lattice mismatch.

Figure 4 shows a schematic diagram of the microtwinned structure of the MnAs epitaxial layer. The epitaxial direction of a MnAs thin film grown on a GaAs (100) substrate without an InAs buffer layer is generally $[\bar{1}100]$. However, when the InAs buffer layer is above a critical thickness, InAs islands, instead of an InAs layer, are formed on the GaAs substrate.[20] These InAs islands generate microtwins in the MnAs epitaxial layer, reducing the strain energy due to the lattice mismatch. The 60° angular relationship between the {0001} MnAs planes is necessary to twinning centering around the $(01\bar{1}\bar{3})$ composition plane between the {0001} MnAs planes. The InAs islands consisted of {111} planes. The angle between the {111} InAs and the GaAs {100} planes is approximately 54°. Therefore, the MnAs {0001} plane maintains the angular relationships with the InAs {111} and the GaAs {001} planes; the angles between the MnAs {0001} and the InAs {111} planes and between the MnAs {0001} and the GaAs {001} planes are about 6° and 60°, respectively. Since the angle between the {0001} planes is also 60°, each grain forms twinned grain boundaries without lattice distortion.

In summary, hexagonal epitaxial MnAs thin films were grown on InAs buffer layers by using MBE. SADP and HRTEM results showed that the MnAs epitaxial layer grown on the InAs buffer layer had a hexagonal crystalline structure. The magnetization curve as a function of the magnetic field at 5 K showed that the MnAs thin film grown on the GaAs substrate with InAs and GaAs buffer layers was ferromagnetic. The magnetization curve as a function of the temperature revealed the T_c value to be as high as 325 K. Microtwins were formed at the grain boundaries of the MnAs thin film on top of the InAs layer. A schematic diagram of the crystal structure for the twinned hexagonal MnAs epitaxial layer was proposed.

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