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Substrate-Orientation Dependence on Structure and Magnetic Properties of MnAs Epitaxial Layers

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Ferromagnetic MnAs layers were grown on (001), (111)A, and (111)B GaAs substrates by molecular-beam epitaxy at substrate temperatures (T_s) in the range from 150 to 400°C. The crystal structure of the MnAs was NiAs type and its orientation was found to change depending on the substrate, ($\bar{1}101$) for the (001) GaAs substrate and (0001) for the (111)A and (111)B substrates at T_s between 300 and 400°C. Polar magneto-optical Kerr-effect measurement demonstrated a crystal-orientation dependence of Kerr rotation and Kerr ellipticity spectra of the MnAs epitaxial layers.

KEYWORDS: MnAs, molecular beam epitaxy, reflection high-energy electron diffraction, x-ray diffraction, polar magneto-optical Kerr-effect

1. Introduction

Ferromagnet/semiconductor heterostructures are attracting much attention because of their potential application in fabricating new devices integrating magnetic, electronic and optical properties.[1] Recent progress in the molecular-beam epitaxy (MBE) technique has demonstrated that excellent-quality ferromagnetic compounds such as CuAu-type MnGa and MnAl can be grown on III–V substrates such as GaAs and InAs.[2,3]

MnSb and MnAs are ferromagnets of the NiAs-type structure, and are grown by MBE and hot-wall epitaxy.[4–9] Akinaga *et al.* reported growth of MnSb layers by MBE:[4,5] the growth direction is [$\bar{1}101$] for the (001) GaAs substrate and [0001] for (0001) sapphire substrate. They also demonstrated a considerable magnetocrystalline anisotropy. Tanaka *et al.* showed that the growth direction and the crystal structure of MnAs epitaxial layers on (001) GaAs substrates depend on both the growth condition for the first few layers and the growth procedure.[7–9] However, there have so far been no detailed reports concerning the growth direction and magnetic properties of MnAs epitaxial layers on GaAs substrates having different orientations. In this paper we discuss the results of a systematic study of the effects of the substrate orientation and the growth temperature on the structure and magnetic properties of MnAs epilayers grown by MBE.

2. Experimental

MnAs MBE growth was carried out in a conventional III–V MBE machine with Mn effusion cell. The substrates were n-type (001), (111)A, and (111)B GaAs. After growing a 200 nm thick undoped GaAs buffer layer on each substrate at 600°C, the substrate temperature (T_s) was lowered to a desired temperature. During the cooling process, the As_4 shutter was closed at T_s of about 480°C. A c(4×4) reconstruction was observed from the (001) surface and a (2×2) from (111)A and (111)B surfaces after closing the As shutter.

MnAs layers were grown under following conditions: an As_4 beam equivalent pressure (BEP) was fixed at about 5×10^{-4} Pa on a beam-flux monitor, and the As_4/Mn BEP ratio was kept constant at about 50. MnAs layers with thicknesses of about 100 and 200 nm were grown on (001) and (111) substrates with growth rates of about 50 and 100 nm/h, respectively, while T_s was varied from 150 to 400°C, as measured by an infrared pyrometer.

The growth direction and the crystal structure of MnAs epitaxial layers grown at various temperatures on each substrate were studied by *in situ* reflection high-energy electron diffraction (RHEED) and *ex situ* X-ray diffraction (XRD). The magnetic properties of MnAs epilayers were investigated using polar Kerr measurements. Polar magneto-optical Kerr rotation and Kerr ellipticity spectra were measured at room temperature for photon energies region from 1.2 to 5.3 eV.

3. Results and Discussion

Figure 1 shows XRD patterns for MnAs epitaxial layers grown at 250 and 350°C on (a) (001), (b) (111)A, and (c) (111)B GaAs substrates. In the case of the (001) substrate, besides a GaAs (002) peak at 31.6° and a huge GaAs (004) peak at 66.0°, small peaks were observed at 27.7, 57.2, and 91.7° in the epilayers grown at T_s below 250°C, corresponding to ($\bar{1}100$), ($\bar{2}200$), ($\bar{3}300$) reflection of hexagonal MnAs, respectively. On the other hand, a strong ($\bar{1}101$) MnAs peak and a ($\bar{2}202$) MnAs peak were observed at 31.9 and 66.6°, respectively, in the epilayers grown at T_s between 300 and 400°C. These results indicate that the growth plane of MnAs epitaxial layers grown at T_s between 300 and 400°C on the c(4×4) GaAs (001) substrate is the ($\bar{1}101$) plane.

For the case of the (111)A GaAs substrate, besides huge GaAs (111) and (333) peaks at 27.3 and 90.2°, respectively, strong ($\bar{1}101$) and ($\bar{2}202$) MnAs peaks were observed at 31.9 and 66.6°, respectively, in the epilayers grown at T_s below 300°C. On the other hand, strong (0002) and (0004) MnAs peaks were observed at 31.3 and 65.3° in the epilayers grown at T_s between 300 and 400°C. Thus, the growth plane of MnAs epitaxial layers on (111)A substrates was found to change depending on the substrate temperature, i.e., ($\bar{1}101$) at T_s below 300°C and (0001) at T_s above 300°C.

At all substrate temperatures, unlike in other cases involving different substrates, (0002) and (0004) MnAs peaks were observed at 31.3 and 65.3°, respectively, in

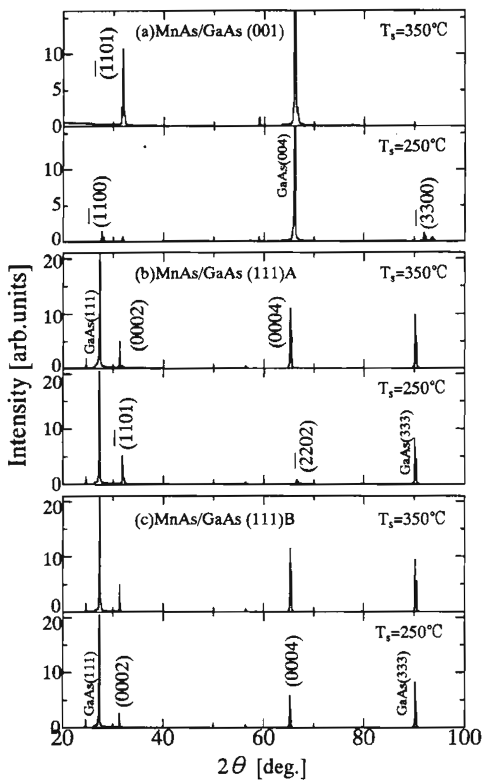

Fig. 1 XRD patterns for MnAs epilayers grown at 250 and 350°C on (a) (001), (b) (111)A, and (c) (111)B GaAs substrates.

the epilayers grown on the (111)B GaAs substrate, indicating that the growth plane of MnAs epitaxial layers on (111)B substrates is the (0001) plane.

Figure 2 shows the substrate temperature dependence of the intensities of the XRD peaks for the MnAs epilayers grown on (a) (001), (b) (111)A, and (c) (111)B GaAs substrates. The intensities of $(\bar{1}101)$ and $(\bar{2}202)$ MnAs peaks, observed for the epilayers grown at T_s between 300 and 400°C on (001) substrates, increased with substrate temperature up to 350°C, and decreased thereafter. For the case of the (111)A GaAs substrate, the intensities of $(\bar{1}101)$ and $(\bar{2}202)$ MnAs peaks increased with substrate temperature up to 250°C, and then decreased above 250°C. On the other hand, (0002) and (0004) MnAs peaks, observed for the epilayers grown at T_s above 300°C, increased in their intensities with substrate temperature up to 350°C, and then decreased thereafter. In the case of the (111)B substrate, the intensities of (0002) and (0004) MnAs peaks increased with substrate temperature up to 350°C.

In the present experiment, the full width at half-maximum (FWHM) of the MnAs peaks obtained at T_s of about 350°C was less than 0.1°, independent of the substrate orientation. The value was about half the width of the epilayers grown at other substrate temperatures, showing that high quality epilayers were grown at about

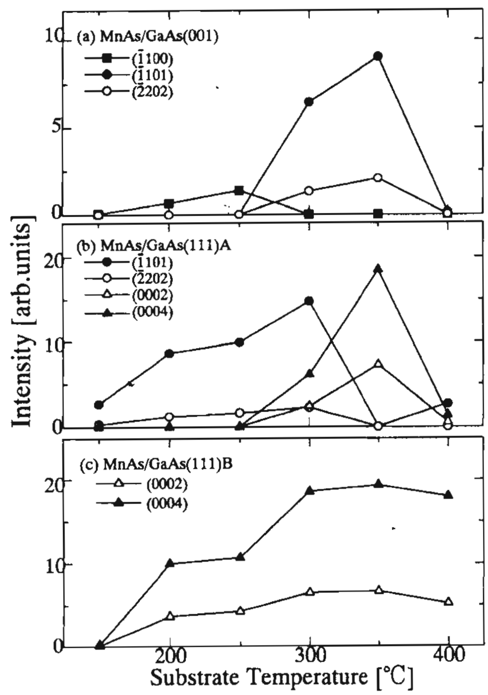

Fig. 2. Substrate temperature dependence of the XRD peak intensities for MnAs epilayers grown on (a) (001), (b) (111)A, and (c) (111)B GaAs substrates.

350°C.

In order to investigate the in-plane relationship between MnAs epilayers and GaAs substrates, RHEED patterns were observed along various azimuths during growth. By measuring the interval between the integral-order streaks, the lattice constant in each plane of MnAs epilayers were estimated: the estimated atomic distances along the various directions were in good agreement with those of bulk hexagonal MnAs, confirming the in-plane relationship between MnAs epilayers and GaAs substrates summarized in Table 1. The structure in the layer

Table I. The in-plane relationship between MnAs epilayers and GaAs substrates.

GaAs	MnAs	in-plane structure
(001)	$(\bar{1}100)$	MnAs$[\bar{1}\bar{1}20]$//GaAs[110]
		MnAs[0001]//GaAs$[\bar{1}10]$
	$(\bar{1}101)$	MnAs$[1\bar{1}02]$//GaAs[110]
		MnAs$[1\bar{1}\bar{2}0]$//GaAs$[\bar{1}10]$
(111)A	$(\bar{1}101)$	MnAs$[1\bar{1}02]$//GaAs$[11\bar{2}]$
		MnAs$[\bar{1}\bar{1}20]$//GaAs$[\bar{1}10]$
	(0001)	MnAs$[1\bar{1}00]$//GaAs$[11\bar{2}]$
		MnAs$[\bar{1}\bar{1}20]$//GaAs$[\bar{1}10]$
(111)B	(0001)	MnAs$[1\bar{1}00]$//GaAs$[11\bar{2}]$
		MnAs$[\bar{1}\bar{1}20]$//GaAs$[\bar{1}10]$

plane of ($\bar{1}$100) MnAs on (001) GaAs agreed with that reported for the MBE growth of MnAs on (001) GaAs at T_s of about 250°C.[7] Moreover, the in-plane structure of ($\bar{1}$101) MnAs epitaxial layers on the c(4 × 4) GaAs (001) substrate was that obtained for the growth at T_s of about 250°C with 1 ML of Mn as a template,[7] and it agreed with that demonstrated for the MBE growth starting with a codeposition of Mn and As fluxes on c(4×4) GaAs (001) substrates.[8] These results indicate that the growth direction depends crucially on the amount of excess As atoms on an As-stabilized (001) surface, and support the conclusion obtained by Tanaka *et al.*

The growth of MnAs with 1 ML of Mn as a template led to a change in the growth direction from [$\bar{1}$101] to [0001] at T_s of 250°C on a (111)A substrate, while the [0001] growth direction remained unchanged for the growth with 1 ML of Mn as a template on a (111)B substrate. Moreover, (0001) MnAs layers were also obtained at T_s of 350°C on both (111)A and (111)B substrates, independent of the results with/without the supply of 1 ML of Mn as a template. Similarly, as described regarding the (001) GaAs case, the interaction of Mn atoms with the starting surface potential of GaAs is responsible for a considerable difference in the growth direction between the MnAs epilayers.

Polar magneto-optical Kerr rotation and Kerr ellipticity spectra were measured at room temperature under a magnetic field of 17 KOe, where the Kerr rotation angle of all the samples were saturated. Figure 3 shows both the Kerr rotation and Kerr ellipticity spectra as a function of energy from 1.2 to 5.3 eV. The solid curve was recorded for a MnAs epilayer facing to the [$\bar{1}$100] direction grown on a (001) GaAs at 250°C, the dash dotted ones for epilayers facing to the [$\bar{1}$101] direction grown on a (001) GaAs at 350°C and facing to the [0001] direction grown on a (111)B GaAs at 350°C, and the dotted one for an epilayer facing to both the [$\bar{1}$101] and [0001] directions grown on a (111)A GaAs at 300°C. As shown in Fig. 3(a), the Kerr rotation for the (0001) MnAs epilayer exhibited two local maxima at about 1.5 and 2.4 eV, and crossed zero around 3.7 eV, where the Kerr ellipticity took a maximum value. In the case of the ($\bar{1}$100) MnAs epilayer, the Kerr rotation reached maxima at about 1.6 and 2.5 eV, and crossed zero around 4.5 eV. For the ($\bar{1}$101) epilayer, the Kerr rotation reached maxima at about 1.6 and 2.4 eV, and crossed zero around 4.3 eV, which shows that the spectrum is characterized as a transient curve between the spectra for the (0001) and ($\bar{1}$100) MnAs epilayers. Almost the same tendency was observed regarding the Kerr rotation and Kerr ellipticity for the epilayers facing to the respective directions grown under various conditions, indicating that magneto-optical properties of the present epilayers are deeply influenced by the crystal orientation. In order to elucidate the origin of the dependence, investigations about optical reflectivity and analysis in terms of the electric conductivity tensor are desired.

In the present experiment, the absolute value of the Kerr rotation angle was smaller than that of the epilayer grown by hot-wall epitaxy.[10] The smaller Kerr rotation angle may arise from the shift of the Mn composition of

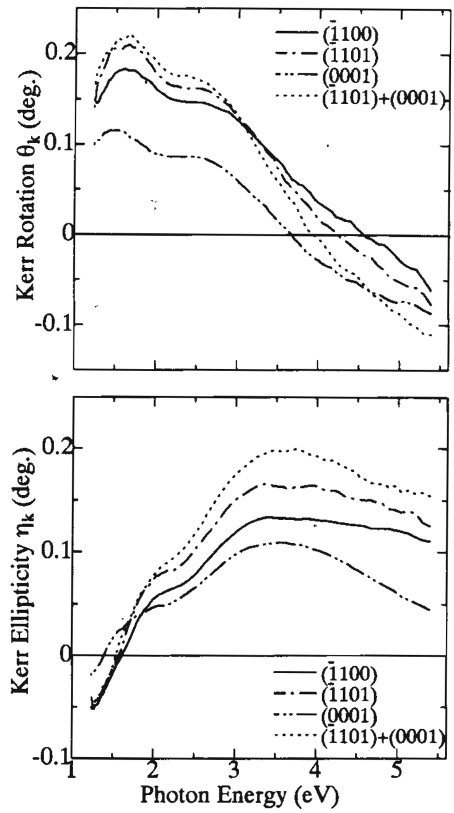

Fig. 3. Polar magnet-optical Kerr rotation and Kerr ellipticity spectra for various MnAs epilayers.

the epilayers grown by MBE, since in the case of MnSb the excess or lack of the composition of Mn atoms was reported to cause a decrease of the Kerr rotation angle.[11] A further investigation is necessary in order to obtain higher quality epilayers by MBE.

4. Summary

MnAs layers were grown on (001), (111)A, and (111)B GaAs substrates by MBE at T_s between 150 and 400°C. XRD measurements and RHEED observations showed that the crystal structure of the MnAs was NiAs type and its orientation was found to change depending on both the substrate orientation and substrate temperature, ($\bar{1}$101) for the (001) GaAs substrate and (0001) for the (111)A and (111)B substrates at T_s from 300 to 400°C. Polar magneto-optical Kerr-effect measurement demonstrated a crystal-orientation dependence of Kerr rotation and Kerr ellipticity spectra of the MnAs epitaxial layers.

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