Growth and properties of (Ga,Mn)As films with high Mn concentration

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(Ga, Mn)As films with high nominal Mn concentration ($0<x<0.55$) are grown by low temperature molecular beam epitaxy (LT-MBE), growth temperature $T_s=180\,°C$. Reflection high energy electron diffraction patterns indicate epitaxial growth of (Ga, Mn)As for $x<0.1$, whereas they show spotty patterns for $x>0.1$, which turn to polycrystalline features when $x>0.3$. X-ray diffraction shows the formation of MnAs together with the growth of (Ga, Mn)As. The lattice constant of the layers suggests that (Ga, Mn)As with high Mn composition as high as 17% can be grown by LT-MBE. [DOI: 10.1063/1.1357841]

I. INTRODUCTION

Ferromagnetic semiconductors (In, Mn)As and (Ga, Mn)As grown by low-temperature molecular beam epitaxy (LT-MBE) open up a new possibility of combining the spin and charge degrees of freedom in III–V based structures.[1–4] The model put forward for the hole-induced ferromagnetism in magnetic III–Vs shows that higher ferromagnetic transition temperature T_c requires higher x and/or higher hole concentration.[5] Although room temperature ferromagnetism is preferable for a number of applications, the highest T_c so far obtained is 110 K for (Ga, Mn)As with Mn composition of 0.053.[6]

In this study, (Ga, Mn)As films with nominal Mn composition x in the range of $0<x<0.55$ are grown at $T_s=180\,°C$ and are investigated by reflection high energy electron diffraction (RHEED), x-ray diffraction (XRD), magnetization, and magnetotransport measurements.

II. EXPERIMENTAL RESULTS AND DISCUSSIONS

The (Ga, Mn)As films with nominal x in the range of $0<x<0.55$ are grown by LT-MBE. Here, x is determined by the beam equivalent pressures of Mn to Ga, using the calibration curve of nominal x versus x determined by XRD for $x<0.07$, where single crystal (Ga, Mn)As layers can be grown. Typical growth rates are 6 nm/min and the As/Ga beam equivalent pressure ratio is typically 100.

On GaAs (100) substrate, a GaAs buffer layer (20–50 nm) is first grown at 560 °C, followed by a 50 nm $(Al_{0.7}Ga_{0.3})As$ buffer layer. Then the substrate is cooled to 180 °C and (Ga,Mn)As (100–180 nm) is grown. For reference, polycrystalline MnAs grown at $T_s=20\,°C$ as well as (Ga, Mn)As grown at $T_s=300\,°C$ with nominal $x=0.31$, in which MnAs exists, are also prepared.

During growth, RHEED pattern is used to monitor the surface condition. At $T_s=560\,°C$, well known (2×4) surface reconstruction is observed. At low substrate temperature of 180 °C, GaAs shows a spot like (1×1) pattern and (Ga,Mn)As shows a streaky (1×2) pattern for $x<0.1$, which is very similar to what has been observed in epitaxial growth of (Ga,Mn)As at 250 °C.[7] For $x>0.1$, the RHEED pattern is spotty as shown in Fig. 1(a) and when x exceeds 0.3, the pattern develops into a ring patterns shown in Fig. 1(b).

Crystallographic analysis is carried out using grazing incidence XRD. The typical patterns with an incident angle of 3° are shown in Fig. 2 where trace 1 is from the $x=0.36$ layer grown at 180 °C, 2 from $x=0.28$, 3 from $x=0.31$ grown at 300 °C, and 4 from the polycrystalline MnAs layer. Some of the peaks that appeared in $x=0.28$ and 0.36 samples can be easily identified as coming from MnAs by comparison of the traces. Relative intensities of MnAs to others increase from $x=0.28$ to 0.36. The relative intensity of the series of MnAs for $x=0.28$ is much weaker than that

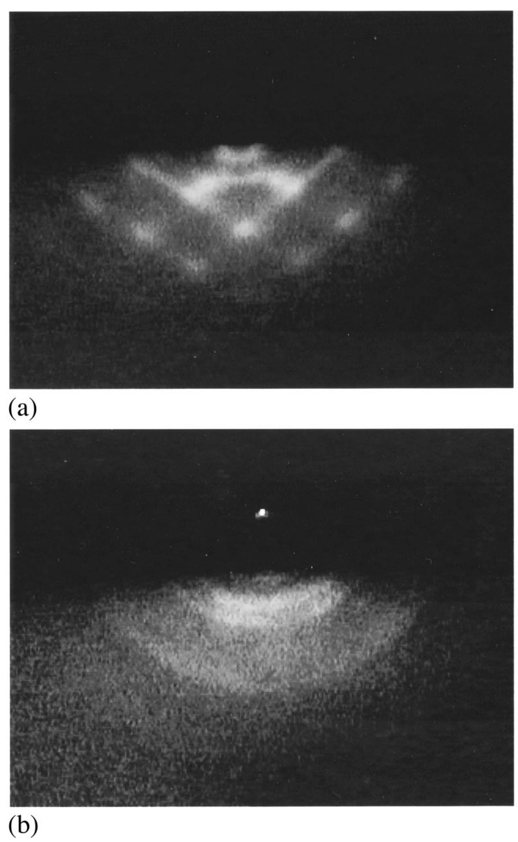

FIG. 1. RHEED pattern after growth (a) for $x=0.23$ and for (b) for $x=0.44$.

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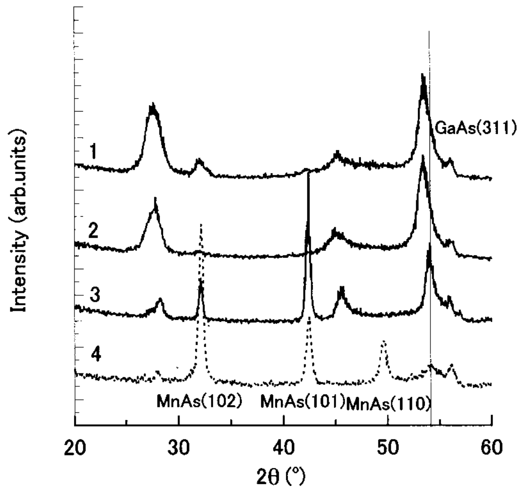

FIG. 2. Grazing incidence x-ray diffraction pattern for $x=0.36$ (trace 1), for $x=0.28$ (trace 2), for the grown layer at $T_s=300\,°C$ (trace 3) and polycrystalline MnAs (trace 4). The position of the line shows the diffraction angle of GaAs (311).

of the films grown at $T_s=300\,°C$ in spite of almost the same nominal Mn composition.

We now turn our attention to the GaAs related peaks. The (311) diffraction peaks from the GaAs substrate (trace 4) and from GaAs grown at $T_s=300\,°C$ (trace 3) appear at almost identical diffraction angle. On the other hand, the same (311) diffraction peaks from the $x=0.28$ and 0.36 samples show clear peak shift toward lower angle. This shows that a considerable amount of Mn is incorporated into the GaAs matrix and (Ga,Mn)As is formed. Assuming no strain (because of the polycrystalline nature of the $x=0.36$ sample) and using the published curve between x and free standing (Ga,Mn)As lattice constant, we estimate the Mn composition in this (Ga,Mn)As to be 17%, which is the highest ever obtained in the alloy form. With increasing $x(0.2<x<0.36)$, the Mn composition in the (Ga,Mn)As matrix is almost constant within the experimental error of 3% Mn composition.

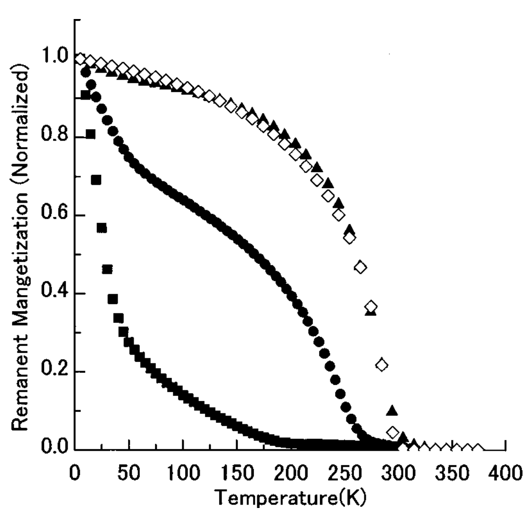

FIG. 3. The temperature dependence of the remanent magnetization. The closed squares are for $x=0.28$, the closed circles for $x=0.36$, and the closed triangles for $x=0.55$. As a reference, polycrystalline MnAs is also shown by the open diamonds.

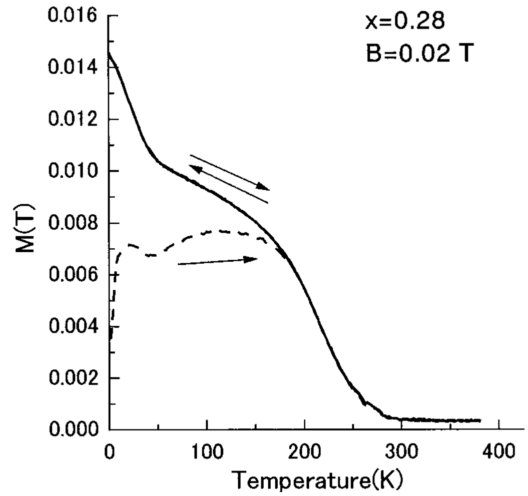

FIG. 4. Temperature dependence of magnetization in the $x=0.28$ sample. Solid line shows the field cooled (0.02 T) case; the broken line is for the zero field cooled case.

The magnetic properties are measured by using a superconducting quantum interference device magnetometer. Figure 3 shows the temperature dependence of remanent magnetization M_r for samples with $x=0.28$, 0.36, 0.55 and polycrystalline MnAs. The samples are magnetized in 2 T at 5 K with magnetic field (B) parallel to the sample plane, and magnetization is measured in zero field as the temperature is increased. As seen in Fig. 3, M_r goes to zero at 180 K and 260 K for $x=0.28$ and 0.36, respectively, in contrast to 310 K of MnAs bulk, whereas the curve for $x=0.55$ almost coincides with that of MnAs. Figure 4 shows the temperature dependence of magnetization under zero field cooled and field cooled conditions for the $x=0.28$ sample, where two curves merge at $T=180$ K. Separate measurements of magnetization curves at 2–340 K of the same sample show that the applied 0.02 T is beyond the coercive field of the film above 100 K. Thus, it is likely that the temperature at which M_r goes to zero for the $x=0.28$ sample in Fig. 3 is the blocking temperature of ferromagnetic clusters in the film, which is most probably MnAs. The shift of the blocking temperature toward high temperatures as Mn concentration increases in Fig. 3 is believed to be due to the increase in the size of the ferromagnetic clusters and/or the reduced distance among them.

Magnetotransport measurements, extensively used to determine the magnetic properties of (Ga, Mn)As films,[6] show nonlinear behavior in the Hall resistance versus field curves below 180 K. Although the presence of MnAs clusters did not show up in magnetotransport of (Ga, Mn)As films grown at 250 °C, we also found similar nonlinearity in the polycrystalline MnAs films, which precludes extraction of magnetic properties from transport measurements. The peculiar temperature dependence below 50 K may be related to the (Ga,Mn)As phase in the film; further study is necessary to clarify the magnetic properties of the phase.

III. SUMMARY

Low temperature ($T_s=180\,°C$) MBE growth of (Ga,Mn)As with high nominal Mn concentration is performed. The grazing incidence XRD measurements show the presence of the (Ga, Mn)As phase with Mn composition of 17%, together with the formation of MnAs. Although it was not possible to determine the magnetic properties of this (Ga, Mn)As phase, the present results show that growth of high Mn concentration epitaxial (Ga, Mn)As, which is a prerequisite for room temperature ferromagnetism, may be possible by advancing the material science of this material.

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