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Epitaxial Growth and Characterization of Zinc-blende CrAs/GaAs/MnAs/GaAs Multilayers

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We report on the growth, structural and transport properties of zinc-blende CrAs/GaAs/MnAs/GaAs multilayers on GaAs(001) substrates. Structural analyses using *in-situ* reflection high-energy electron diffraction and *ex-situ* cross-sectional transmission electron microscopy confirmed the realization of a zinc-blende crystal structure. Room temperature Hall measurements reveal that the as-grown multilayer exhibits *p*-type conductivity with a very low resistivity of 0.052 Ωcm, a high carrier concentration of 6.2×10^{19} cm^{-3} and a Hall mobility of 1.8 cm^2/Vs. We also observed a clear decrease of the resistivity in samples after low temperature annealing.

Key words : half-metallic ferromagnets, zinc-blende multilayer, hall effect

1. Introduction

Half-metallic ferromagnets are expected as a key ingredient in future high performance spintronic devices, because they have only one electronic spin channel at the Fermi energy and, therefore, show nearly 100% spin polarization [1, 2]. Since de Groot *et al.*'s discovery [3] in 1983, many kinds of half-metallic ferromagnets have been theoretically predicted and some of them, such as Heusler alloys [4, 5], CrO_2 [6], and transition-metal perovskites [7] have been confirmed experimentally. On the other hand, it is highly desirable to grow half-metallic ferromagnetic thin films on semiconductors because of the possibility of an application to "spintronic" devices. For this purpose, considerable efforts have been made on the epitaxial growth of the transition-metal pnictides, such as MnAs [7], MnSb [8, 9], and MnBi [10] on GaAs substrates.

Using the full-potential linearized augmented-plane-wave (FLAPW) method, Akinaga *et al.* [11] first calculated that in the ferromagnetic case CrAs with a zinc-blende structure could be a half-metallic ferromagnet. Following the prediction, they have succeeded to grow thin films of CrAs on GaAs(001) substrates with the atomically flat interfaces by molecular-beam epitaxy [11]. They found that CrAs is ferromagnetic at room temperature with the Curie temperature higher than 400 K and they have deduced a total spin-moment of 3 μ_B in agreement with the theory. Nevertheless, the very thin critical thickness (3 nm) of zinc-blende CrAs film is one of the biggest challenges faced in advancing this material toward applications. More recently, *ab initio* calculations indicated the half-metallic properties are preserved in zinc-blende CrAs/GaAs hybrid multilayers [12]. Experimentally, zinc-blende CrAs/GaAs multilayers up to 20 nm have been successful grown by molecular-beam epitaxy [13], but with a very high resistivity and low carrier concentration. Theoretical calculation predicted that Mn dopant substitutes for the Ga site and acts as an acceptor providing itinerant holes [14]. For this reason, we assume that the existence of MnAs layer may result in the increase of the carrier concentration in CrAs/GaAs system. Therefore, in this paper, we report on the growth, structural and transport properties of zinc-blende CrAs/GaAs/MnAs/GaAs hybrid multilayer on GaAs(001) substrates.

2. Experimental Details

A molecular-beam-epitaxy (MBE) system (Riber 32P) was used to grow zinc-blende CrAs/GaAs/MnAs/GaAs multilayer. Figure 1 shows the schematic structure of the multilayer study in this work. The multilayer was grown on an "epi-ready" semi-insulating GaAs(001) substrate.

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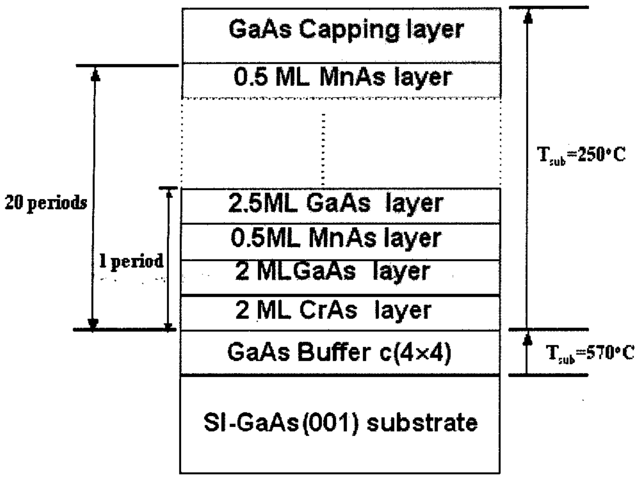

Fig. 1. A cross-sectional schematic drawing of the CrAs/GaAs/MnAs/GaAs multilayer.

The substrate was set on a molybdenum holder with indium solder. After being degassed at 400°C in a loading chamber, the substrate was transferred into the MBE growth chamber without breaking vacuum. Before starting the growth, the substrate was heated up to about 590°C and kept for 10 minutes for thermal cleaning to remove the surface oxidization layer. Then, a GaAs buffer layer with the thickness of 20 nm was grown at 570°C to prepare a flat surface. On top of the buffer layer, 2 monolayers (ML) of CrAs, 2 ML of GaAs, 0.5 ML of MnAs and 2.5 ML of GaAs were grown alternatively at 250°C. Growth of the CrAs, GaAs, and MnAs layers was performed by the opening of Knudsen cells for each element of Cr, Ga and Mn under exposure of As with a beam pressure of about 4×10^{-6} Pa. Finally, a GaAs capping layer with a thickness of 10 nm was deposited to prevent oxidation of the multilayer. The surface was characterized by *in-situ* reflection high-energy electron diffraction (RHEED) during the growth. The structural properties were examined by *ex-situ* cross-sectional transmission electron microscopy (TEM). Transport measurements were carried out at room temperature in Hall-bars fabricated by photolithography and wet etching.

3. Results and Discussion

Figure 2 shows the RHEED patterns of the GaAs surface and the last MnAs layer of the multilayer sample with electron azimuth along the [110] and [1$\bar{1}$0] axes of GaAs, respectively. For the GaAs substrate, the RHEED patterns [Figs. 2(a) and 2(b)] were taken at 570°C after the growth of 20 nm GaAs buffer layer. We note that along the [1$\bar{1}$0] direction the reconstruction shows four lines in comparison to two lines along [110], indicating a smooth (2×4)-reconstructed GaAs(001) surface. Upon cooling, the (2×4)-reconstructed surface changes to "As rich" c (4×4)-reconstructed surface at around 520°C and As shutter was closed below 400°C. The GaAs substrate retains the c (4×4)-reconstructed surface before the deposition of the initial CrAs layer at 250°C. Soon after the deposition of 1st CrAs layer at 250°C, we found that the reconstruction lines in the substrate's RHEED patterns disappear immediately, and the c (4×4) RHEED pattern changes to the (1×1) one. Importantly, the principal streaks lines remained during several repetitions of the alternate growth, which suggests the success of an epitaxial growth of the CrAs/GaAs/MnAs/GaAs multilayer. Moreover, we found that the streaky pattern gradually changed to slightly spotty pattern with the increase of the periods, and an obvious spotty pattern was observed when the last growth period was performed. Figure 2(c) and (d) show the RHEED patterns of the last MnAs layer surface of the multilayer.

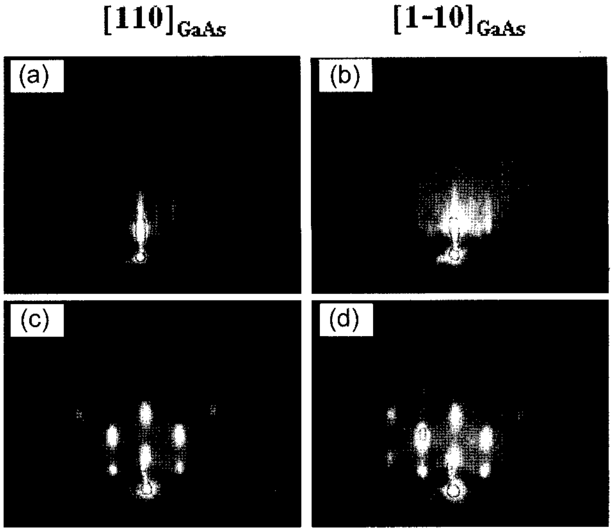

Fig. 2. RHEED patterns from the surface of (a)-(b) 20 nm GaAs buffer layer and (c)-(d) the last MnAs layer for the [110] azimuth (left-hand side) and the [1-10] azimuth (right-hand side).

The structural properties of the multilayer were further checked by high-resolution TEM. Figure 3(a) shows a representative wide-view cross-sectional TEM image of the zinc-blende multilayer. We can see that the multilayer was grown nearly as a planned structure because the thickness of ~20 nm was very close to the designed value of 23 nm. Figure 3(b) shows an enlarged interface zone of the multilayer. It can be clearly observed that the lattice image of multilayer forms continuous lines with the GaAs substrate, which indicated that CrAs, GaAs and MnAs composing the multilayer maintain zinc-blende structure.

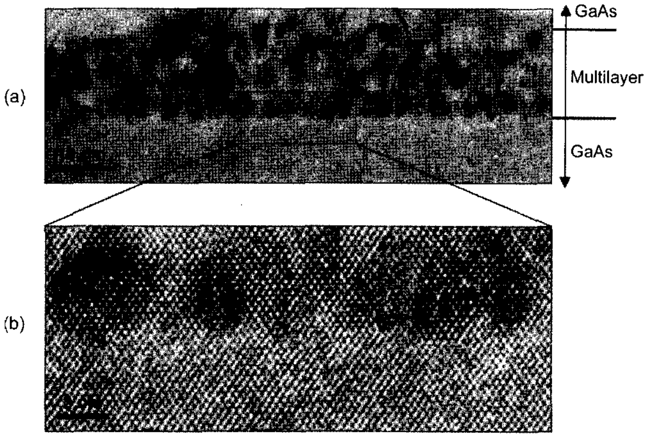

Fig. 3. (a) Cross-sectional TEM image of the multilayer with the structure of [CrAs(2ML)/GaAs(2ML)/MnAs(0.5ML)/GaAs(2.5ML)]×20 layers, grown on a 20 nm GaAs buffer layer and capped by a 10 nm GaAs layer. (b) An enlarged interface zone between GaAs substrate and the multilayer.

On the other hand, however, it should be pointed out that a lot of tiny clusters (zinc-blend type) with around 2~3 nm in diameter are observed in the whole multilayer region, as shown in our TEM images. Formation of defects and clusters have been observed in highly mismatched structures like InAs/GaAs where the lattice mismatch is 7.2%, and have been explained by the relaxation of the lattice strain [15]. The lattice mismatch between the hypothetical zinc-blende MnAs and GaAs is predicted to be around 6.0%. Therefore, we assume that lattice mismatch between zinc-blend MnAs and GaAs resulted in the formation of defects/clusters in our hybrid multilayer. In fact, gradually spotty RHEED patterns observed during MBE growth of the multilayer in our present work supporting above assumption. It means even for 0.5 ML MnAs layer, lattice strain relaxation has induced tiny clusters. Therefore, our ongoing works are to maximize the concentration of Mn substitutionally incorporated in the cation site of GaAs to act as acceptors rather than as compensating interstitial defects or clusters.

Hall effect and electrical resistivity measurements were carried out at room temperature in a Hall-bar fabricated by photolithography and wet etching. The results are summarized in Table 1. The as-grown multilayer exhibits p-type conductivity with a very low resistitivity of 0.052 Ωcm, a high carrier concentration of 6.2×10^{19} cm^{-3} and a mobility of 1.8 cm^2/Vs at room temperature. Moreover, *ex situ* postgrowth low-temperature annealing (at 200°C for 1 hour) was also performed to reduce the interstitial defect density. The results of Hall measurements for the annealing sample are also summarized in Table 1. We can see that, as expect, the resistivity decreases and the carrier concentration increases in sample after low-temperature annealing, although the mobility remain almost unchanged.

Table 1. Transport properties at room temperature of [CrAs (2ML)/GaAs(2ML)/MnAs(0.5ML)/GaAs(2.5ML)]×20 multilayers.

Sample	Type	Mobility (cm^2/Vs)	Resistivity (Ωcm)	Carrier density (cm^{-3})
As-grown	p	1.81	0.052	6.2×10^{19}
Anneal (at 200°C)	p	1.82	0.042	6.5×10^{19}

4. Conclusion

We have demonstrated the successful epitaxial growth of CrAs (2ML)/GaAs(2ML)/MnAs(0.5ML)/GaAs(2.5ML) multilayer on GaAs(001) substrates by MBE. *In-situ* RHEED patterns during the growth and *ex-situ* cross-sectional TEM images confirmed the realization of a zinc-blende crystal structure. Room temperature Hall measurements reveal that the as-grown multilayer exhibits *p*-type conductivity with a very low resistivity of 0.052 Ωcm, a high carrier concentration of 6.2×10^{19} cm^{-3} and a Hall mobility of 1.8 cm^2/Vs. The resistivity decreases clearly in samples after low temperature annealing.

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References

[1] R. A. Groot, F. M. Mueller, P. G. van Engen, and K. H. J. Buschow, Phys. Rev. Lett. **50** 2024 (1983).
[2] G. A. Prinz, Phys. Today **48**, 58 (1995).
[3] T. Ambrose, J. J. Krebs, and G. A. Prinz, Apply. Phys. Lett. **76**, 3280 (2000).
[4] W. Van Roy, J. De Boeck, B. Brijs, and G. Borghs, Apply. Phys. Lett. **77**, 4190 (2000).

[5] W. H. Wang, M. Przybylski, W. Kuch, L. I. Chelaru, J. Wang, Y. F. Lu, J. Barthel, H. L. Meyerheim, and J. Kirschner, Phy. Rev. B **71**, 144416 (2005).

[6] N. E. Brener, J. M. Tyler, J. Callaway, D. Bagayoko, and G. L. Zhao, Phys. Rev. B **61**, 16582 (2000).

[7] J. Z. Sun, W. J allagher, P. R. Duncombe, L. Krusin-Elbaum R. A.Altma, A. Gupta, Yu. Lu, G. Q. Gong, and G. Xiao, Appl. Phys. Lett. **69**, 3266 (1996).

[8] S. Miyanishi, H. Akinaga, W. van Roy, K. Tanaka, App. Phys. Lett. **70**, 2046 (1997).

[9] M. Mizuguchi, H. Akinaga, K. Ono, and M. Oshima, Appl. Phys. Lett. **76**, 1743 (2000).

[10] R. Hayashi, K. Terayama, T. Ishibashi, and K. Sato, J. Magn. Soc. Japan **23**, 99 (1999).

[11] H. Akinaga, T. Manago, and M. Shirai, Japan. J. Appl. Phys. **39**, L1118 (2000).

[12] C. Y. Fong and M. C. Qian, J. Phys.: Condens. Matter **16**, S5669 (2004).

[13] H. Akinaga and M. Mizuguchi, J. Phys.: Condens. Matter **16**, S5549 (2004).

[14] T. Dietl *et al.*, Science **287**, 1019 (2000).

[15] S. Dosanjh, P. Dawson, M.R. Fahy, B. A. Joyce, T. Murry, H. Toyoshima, X. M. Zhang, and R. A. Stradling, J. Appl. Phys. **71**, 1242 (1992).