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Zinc-blende CrAs/GaAs multilayers grown by molecular-beam epitaxy

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Abstract

The epitaxial growth of zinc-blende CrAs/GaAs multilayers has been achieved by using the molecular-beam epitaxy method. The crystallographic quality was evaluated by reflection high-energy electron diffraction (RHEED) and cross-sectional transmission electron microscopy (TEM). The increase of the substrate temperature during growth up to 300 °C brings the RHEED pattern to a streak, in contrast to the case at 200 °C. TEM images show the atomically flat surface and interface of the multilayer.

1. Introduction

Since one of major players in present semiconductor-based electronics is the zinc-blende-type semiconductor, such as GaAs and InP, zinc-blende materials showing the ferromagnetic transition at temperatures much higher than room temperature are very much required for novel functions. We reported the successful epitaxial growth of a zinc-blende CrAs thin film and the magnetic properties, together with the prediction of half-metallic behaviour by *ab initio* calculations using the full-potential linearized augmented-plane-wave method [1, 2]. The epitaxial growth of a zinc-blende CrSb was also demonstrated by Zhao *et al* [3]. These reports gave rise to intensive theoretical studies to reveal the potential as the ideal ferromagnet combined with the zinc-blende semiconductor. Sakuma investigated theoretically the stability of the ferromagnetism in zinc-blende CrAs [4]. The first-principles calculations showed a very large effective exchange constant of 150 meV in zinc-blende CrAs with a lattice constant above the experimental one of GaAs, which corresponds to the very high Curie temperature of 1200 K in the framework of the molecular field approximation. The stability of the ferromagnetism as a function of the lattice constant in the range of the experimental values between GaAs and InAs was also pointed out by Sanyal *et al* [5]. The Curie temperature was estimated by adopting mean field theory as 1320 and 1100 K in zinc-blende CrAs with the lattice constant

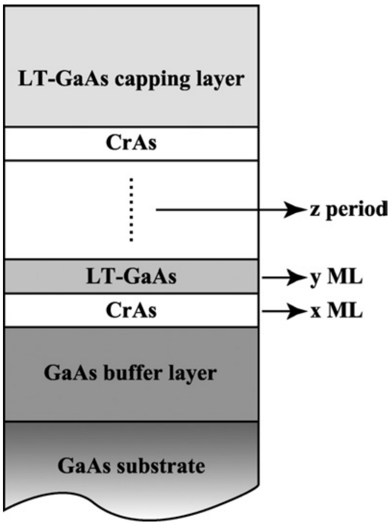

Figure 1. A cross-sectional schematic drawing of the CrAs and GaAs multilayer. The notation LT means 'low-temperature grown'.

of GaAs and InAs, respectively. A Monte Carlo simulation gave the relatively lower Curie temperature of 980 and 790 K for zinc-blende CrAs with the lattice constant of GaAs and InAs, respectively. These values agree well with the calculation by Sakuma, and show the suitability of the material for device applications. This fact contrasts with the case of the zinc-blende transition metal mono-pnictide, such as MnAs, which possesses the possibility to show the half-metallic property but only in the case that the lattice constant exceeds 0.58 nm [6]. The spin-polarization ratio at the surface of zinc-blende CrAs was studied by Galanakis [7]. The full-potential screened KKR method showed that the Cr-terminated (001) surface of zinc-blende CrAs should be half-metallic for both GaAs and InAs experimental lattice constants, while the As-terminated surface loses the half-metallicity.

From the experimental point of view, one of the largest difficulties to utilize this material in a practical device is the very thin critical thickness in the epitaxial growth of zinc-blende CrAs. The zinc-blende crystal structure remains only when the thickness does not exceed 3 nm [2]. To make this material full-fledged, we tried to grow zinc-blende CrAs/GaAs multilayers [8], since *ab initio* calculations showed high spin polarization throughout the entire region of the multilayer in the case that two-monolayer zinc-blende CrAs and two-monolayer GaAs stack alternatively [9]. Our first trial was not successful in terms of the crystal quality [8]. The island-like pattern of the reflection high-energy electron diffraction (RHEED) indicated that the surface and the interface of the multilayer were not completely flat.

Here, in this paper, we show the epitaxial growth of zinc-blende CrAs/GaAs multilayers with an atomically flat surface and interfaces. By optimizing the growth temperature, the crystallographic quality was improved dramatically. A superconducting quantum interference device magnetometer (SQUID) measurement showed a total magnetic moment of about $2\ \mu_{\mathrm{B}}/\mathrm{f.u.}$ of zinc-blende CrAs.

2. Experiment

A molecular-beam-epitaxy (MBE) system (Riber 32P) was used to fabricate zinc-blende CrAs/GaAs multilayers. Figure 1 shows the schematic structure of the multilayer. The

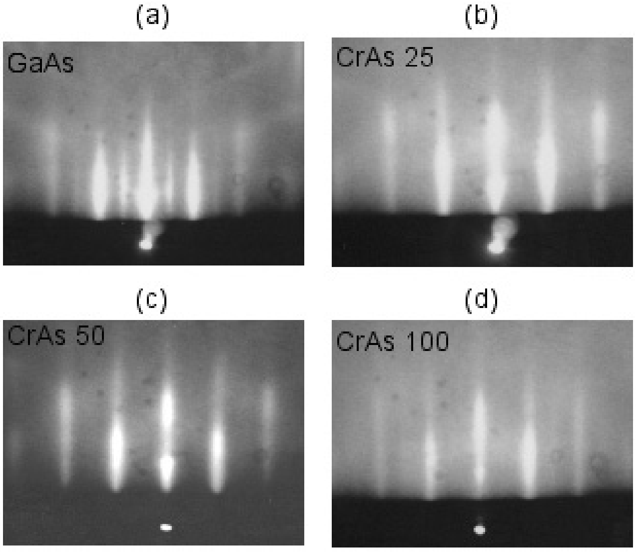

Figure 2. (a) RHEED pattern from the GaAs surface. (b)–(d) RHEED patterns from CrAs surfaces after the growth of 25, 50 and 100 layers of CrAs, respectively.

multilayer was grown on a semi-insulating GaAs(001) substrate. Before starting the growth, the substrate was heated up to about 600 °C in ultrahigh vacuum for thermal cleaning to remove the surface oxidization layer. A buffer layer of GaAs with a thickness of 20 nm was grown at about 580 °C. On the buffer layer, x monolayers (ML) of CrAs and y ML of GaAs were grown alternatively. Growth of the CrAs and GaAs layers was performed by the opening of Knudsen cells for each element of Cr and Ga under exposure of As with a beam pressure of about 6×10^{-4} Pa. The substrate temperature was set at 300 °C, which is about 100 °C higher than that reported in our previous paper [8]. The beam pressure ratio of As/Cr was kept at more than 1000 during the growth. The multilayer was capped by a GaAs layer with a thickness of 10 nm. The surface was characterized by RHEED during the growth.

3. Results and discussion

RHEED patterns obtained from the surface of the GaAs buffer layer and the CrAs layers are shown in figure 2. These patterns were taken during the growth of a sample with $(x, y, z) = (2, 2, 100)$. Although the two-fold reconstruction pattern disappears when the CrAs growth starts, the principal streak lines remain in the pattern from the CrAs surface, even after the 100th growth of the CrAs layer. This result is completely different from that obtained in our previous study [8]. When the substrate temperature was about 200 °C, the pattern from the CrAs surface became weaker and spotty with the increase of the number of the CrAs layers.

Figure 3 shows the cross-sectional transmission electron microscopy (TEM) image of a zinc-blende CrAs/GaAs multilayer with $(x, y, z) = (4, 4, 10)$. Although one can find occasionally a crystallographic defect of the multilayer in the wide view of the TEM image, the zinc-blende atomic image is clearly confirmed in the whole CrAs/GaAs multilayer. The dark-field TEM image is shown in figure 4. A couple of light and dark layers appear in the image with the thickness almost as designed. These TEM images proved the realization of a zinc-blende CrAs layer sandwiched by GaAs layers. It should be noted that no Cr diffusion into the GaAs cap and buffer layers was evidenced in the depth profile of the Cr concentration

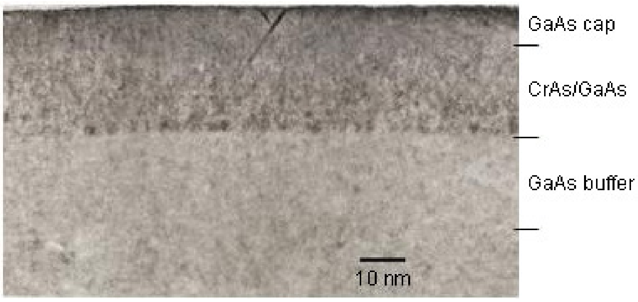

Figure 3. Cross-sectional TEM image of a sample with the structure of [CrAs (4 ML)/GaAs (4 ML)] × 10 layers, capped by a 10 nm GaAs layer and grown on a 20 nm GaAs buffer layer.

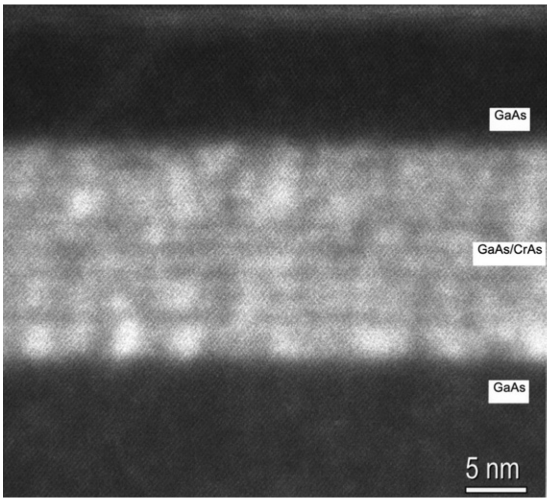

Figure 4. Dark-field cross-sectional TEM image of a sample with the structure of [CrAs (4 ML)/GaAs (4 ML)] × 10 layers, capped by a 10 nm GaAs layer and grown on a 20 nm GaAs buffer layer.

investigated by secondary ion mass spectroscopy (SIMS). Although the SIMS spatial resolution along the direction of the film thickness is not fine enough to identify each CrAs layer, the total thickness of the layer including Cr atoms agreed well with the nominal thickness.

The magnetic properties of a multilayer with $(x, y, z) = (2, 2, 100)$ were investigated by SQUID. Figure 5 shows the magnetization curve measured at room temperature. The curvature shows the so-called soft magnetization. The remnant magnetization is as small as that observed in a single film of zinc-blende CrAs [2]. The saturation magnetization is determined to be 400 kA m^{-1}, corresponding to about 2 $\mu_{\mathrm{B}}/\mathrm{f.u.}$ of CrAs. This value is smaller than 3 μ_{B} predicted by theoretical calculation [9]. The temperature dependence of the magnetization indicated a ferromagnetic transition temperature of about 800 K, which is also lower than the theoretical prediction in the bulk zinc-blende CrAs. It is worth noting that the present magnetization curve does *not* show the irregular change of the curvature. This fact shows a striking contrast to the report in [10], which possibly indicates a second magnetic phase in the sample.

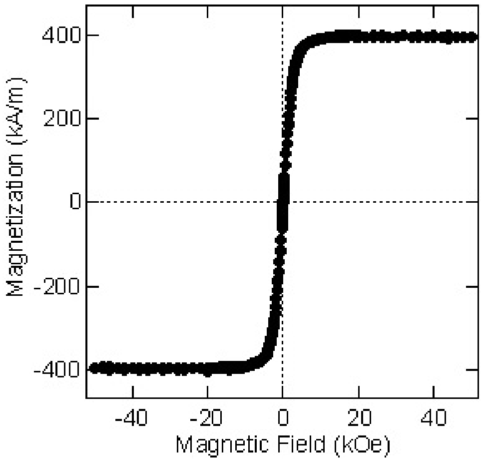

Figure 5. The magnetization hysteresis curve of a sample with the structure of [CrAs (2 ML)/GaAs (2 ML)] × 100 layers, capped by a 10 nm GaAs layer and grown on a 20 nm GaAs buffer layer. The measurement was performed at room temperature. The magnetic field was applied perpendicular to the film plane. The magnetization per volume was calculated by using the nominal and total thickness of all CrAs layers in the sample.

4. Conclusion

We have demonstrated the epitaxial growth of zinc-blende CrAs/GaAs multilayers. By increasing the substrate temperature to 300 °C, the interface and surface flatness were improved. Although the experimentally obtained magnetic moment of the present multilayer was smaller than that predicted theoretically, the multilayer with its high crystallographic quality is certainly a good candidate for a spin-polarized electron source combined with zinc-blende-type semiconductors.

Acknowledgments

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