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Epitaxial growth of zinc-blende CrAs/GaAs multilayer

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Zinc-blende CrAs/GaAs multilayers were grown by molecular beam epitaxy. It was certified that each CrAs layer maintains an epitaxial relationship with the zinc-blende GaAs structure judging from the reflection high-energy electron diffraction observation. The film contains thicker zinc-blende CrAs layers in total than the CrAs thin film directly grown on the GaAs substrate which has the critical thickness of 3 nm. It was clarified that the optimum thicknesses of CrAs and GaAs to keep a good epitaxial relationship are 2 ML and 2 ML, respectively. The electronic structure of the multilayer is thought to be close to that of the (Ga, Cr)As thin film which has 50% of Cr content judging from x-ray absorption spectroscopy measurements. [DOI: 10.1063/1.1455612]

I. INTRODUCTION

Growth of ferromagnetic thin films on semiconductors is now one of the most interesting topics in the material science field because of the possibility of an application to ‘‘spintronic’’ devices. Particularly, the growth of ferromagnetic compounds like MnAs,[1,2] MnSb,[3,4] and (Ga, Mn)As[5,6] on GaAs substrate by a molecular beam epitaxy (MBE) method has been established in recent years. Recently, we have reported the growth of zinc-blende CrAs thin film on GaAs substrates.[7] It is predicted by a theoretical calculation that zinc-blende (zb)-type CrAs has a half metallic band structure.[8] Although it is also clarified that this film has ferromagnetic properties at room temperature by superconducting quantum interference device magnetometer measurements, this film can be grown only with a thickness below 3 nm. We think this limitation is determined by the lattice mismatch between GaAs and CrAs. In this article, we report on the epitaxial growth of zinc-blende CrAs/GaAs multilayer with thicker zb–CrAs layer in total. Electronic structures were also investigated by x-ray absorption spectroscopy (XAS) using synchrotron radiation. We compared a XAS spectrum of a fine multilayer with that of (Ga, Cr)As films consisting of 1% and 50% of Cr atoms as references.

II. EXPERIMENT

A MBE system (Riber 32P) was used to fabricate all the multilayers. Figure 1(a) shows a schematic structure of fabricated multilayers. CrAs/GaAs multilayers were grown on semi-insulating GaAs(001) substrates. Thermal cleaning was performed by annealing the substrate at 600 °C for 10 min just after loading in ultrahigh vacuum to remove the surface oxidation layer. After the growth of a high-temperature GaAs buffer layer with the thickness of 20 nm at 580 °C, CrAs layer (*x* ML) and GaAs layer (*y* ML) were grown alternately. Growths of CrAs and GaAs were performed by opening shutters of Knudsen cells for each element at once at 220 °C.

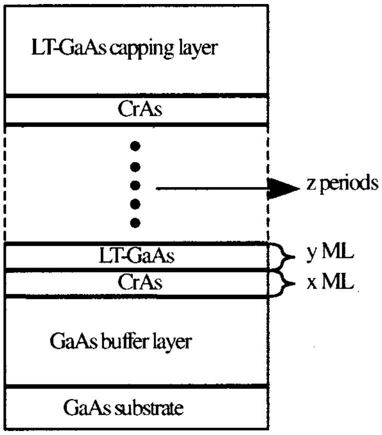

FIG. 1. A cross-sectional schematic structure of CrAs/GaAs multilayers. Thickness of CrAs and GaAs and a growth period were varied.

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TABLE I. Sample names and growth conditions of CrAs/GaAs multilayers. We used (Ga, Cr) As thin films as a reference for the XAS study.

Sample names	Multilayer A	Multilayer B	Multilayer C	Multilayer D	(Ga,Cr)As–E	(Ga,Cr)As–F
x (ML)	2	2	3	2	⋯	⋯
y (ML)	2	1	3	2	⋯	⋯
z (period)	10	10	10	100	⋯	⋯
RHEED	Streaky	Spotty	Spotty	Spotty	Spotty	Streaky

The vapor pressure ratio of As/Cr was set at 100 to 1000. The period of the growth (z period) was set at 10 or 100. The thickness of the last GaAs layers was set at 5 nm to prevent the oxidation of multilayer structures. Combinations of (x, y, z) of fabricated structures are shown in Table I. Surface conditions were monitored by reflection high-energy electron diffraction (RHEED) during all the growth processes.

XAS measurements using synchrotron radiation were performed at the Beam Line 11A of Synchrotron Radiation Research Center in Taiwan to investigate the electronic properties. The photon energy was varied from 570 to 595 eV continuously, and the incidence angle was set at 60° from the normal. The detection angle was set normal to sample surfaces. XAS was measured using partial fluorescence yield using Si detector where the fluorescence from Cr L edge was detected.

III. RESULTS AND DISCUSSIONS

The RHEED pattern from the surface of the first CrAs (2 ML) layer is shown in Fig. 2(a). Although the streak pattern from the zb–CrAs layer was observed, the streaks were weaker than that of the GaAs buffer layer. The first GaAs layer was grown for multilayers A, B, and D on this CrAs layer. As for the multilayer C, the first GaAs layer was grown on the CrAs layer with the thickness of 3 nm which is the critical thickness for epitaxial growth at present. Figure 2(b) is a RHEED pattern from the surface of the tenth CrAs layer of the multilayer A. The streaky pattern remained. We believe that epitaxial growth of zb–CrAs/GaAs can be achieved under the growth condition of the multilayer A. The total thickness of CrAs layer for multilayer A is estimated to be about 6–7 nm. It can be said that the zb–CrAs layers twice thicker than the critical thickness of the single CrAs film have grown on GaAs substrate by fabricating the multilayer A. On the other hand, different RHEED patterns were observed when other series of multilayers (multilayer B and multilayer C) were grown. The RHEED patterns from the surface of the tenth CrAs layer of the multilayer B and the multilayer C are shown in Fig. 2(c) and 2(d), respectively. In both cases, spotty patterns were observed. It can be said that when the thickness of CrAs layer is greater than 2 ML ($x>2$) and the thickness of GaAs layer is smaller than 2 ML ($y<2$), CrAs and GaAs do not grow epitaxially. We believe that the set of $(x,y)=(2,2)$ is the optimum condition to realize the epitaxial zb–CrAs/GaAs multilayer.

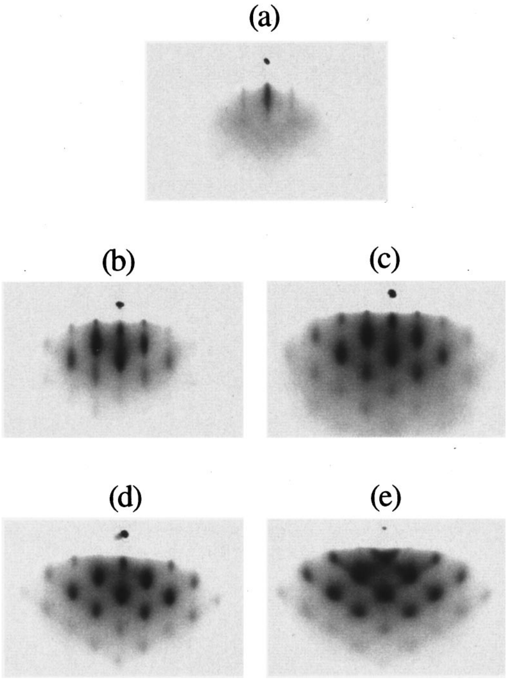

FIG. 2. RHEED patterns from the surfaces of: (a) first CrAs layer of multilayer A, (b) tenth CrAs layer of multilayer A, (c) tenth CrAs layer of multilayer B, (d) tenth CrAs layer of multilayer C, and (e) one hundredth CrAs layer of multilayer D. These patterns were taken along the direction of GaAs[110].

We also investigated the structural property of thicker films than the multilayer A with the growth condition of $(x,y)=(2,2)$. When the growth was continued after the growth of the multilayer A, it was observed that the streaky RHEED pattern gradually changed to a spotty pattern with increasing the growth period. Extra spots which indicate a formation of unknown phase have appeared at around $z=50$. Figure 2(e) shows a RHEED pattern from the surface of the hundredth CrAs layer of multilayer D ($z=100$). A perfectly spotty pattern from a top CrAs layer was observed. It is thought that CrAs layers do not keep zinc-blende structure in this thicker multilayer judging from the RHEED observation.

Figure 3 shows the XAS spectra of the multilayer A, $(\mathrm{Ga,Cr})\mathrm{As–E}(\mathrm{Cr}=50\%)$, and $(\mathrm{Ga,Cr})\mathrm{As–F}(\mathrm{Cr}=1\%)$. Solid triangles and open triangles indicate the Cr L_2 peak positions and Cr L_3 peak positions, respectively. Three spectra were normalized by peak heights, and integral backgrounds were subtracted for each spectrum. It should be noted that the multilayer A has Cr L_2 edges and Cr L_3 at almost the same photon energies as the $(\mathrm{Ga,Cr})\mathrm{As–E}(\mathrm{Cr}=50\%)$, while the (Ga, Cr)As–F ($\mathrm{Cr}=1\%$) has Cr L_2 and Cr L_3 edges at about 1 eV lower photon energies. These results suggests the similarity between the multilayer A and the (Ga, Cr)As–E in terms of the electronic structures and

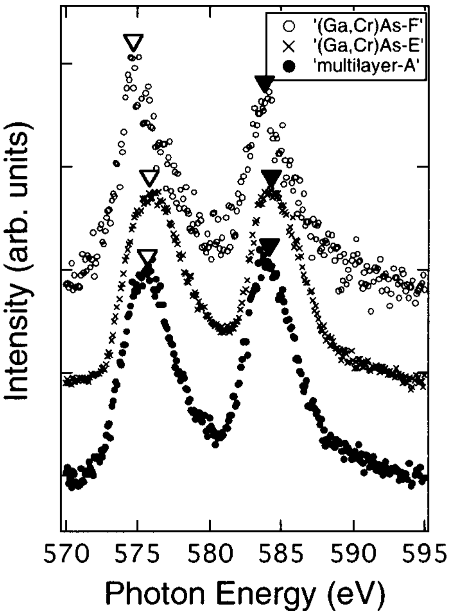

FIG. 3. XAS spectra of the multilayer A, (Ga, Cr)As–E(Cr=50%), and (Ga, Cr)As–F(Cr=1%). Measurements were performed by a soft x-ray partial fluorescence yield method. Solid triangles and open triangles indicate the Cr L_2 peak positions and Cr L_3 peak positions, respectively. Three spectra were normalized by peak heights, and integral backgrounds were subtracted for each spectrum.

the chemical bonding around Cr atoms. Although the reason why the electronic structures are quite different between the (Ga, Cr)As–E and the (Ga, Cr)As–F is not clear at present, we think that the (Ga, Cr)As–E is in the CrAs-segregated state, while the (Ga, Cr)As–F is in the diluted magnetic semiconductor state. It might be also supported by the fact that Cr content of these two films is almost the same if these were fabricated as designed. We believe the coincidence of peak position is derived from the consistency in the chemical shift of the two films. This fact may also support our assumption that a (Ga, Cr)As film with the Cr content of 50% includes Cr–As precipitates which will be shown elsewhere.[9] It has been reported that Mn atoms interdiffuse into GaAs layers in MnAs/GaAs multilayers which were fabricated by a digital growth.[10] An extent of interdiffusion for our zb-Cr–As/GaAs system has not been clarified yet. More detailed discussion is needed to clarify the electronic structures of multilayers.

IV. CONCLUSIONS

In conclusion, the epitaxial zinc-blende CrAs/GaAs multilayers which include 20 ML CrAs layers in total has been successfully grown. It is found that when the thickness of CrAs layer is greater than 2 ML and the thickness of GaAs layer is smaller than 2 ML, CrAs and GaAs do not grow epitaxially. The multilayer with a total thickness of 200 ML CrAs has been grown by an island growth mode judging from the RHEED pattern, which means the existence of limitation of epitaxial growth of multilayer. XAS study for epitaxial multilayers has revealed that the electronic structure is very close to that of (Ga, Cr)As with the Cr content of 50%.

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