Structural and magnetic properties of epitaxial Fe$_3$Si/GaAs heterostructures

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Abstract

High-quality epitaxial Fe$_3$Si films (1 0 0) were grown on the GaAs (1 0 0) surface using molecular beam epitaxy (MBE). High-resolution X-ray diffraction analysis using an X-ray energy of 12.38 keV from synchrotron radiation gave a narrow rocking curve of $\sim$0.014° for the Fe$_3$Si (0 0 6) reflection, with a lattice mismatch between the film and GaAs of $\sim$0.25% in the normal direction. Square-shaped M–H loops with a typical moment of 660 emu/cm$^3$ at 10 K and the easy axis along [1 0 0] were obtained for films grown at a substrate temperature ($T_s$) of 150°C, and the M–H loops at low fields show fine features for films grown at a $T_s$ of 200–300°C. A two-step growth procedure with the initial growth at 150°C and subsequently ramping to 250°C was applied to minimize the interfacial reactions, thus to achieve abrupt interfaces.

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1. Introduction

Fe$_3$Si, a ferromagnet with a $T_c$ of 840 K, has a cubic DO$_3$ structure with its lattice constant in close match with GaAs (1 0 0) [1–6]. Fe$_3$Si has recently gained much attention, because it can be considered as a Heusler alloy of Fe$_2$FeSi having two distinct crystallographic and magnetic Fe sites. Since some Heusler alloys are predicted to be half metals with 100% spin polarized at the Fermi level, Fe$_3$Si may be a promising candidate for effective spin injection [7]. Hong et al. [1–4] reported in 1990s the first success of achieving high-quality epitaxial Fe$_3$Si of only several monolayers thick on GaAs (1 0 0). More recently, Herfort et al. [5,6] have established an optimized growth temperature range and fine tuned the Fe$_3$Si composition. Ionescu et al. [8] have reported extensive studies of structural, magnetic, electronic, and spin transport properties.

In this work a high degree of structural perfection with nearly perfect lattice match was achieved in the hetero-epitaxy between a metallic compound Fe$_3$Si and a compound semiconductor GaAs. High-resolution X-ray diffraction (XRD) measurements gave a narrow rocking curve of about 0.014° for the Fe$_3$Si (0 0 6) reflection with a lattice mismatch between the film and GaAs determined to be $\sim$0.25% in the normal direction. Square-shaped M–H loops were obtained for films grown at a substrate temperature ($T_s$) of 150°C with the easy axis along [1 0 0], and the M–H loops at low fields show fine features for films grown at a $T_s$ of 200–300°C. A two-step growth procedure with the initial growth at 150°C and subsequently ramping to 250°C was applied to minimize the interfacial reactions, as confirmed by in situ X-ray photoelectron spectroscopy (XPS) and
high-resolution cross-sectional transmission electron microscopy (HRTEM).

2. Experimental procedure

The epitaxial growth of FeSi on GaAs was carried out in a multi-chamber molecular beam epitaxy (MBE) system, which includes a solid-source GaAs-based III–V MBE chamber and a metal deposition chamber. After the initial GaAs (1 0 0) epitaxial growth in the III–V chamber, the samples were transferred in situ to the metal chamber. A Ga-stabilized surface exhibiting a 4 × 6 reconstructed RHEED pattern of reflection high-energy electron diffraction (RHEED) was obtained after reheating the substrates to 550 °C before FeSi deposition. The substrate temperature ($T_s$) was then lowered to 150–300 °C to commence the FeSi growth with the film surface quality continuously monitored by RHEED. Typical growth rate is 1.17 nm/min. An amorphous Si capping layer was deposited to protect the FeSi films from being oxidized or contaminated. The XPS spectrum shows the absence of iron oxide, suggesting the effectiveness of the amorphous Si in protecting FeSi.

XRD measurements were carried out at BL17B beam line of National Synchrotron Radiation Research Center (NSRRC) in Taiwan, and also at SPring-8 in Japan. X-rays at NSRRC are monochromatized by a pair of Si (1 1 1) crystals with a beam energy of 8 keV for most measurements. An eight-circle diffractometer with two pairs of slits between the sample and detector was employed. A typical resolution of better than 4 × 10$^{-4}$ nm$^{-1}$ was adopted for this work. X-ray reflectivity (XRR) was measured using a two-circle diffractometer operated at 50 kV and 200 mA with Cu-K$_{α1}$ radiation ($λ = 1.54$ Å). By using the modeling fits of Bede REFS Mercury Code, the film thickness, interfacial roughness and electron density distribution were determined [9,10]. For the HRTEM study a JEOL 2100F field-emission TEM was used to examine the film microstructure and interfaces. The magnetic properties of FeSi were studied using the superconducting quantum interference device (SQUID) magnetometer. After depositing FeSi films a few monolayers thick, the wafers were transferred in situ under a vacuum of 10$^{-10}$ Torr to the XPS analysis chamber equipped with a SPECS-PHOIBOS-150 analyzer.

3. Results and discussion

Bright streaky reconstructed RHEED patterns were observed immediately in the initial growth of FeSi 1–2 ML thick deposited on GaAs (1 0 0) at a $T_s$ of 250 °C, and the patterns continued to sharpen with increasing film thickness as shown in Fig. 1. The presence of Kikuchi arcs indicated that a high-quality crystalline surface is attained. This is in strong contrast to the pure Fe (1 0 0) on GaAs (1 0 0) in which the initial growth showed broad RHEED patterns, suggesting pronounced interactions between Fe and GaAs.

X-ray diffraction and RHEED studies showed that the film structural quality improves with the $T_s$ increasing from 150 to 250 °C. For example, a FeSi film 22 nm thick deposited with a $T_s$ of 250 °C exhibits interference fringes (Pendello¨sung) up to 20th order surrounding the diffraction peak of FeSi (0 0 2), indicating a high degree of crystallinity and sharp interfaces. However, it was very difficult to separate the diffraction peak of the films from the main GaAs reflection due to the lattice parameters being too close. Consequently, it was not possible to measure the rocking curves on the film diffraction peaks. Equal difficulties are met in the in-plane scans to study in-plane film structural order.

Nevertheless, by changing the energy of incident X-ray beam from 8 to 12.38 keV at 12B2 beam line in SPring-8, we were able to separate FeSi (0 0 6) and GaAs (0 0 6) reflections, with the fitting carried out using the dynamical diffraction theory. The inset shows the FeSi (0 0 6) rocking curve.

Fig. 1. RHEED patterns taken at the initial growth (1 nm) and the end (14 nm) of the FeSi film on GaAs (1 0 0) along the FeSi in-plane [1 1 0] and [1 0 0] axes.

Fig. 2. Measured (dots) and fitted (line) XRD normal scan of FeSi (0 0 6) and GaAs (0 0 6) reflections, with the fitting carried out using the dynamical diffraction theory. The inset shows the FeSi (0 0 6) rocking curve.
The scattered intensity was simulated with the Bede scientific rocking curve analysis using a dynamical simulation (RADS) program which includes dynamical diffraction. The simulated results matched very well with the experimental data, as shown in Fig. 2. With the aid from the simulation we determined a lattice mismatch ~0.25% between Fe3Si and GaAs.

The magnetic properties were measured at both 10 and 300 K, with the M–H loops (10 K) taken with the applied magnetic field parallel to the [1 0 0] direction in the plane (Fig. 3). Typical magnetic moments are 660 emu/cm3 at 10 K, 600 emu/cm3 at 300 K, respectively. The hysteresis loops at low fields showed fine features that varied with film growth temperature. M–H loops with regular normal shapes were obtained for films grown at Ts of 150°C; however, the M–H loops with a-step shape were observed for films grown at higher Ts of 200, 250, and 300°C. These may be resulted from interfacial reactions occurring between the film and the substrate at Ts greater than 150°C. Therefore, we have adopted a two-step growth procedure with the initial Fe3Si 2 nm thick grown at 150°C to minimize the interfacial reactions, then the Ts was ramped to 250°C to attain high-quality epitaxial growth.

In order to probe the interfacial reactions, a Fe3Si (3 nm)/GaAs film was deposited at Ts of 150°C and in situ transferred to the XPS chamber to determine the chemical states. Fig. 4 shows in situ XPS spectra measured on the as-grown Fe3Si (3 nm)/GaAs. All of the Ga 3d peaks can be fitted with two components: the first component, labeled as Ga–As, is the Ga 3d of bulk GaAs. The second component, labeled as Ga–Fe, is shifted by 1.1 eV to a lower binding energy with respect to the peak of bulk GaAs, and is probably due to the formation of a reacted phase at the Fe3Si/GaAs interface. The intensity of the Ga–As peak decreased monotonically, as expected, when the incident angle of soft X-ray beam was changed from 0° to 75° relative to surface normal. However, the intensity of Ga–Fe peak was almost unchanged (Fig. 4(b)), indicative of the occurrence of chemical reaction at the interface. The interfacial reaction is expected to be more pronounced...
when the deposition takes place at higher $T_s$ of 250 °C. The XPS measurement indicated that the interfacial reaction may be limited to a first few MLs (~2–3 ML) at $T_s$ of 150 °C. However, the phenomenon was not revealed from the M–H loops for the reason that the un-reacted film, relatively thick in comparison with the interfacial layer, dominated the overall magnetic property.

The cross-sectional HRTEM image (Fig. 5) showed that a high-quality epitaxial Fe$_3$Si thin film on GaAs was obtained using the two-temperature growth procedure. The atomically smooth and chemically abrupt interface may be one of the sharpest transitions from metals to semiconductors ever demonstrated. The thickness of the Fe$_3$Si film estimated from the TEM images is 14.4 nm, consistent with the thickness determined from the XRR analysis (Fig. 5 inset) which also gave a roughness of the Fe$_3$Si/GaAs interface of ~0.4 nm from a theoretical modeling of the bi-layer.

4. Conclusion

In this work single crystal films of Fe$_3$Si with lattice constants closely matched to GaAs were epitaxially grown on GaAs (100), achieving a high degree of structural perfection and atomically smooth interface. Using an X-ray energy of 12.38 keV in XRD analysis we determine a narrow rocking curve of about 0.014° for the Fe$_3$Si (0 0 6) reflection, with a lattice mismatch between the film and GaAs of ~0.25% normal to the film plane. With the diagnosis of in situ XPS chemical analysis for the interface and ex situ magnetic measurements, we have applied a two-step temperature growth procedure to minimize the interfacial reactions between Fe$_3$Si and GaAs to achieve better spin injection efficiency as required for spintronic applications.

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