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Atomic structure of postgrowth annealed epitaxial Fe/(001)GaAs interfaces

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The interfacial atomic structure of epitaxial Fe films grown by molecular beam epitaxy on $c(4 \times 4)$ reconstructed (001) GaAs was investigated using high-angle annular dark-field imaging in scanning transmission electron microscopy. No extended interfacial reaction phase is observed and the image contrast is discussed in terms of the interface atomic configuration. The images show an As-terminated semiconductor. The interface consists of a single partially occupied plane inserted between the Fe film and the GaAs, which most likely is occupied by Fe. This interface structure provides strong evidence for preferential Fe–As bonding across the interface. © 2008 American Institute of Physics. [DOI: 10.1063/1.2990622]

Epitaxial Fe/GaAs heterostructures are prototypical ferromagnetic/semiconductor spin injection systems with many attractive properties, combining small lattice mismatch and the high Curie temperature and spin polarization of Fe with the high carrier mobilities of GaAs.¹ Theoretical predictions suggest that a prerequisite for efficient spin injection are Fe/GaAs interfaces that are abrupt and devoid of interdiffusion, roughness, or reaction products.² Furthermore, even for abrupt interfaces, the spin injection efficiency and polarization depend sensitively on the interface atomic arrangement and bonding^{2,3} and postgrowth annealing.^{4,5} While scanning tunneling microscopy has been widely applied to the investigation of the initial stages of Fe growth on GaAs,^{6–8} the technique cannot provide information about the interface once a film has formed. Photoemission studies have suggested interfacial reactions with As surface segregation.⁹ *In situ* x-ray photoelectron spectroscopy (XPS) studies of Fe growth at -15°C on (001) GaAs found that approximately 1 ML of Ga atoms at the interface has more metallic bonding than in GaAs, which would be consistent with the formation of an ultrathin (≤ 3 ML) $\text{Fe}_3\text{Ga}_{2-x}\text{As}_x$ layer.⁸ No change in bonding was observed by *in situ* XPS for anneals $\leq 250^\circ\text{C}$.¹⁰

Cross-section high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM or Z-contrast imaging) is capable of resolving the projected interface atomic structure after growth and annealing treatments. To date, only one study has applied Z-contrast imaging to the Fe/GaAs interface.¹¹ In this study it was concluded that the interface consists of a Ga-terminated semiconductor followed by a plane in which Fe and As columns alternate. This particular structure was also considered in simulations employing the density-functional theory, where it was found to be relatively stable at low Fe coverage.¹² For thicker Fe layers the abrupt interface was predicted to be most stable.^{12,13} In Ref. 11 images were only obtained along one direction, which can make unambiguous identification of the interface structure difficult. In this letter, we revisit the atomic structure of molecular beam epitaxy (MBE) grown

Fe/GaAs interfaces. By imaging the interface along two perpendicular directions, we find an interface structure that differs from the one previously reported.

Substrates were (001) GaAs covered with a 500 nm thick GaAs buffer layer, which was grown by MBE at 580°C . The in-plane orientation was confirmed using *in situ* reflection high energy electron diffraction of the $(2 \times 4)/c(2 \times 8)$ surface reconstruction following the growth of the GaAs buffer. Cooling under an As_4 flux was used to obtain the $c(4 \times 4)$ As-rich reconstructed surface. Epitaxial Fe layers with the bcc structure were grown to a thickness of 2.5 nm by MBE at a substrate temperature of $\leq 50^\circ\text{C}$ using a high-temperature effusion cell for Fe and a procedure reported elsewhere.⁸ Following Fe growth, the samples were *in situ* annealed at 200°C for 1 h. A 5 nm thick Al capping layer was grown to prevent oxidation. Epitaxial Fe contacts fabricated in the same manner on $\text{Ga}_{1-x}\text{Al}_x\text{As}$ spin light emitting diode structures have led to steady state spin polarized injection signals of $\sim 30\%$, corresponding to spin injection efficiencies $\sim 50\%$.¹⁴ Cross-section TEM samples along two perpendicular $\langle 110 \rangle_{\text{GaAs}}$ directions were prepared by standard wedge polishing followed by Ar-ion milling for final thinning. In the following, we will use the conventional notation for the specific $\langle 110 \rangle_{\text{GaAs}}$ directions, which places the Ga atoms at the (0,0,0) positions of the zinc blende structure. The As atoms in each dumbbell then point toward the interface along $[110]_{\text{GaAs}}$ and away from it along $[1\bar{1}0]_{\text{GaAs}}$. Note that this is different from the notation used in Ref. 11. A FEI Titan 80–300 TEM/STEM with a supertwin lens ($C_s = 1.2$ mm) and field-emission gun operated at 300 kV was used for HAADF imaging. The convergence semiangle was 9.6 ± 0.15 mrad and the inner detector semiangle was 43 ± 1 mrad.

Figures 1(a) and 1(b) show HAADF images along $[1\bar{1}0]_{\text{GaAs}}$ and $[110]_{\text{GaAs}}$, respectively. The interface between zinc blende GaAs and Fe appears atomically abrupt and free of readily identified reaction phases. The images did not allow for distinguishing between As and Ga columns. The dumbbells were, however, clearly visible and, along with the known orientation of the sample, allowed for determination of the interface termination. Along $[1\bar{1}0]_{\text{GaAs}}$, a step is apparent near the center of the image [see arrow in Fig. 1(a)]. The interface atomic structure must be determined using regions

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FIG. 1. (Color online) HAADF images of the GaAs/Fe interface along (a) $[1\bar{1}0]_{\text{GaAs}}$ and (b) $[110]_{\text{GaAs}}$. The arrow in (a) indicates the location of an interface step. Arrows in (b) indicate additional intensity between the first complete Fe layer and the terminating As layer. Higher magnification images along $[1\bar{1}0]_{\text{GaAs}}$ and $[110]_{\text{GaAs}}$ are shown in (c) and (d), respectively. The overlays show the position of the atom columns near the interface. The image in (a) has been corrected for drift.

sufficiently thin to not contain overlapping steps along the beam direction. Magnified images from thin areas along the two directions [Figs. 1(c) and 1(d)] showed that the GaAs surface was terminated by an As layer, as expected from the As-rich $c(4 \times 4)$ reconstruction. Along $[110]_{\text{GaAs}}$, but not along $[1\bar{1}0]_{\text{GaAs}}$, intensity was present between the terminating As and the first complete Fe layer at the positions indicated by arrows in Fig. 1(b) and squares in Fig. 1(d). The intensity of these positions was much lower than that of the Fe, Ga, or As columns on either side of the interface, indicating that the atomic columns in this plane were only partially occupied. The lower intensity at these positions could also be seen from the intensity line scan across an image with the high frequency (noise) components removed by Fourier filtering with a 15 nm^{-1} diameter circular aperture (Fig. 2). The partially occupied columns were located close to positions normally occupied by Ga in bulk GaAs, but the projected distance between them and the As columns was $0.223 \pm 0.01 \text{ nm}$, which is smaller than the projected As–Ga distance. As a result, interfacial dumbbells that would appear along $[1\bar{1}0]_{\text{GaAs}}$ were separated by only 0.12 nm , which is below the resolution of the instrument. The presence of the

FIG. 2. (Color online) (a) Filtered image along $[110]_{\text{GaAs}}$ to remove the noise. (b) Intensity line profile across the interface along the line indicated in (a). The position marked with an arrow in (b) is the same position shown by the arrow in (a).

partially filled interfacial plane resulted in a relatively large distance between the As plane and the first complete Fe film plane ($0.25 \pm 0.03 \text{ nm}$). One possible explanation for weak contrast in the interface plane could be that it is occupied by residual As from the As-rich $c(4 \times 4)$ reconstructed GaAs surface; however, in this case As–As or Ga–As dimers¹⁵ should have been observed along $[1\bar{1}0]_{\text{GaAs}}$. Thus, the atomic columns in the interface plane corresponded most likely to partially occupied (relative to bcc) Fe columns. The atom columns in the first few Fe layers occasionally appeared slightly brighter than the surrounding Fe. The change in contrast was random, especially for the thinnest sample regions. The constant change could have been caused by outdiffusion of As, which is expected given the As-rich growth surface. Alternatively, the contrast could also result from the outdiffusion of Ga, which would correspond to the more metallic bonded Ga atoms detected by XPS at the interface.⁸

An interface structure model with a partially occupied Fe plane that is consistent with the contrast in the HAADF images is shown in Fig. 3. Note that the model requires an Fe interface plane with not only missing Fe columns compared

FIG. 3. (Color online) Structure model of the interface (plan-view and cross-sections along two perpendicular $\langle 110 \rangle_{\text{GaAs}}$ directions) based on the experimental HAADF images. The model assumes a partially occupied Fe plane between the terminating As plane and the first fully occupied Fe layer. In the plan-view image ($[001]$ projection) three layers are shown (from substrate to interface): Ga layer, As layer, and the partially occupied Fe layer. In the cross-section view (right), the projected distances estimated from the experimental images are indicated.

to a bcc Fe layer, but also partial occupancy of the remaining columns. From the image contrast, it was estimated that about half the Fe was missing in the partially occupied columns, as shown in Fig. 3. The structure agrees with the preference for Fe–As bonding predicted in first-principles calculations of the cohesive energy of Fe–As relative to that of Fe–Ga.³ The interface structure differs from the one determined in Ref. 11; in particular, in Ref. 11 it was proposed that the interface plane consisted of a single mixed As–Fe plane with no partially occupied interface plane. The interface structure shown in Fig. 3 has not yet been considered in theoretical simulations. Models in the literature have either assumed an abrupt interface or additional Fe atoms at interstitial sites in the semiconductor,^{12,13} which were not observed here. Furthermore, unannealed samples showed the same interface structure (though note that TEM sample preparation involves an anneal at 120–150 °C), indicating that the structure reported here is a preferred one for the Fe/GaAs interface.

The magnitude and sign of the spin injection has been found to depend critically on the interfacial formation.^{5,10} We speculate that the specific interfacial structure observed here is critical to the high spin injection efficiency and that this structure may be best obtained by postgrowth annealing at a moderate temperature that avoids extensive Fe–GaAs interfacial reactions.¹⁶ We suggest that future theoretical simulations should compare the stability and magnetic spin and transport properties of the interface structure shown in Fig. 3 with respect to those previously considered,^{11–13} as complete understanding of this interface is critical in maximizing the potential of the GaAs/Fe system for spin injection applications.

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