

## Kinetic stabilization of Fe film on GaAs(100): An *in situ* X-ray reflectivity study

D.Y. Noh<sup>a</sup>, T.C. Kim<sup>a</sup>, Y. Kim<sup>a</sup>, J.-M. Lee<sup>b</sup>, S.-J. Oh<sup>b</sup>, J.-S. Kim<sup>c,\*</sup>

<sup>a</sup> School of Photon Science and Technology, Department of Materials Science and Engineering, Gwangju Institute of Science and Technology, Gwangju 500-712, Republic of Korea

<sup>b</sup> Department of Physics and Astronomy and Center for Strongly Correlated Materials Research, Seoul National University, Seoul 151-742, Republic of Korea

<sup>c</sup> Department of Physics and Astronomy Sook-Myung Women's University, Seoul 140-742, Republic of Korea

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### Abstract

We study the growth of Fe films on GaAs(100) at a low temperature, 140 K, by *in situ* X-ray reflectivity (XRR) using synchrotron radiation. The XRR curves are well modeled by a single Fe layer on GaAs both at the growth temperature and after annealed at the room temperature. We found that the surface became progressively rougher during the growth with the growth exponent,  $\beta_S = 0.43 \pm 0.14$ . The observed  $\beta_S$  is attributed to the restricted interlayer diffusion at the low growth temperature. The change of the interface width during growth was minimal. When the Fe film was annealed to room temperature, the surface smoothed, keeping the interface width almost unchanged. The confinement of the interface derives from that the diffusion of Ga and As proceeds via the inefficient bulk diffusion, and the overlying Fe film is kinetically stabilized.

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Recently, ferromagnetic metal–semiconductor heterostructures have drawn intense attention for their applications in the spintronic devices such as spin-polarized field effect transistors and spin-polarized light emitting diodes [1]. Such spintronic devices require a highly polarized spin source and efficient spin injection through the interface of the heterostructures. The Fe/GaAs system has been studied as a prototype of the heterostructures, because high quality, epitaxial body centered cubic (bcc) Fe films can be grown on GaAs surfaces due to their small lattice mismatch of about 1.3% [2,3]. Moreover, the spin injection from the Fe film into GaAs(100) has been realized even at room temperature [4]. However, the system has enjoyed limited success, because the magnetic polarization of the Fe film and in turn the spin injection efficiency has often seri-

ously degraded due to the formation of nonmagnetic or antiferromagnetic Fe alloys with Ga and especially As out-diffused from the bulk [5–7].

There have been several attempts to suppress the alloy formation: sulfuric passivation of GaAs surface [8] and insertion of a thin interlayer such as Er layer between Fe film and GaAs [9]. These have reduced the outdiffusion and the alloy formation of Ga and As. Segregated As atoms, however, have still been observed on the surface [9]. Chye et al. [10] grew Fe films on GaAs(100) at a low temperature ( $\sim 120$  K), and found that magnetic properties were improved significantly. The alloy formation seemed to be much suppressed. However, structural and chemical analysis confirming the assumed growth behavior of Fe/GaAs(100) at the low temperature has not been done. Only recently, photoelectron spectroscopy has been performed on Fe films grown on GaAs(100) at 130 K, and suggested that the alloy formation and outdiffusion of both

\* Corresponding author.

E-mail address: [jskim@sookmyung.ac.kr](mailto:jskim@sookmyung.ac.kr) (J.-S. Kim).

Ga and As be effectively suppressed [11]. The atomic structure of the interface has also been studied by Z-contrast electron microscopy [12].

In the present work, we studied the growth and thermal stability of Fe films grown on GaAs(100) at 140 K by *in situ* X-ray reflectivity (XRR). Most importantly, we observed the formation of virtually pristine Fe film on GaAs(100). The surface width ( $\sigma_S$ ) of the film increases following a power law with the growth exponent  $\beta_S \approx 0.43$ . This reflects that the interlayer diffusion is restricted at the low growth temperature due to the insufficient thermal energy of the adatoms to overcome the step Ehrlich Schwoebel barrier. The interface width was kept below 5 Å indicating that the alloy formation was virtually suppressed. Further, the Fe film was thermally stable against annealing up to room temperature. The confinement of the interface alloy is ascribed to the inefficiency of bulk diffusion process by which Ga and As diffuse into the Fe film.

All the measurements have been performed in an ultra-high vacuum (UHV) X-ray scattering chamber at 5C2 beam line of Pohang light source (PLS). The base pressure of the chamber was below  $8 \times 10^{-10}$  Torr. Clean GaAs(100) substrate surface was obtained by repeated cycles of 0.5 keV  $\text{Ar}^+$  ion sputtering with the incident angle of  $45^\circ$  from the surface normal around 300 K and annealing up to 850 K. The sample temperature was monitored by both an optical pyrometer and a K-type thermocouple attached near the sample.

In order to observe the evolution of the surface morphology upon Fe deposition, specular XRR was measured. The incident X-ray was focused vertically by a mirror and monochromatized to a wavelength  $\lambda = 1.24$  Å by a double bounce Si(111) monochromator. The deposition of Fe is carried out in a UHV chamber mounted on four-circle X-ray diffraction goniometer (2 + 2 mode).

A commercial electron beam evaporator (EFM3, Omicron) was used to evaporate Fe (purity of 99.99%) at the rate of 0.026 Å/s. During the deposition, the chamber pressure was maintained below  $1 \times 10^{-9}$  Torr and the substrate was held at 140 K. The deposition of Fe and the XRR measurement were alternated. The referred nominal thickness  $t$  of the Fe film is the accumulated amount of the deposited Fe.

Fig. 1a shows the specular XRR curves as a function of the out-of-plane momentum transfer,  $q_z$ . We subtracted the longitudinal diffuse reflectivity, which was measured by tilting the sample off in the  $\theta$  rocking direction just outside the specular rod, from the measured specular XRR to separate out the true specular component. In a clean GaAs(100), significant amount of the reflected intensity was observed at  $q_z$  as large as  $\approx 0.7$  Å $^{-1}$ , which indicates that the clean GaAs substrate was smooth. The XRR decays exponentially,  $\sim e^{-\sigma^2 q_z^2}$  at large  $q$  values. The surface roughness  $\sigma_S$  obtained by fitting the XRR to Parratt's formula [13], is  $2.3 \pm 0.2$  Å. As the Fe film was deposited, however, the slope of the specular XRR curve decayed more steeply, implying that the growth front became rougher. The mod-

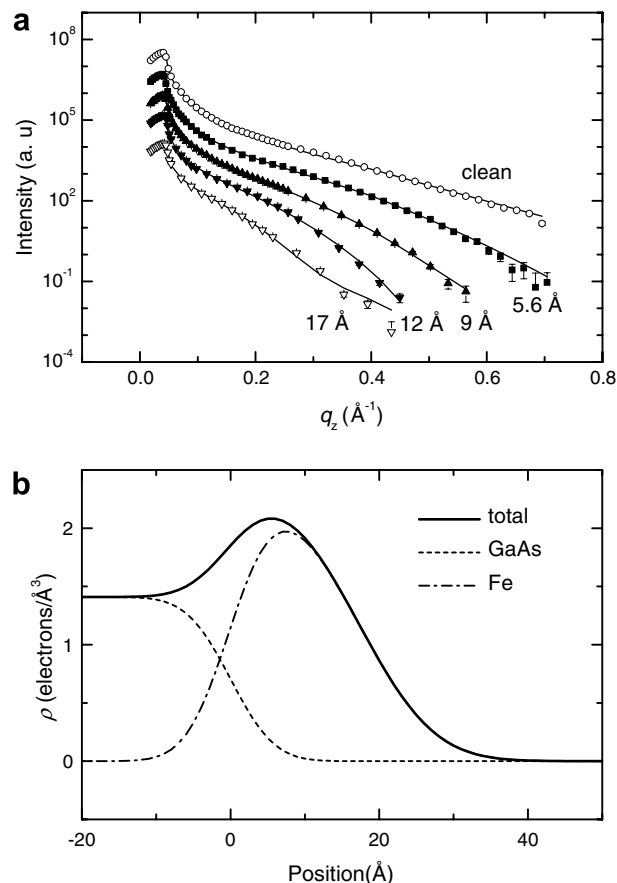


Fig. 1. (a) Specular X-ray reflectivity as a function of the out-of-plane momentum transfer,  $q_z$  for various Fe film thickness,  $t$  at 140 K. The solid lines are the results of the fit by the effective density model based on Parratt's formalism. The curves are vertically shifted with respect to each other for clarity. (b) The best-fit electron density profiles of 17 Å-thick Fe film.

ulation in the XRR curves is extremely weak since the interference between the X-rays reflected from the rough surface (vacuum/Fe) and those reflected from the interface (Fe/GaAs) hardly occurs.

In a typical analysis using the Parratt's formalism, a system is described by a stack of thin slabs with finite interface roughnesses that are much smaller than the thicknesses of the slabs. The root-mean-square roughness or the interface roughness  $\sigma_I$  between the  $i$ th and  $i + 1$ th slabs are defined as  $\sigma_I \equiv \sqrt{\langle h_i^2(\vec{r}) \rangle}$ , where  $h_i(\vec{r})$  represents the height variation of the interface at lateral position  $\vec{r}$ . The XRR is evaluated by calculating the interference of X-rays reflected by each interface.

In the present case, however, the film thickness is rather thin, and the interface roughness can be comparable to the thickness. To take this fact into account, we adopted so called 'effective density model' [14]. In this model, the electron density profiles of a film  $\rho_f(z)$  and a substrate  $\rho_s(z)$  are evaluated for given set of interface widths,  $\sigma_S$  and  $\sigma_I$ . The electron density profile of the substrate is described using an error function  $0.5\rho_s^0\{\text{erf}(z/\sqrt{2}\sigma_I) + 1.0\}$ , and the film density is given by  $0.5\rho_f^0\{\text{erf}[(z-t)/\sqrt{2}\sigma_S] - \text{erf}(z/\sqrt{2}\sigma_I)\}$ ,

where  $t$  is the film thickness. In calculating the XRR, the system (the film on top of the substrate) is sliced into many layers of atomic scale thickness,  $\sim 1$  Å. The effective electron density of each slice is given by the sum of the electron densities of the film and the substrate at the position of the slice. From the effective electron density, one can obtain the index of refraction of the slice,  $n = 1 - \lambda^2 r_e \rho_{\text{eff}} / 2\pi + i\beta$ , where  $\lambda$  is the wavelength of X-ray,  $r_e$  is the classical electron radius,  $\rho_{\text{eff}}$  is the effective electron density, and  $\beta$  is the imaginary part representing the absorption. The XRR is calculated by the interference of X-rays scattered by each layer.

Solid lines in Fig. 1a represent the best-fits of the experimental specular XRR curves obtained on the Fe film grown on GaAs(100) at 140 K. The major fitting parameters are  $\sigma_s$ ,  $\sigma_i$ , and the film density  $\rho_f^0$ . The model reproduces the experimental XRR curves rather well. The electron density profile of the substrate and the film obtained from the fit is plotted in Fig. 1b in the case of the 17 Å thick film. We note that XRR provides only the overall density profile, and one cannot separate the contributions of each chemical species. However, the best-fit values of the film electron density  $\rho_f^0$ , 2.37 electrons/Å<sup>3</sup> is close to that of bcc Fe, and we conclude that the film consists mostly of Fe atoms. We note that it was not necessary to include any independent interfacial alloy layers or an As-segregated layer to describe the data, although the interface roughness is increased to 4–5 Å. Considering that the roughness of the clean substrate was about 2.3 Å, the interfacial alloy formation must be limited to one or two monolayers. This suggests that no significant chemical reactions of the Fe and the Ga(As) have occurred. This result is consistent with the observation of the recent photoelectron spectroscopy that finds effective suppression of the outdiffusion of both Ga and As, and negligible interface alloying in an Fe film grown on GaAs(100) at 130 K [11].

In Fig. 2, summarized are the  $\sigma_s$  and  $\sigma_i$  as a function of the growth time. The error limit was estimated by drawing constant  $\chi^2$ , the goodness of the fit, contours in the fitting parameter space, and selecting a boundary that takes on a value greater by one than the minimum values. This way of estimating the error limit was necessary to take care of the correlation between the fitting parameters such as the film density and the roughness. The linear dependence of the  $\sigma_s$  and  $\sigma_i$  on  $t$  in a double logarithmic scale implies a power law growth,  $\sigma_{s,i} \sim t_{s,i}^{\beta}$ , where  $\beta_s$  and  $\beta_i$  are  $0.43 \pm 0.14$  and  $0.17 \pm 0.18$ , respectively. The fact that  $\beta_s$  is different from  $\beta_i$  indicates that the surface width does not result from simple propagation of the interface profile. The small value of  $\beta_i$  indicates that the interface width was practically unchanged during the growth, consistent with the Z-contrast electron microscopy result [12].

Such a high value of  $\beta_s$  close to 0.5 has been predicted previously: The random deposition model that allows no lateral relaxation [15] gives rise to  $\beta \approx 0.5$ . A more realistic model, where surface diffusion is allowed, predicts  $\beta \sim 0.5$  when there is a destabilizing uphill current induced by step

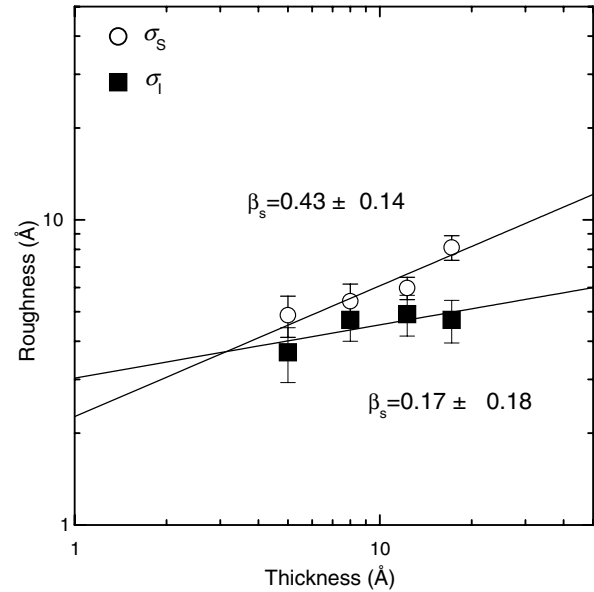


Fig. 2. Surface roughness,  $\sigma_s$  and interface roughness,  $\sigma_i$  as a function of the deposited Fe film thickness,  $t$  at 140 K. The solid lines show the results of fitting the  $\sigma_s$  and  $\sigma_i$  to a power law,  $\sigma \sim t^\beta$ .

Ehrlich-Schwöbel (ES) barrier [16]. Atomistic picture for the growth kinetics was also obtained by kinetic Monte Carlo (KMC) simulations [17,18] that explicitly took the step ES barrier into account; they found that  $\beta_s$  was maximal, 0.5 at temperatures where (1) the step crossing is virtually frustrated, and (2) the other smoothing process such as downward funneling is still not effective. In regards to the predictions of both the continuum models and the KMC simulations, the ES barrier for the present system seems to be insurmountably high at the deposition temperature, 140 K, and gives rise to the high uphill current of the adatoms. Hence, there develops very rough surface with  $\beta_s \sim 0.5$ .

We also examined the thermal stability of the Fe film at ambient temperature that is critical for the device applications. After depositing an Fe film of 23 Å at 140 K and annealing to 300 K, the structure was investigated by XRR. As shown in Fig. 3a, the overall intensity of the XRR was enhanced by the annealing, and the modulation of the intensity with respect to  $q_z$  was much improved as compared with the as-grown sample at 140 K, e.g. 17 Å-thick film in Fig. 1a. This indicates that the surface roughness was significantly reduced by the smoothing effects at 300 K. Upon further annealing for one more hour, the XRR curve did not show any further change as shown in Fig. 3a, which indicated that the film was already in a steady state.

The XRR of the annealed sample was well described by the previously adopted model, a single Fe layer on GaAs(100). The best-fit values of the film surface width  $\sigma_s$  was  $4.5 \pm 0.5$  Å, and the interface width  $\sigma_i$  was  $4.4 \pm 0.5$  Å. While the film surface was smoothed significantly during annealing, the interface width stayed mostly unchanged. This suggests that the ultrathin interfacial alloy

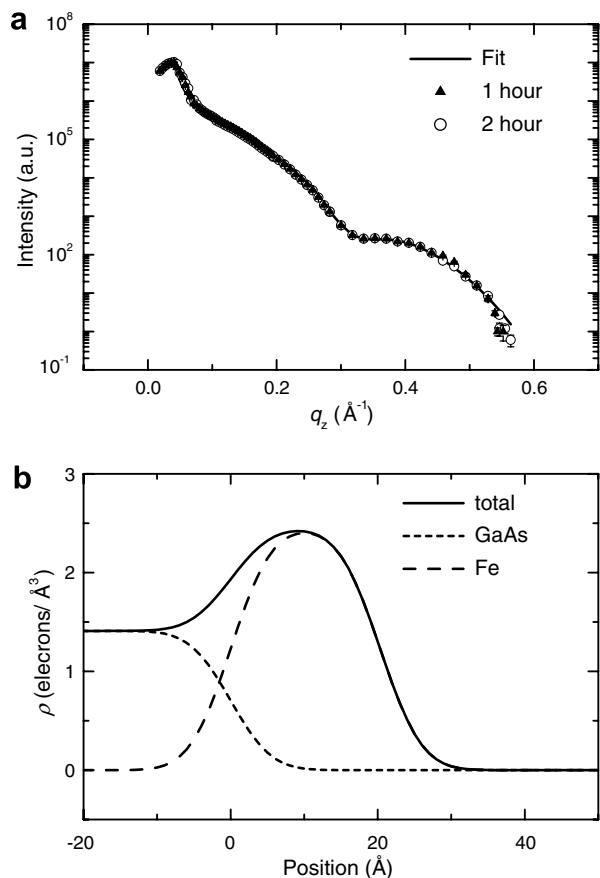


Fig. 3. (a) Specular X-ray reflectivity curves after annealing a 23 Å-thick Fe film at 300 K for one and two hours, respectively. (b) The best-fit, electron density profile of the annealed sample.

of one or two atomic layer formed during the growth prohibited further outdiffusion of Ga or As atoms effectively. The limited interface width is the consequence of the inefficiency of the bulk diffusion of the Ga and As from the substrate to the overlying film. Fig. 3b shows the electron density profile obtained from the best-fit values. We note that the maximum electron density of the Fe film is  $2.4 \text{ electrons}/\text{Å}^3$  which is slightly larger than the electron density of bcc Fe. This might be attributed to the small amount of Ga or As atoms that have out-diffused through the interface. Actually, recent photoelectron spectroscopy of the Fe film grown on GaAs(100) at 130 K observes the appearance of As peak, although very small, when the film is annealed above room temperature, and attributes it to the outdiffused As atoms far below the surface [11]. We conclude that the pristine Fe film is mostly conserved or kinetically stabilized at room temperature, although small amount of As atoms might have diffused through the interface.

## Summary

We have investigated the growth of the Fe films on GaAs(100) at 140 K, by *in situ* X-ray reflectivity. At 140 K, a rough Fe film forms on GaAs(100) due to restrictive surface relaxation. The chemical reaction at the interface and surface segregation of As seems to be virtually suppressed during the growth at 140 K. After annealing the sample to room temperature, the interface is maintained sharp, and the pristine Fe film is mostly preserved.

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