Anisotropic lattice relaxation and uniaxial magnetic anisotropy in Fe/InAs(100)-4×2

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(Received 14 April 2000)

The magnetic anisotropy and the lattice relaxation of epitaxial Fe films grown on InAs(100)-4×2 at room temperature have been studied using *in situ* magneto-optical Kerr effect and reflection high-energy electron diffraction. The experimental results demonstrate that the symmetry breaking associated with the intrinsic atomic scale structure of the reconstructed semiconductor surface induces an in-plane anisotropic lattice relaxation and an in-plane uniaxial magnetic anisotropy in the ultrathin region. We propose that this is a general phenomenon in ferromagnetic/semiconductor heterostructures.

One of the fundamental issues in heterostructure epitaxial growth is the effect of lattice mismatch on growth morphology and physical properties. The stress caused by the mismatch between the epitaxial overlayer and the substrate produces a driving force that modifies the structure and morphology, which in turn determine the physical properties of the heterostructures. Recent studies of the growth morphology[1–4] using high-resolution scanning electron microscopy and scanning tunneling microscopy have revealed that strain not only drives the initial two-dimensional–three-dimensional (2D-3D) transition, but can also drive the transition from one island shape to another. Another important issue in heterostructure epitaxy is the evolution of the lattice strain. In general, the epitaxial overlayer grows in registry with the substrate lattice mesh in the initial pseudomorphic phase, and then the strain is partially relaxed over a certain thickness range before reaching the bulk lattice constant. However, the effect of the surface morphological and electronic structure of the substrate on the lattice relaxation of the epitaxial overlayer has rarely been investigated. This should be particularly important for heterostructure epitaxy on a semiconductor substrate, as various atomic scale structures occur on the reconstructed semiconductor surface.[5–7] These atomic scale structures may lead to an ''anisotropic lattice relaxation,'' as compared with the ''anisotropic growth morphology'' observed recently in Ge/Si-based systems by several groups.[3,4,8]

Knowledge of the surface and interface structure at the atomic and nanometer scale is central to understanding the magnetic properties of ultrathin ferromagnetic films.[9] An oscillation of the magnetic anisotropy was observed in Co/Cu(100) (Ref. 10) due to the periodic variation of the film roughness alternating between filled and incompletely filled atomic layers. Symmetry breaking at atomic steps of the ultrathin Fe films grown on stepped Ag(100) was found to create a uniaxial magnetic anisotropy, for example.[11] Strain has been widely shown to contribute to the magnetic anisotropy in epitaxial heterostructures in several studies.[12–15] Ferromagnetic metal/semiconductor heterostructures have attracted great attention recently for the study of fundamental magnetic properties of ultrathin films and for the development of next generation magnetoelectronic devices.[16,17] An in-plane uniaxial magnetic anisotropy (UMA), unexpected from the crystal symmetry of bulk bcc Fe, was observed in the Fe/GaAs system,[18–23] but the origin of this uniaxial anisotropy remains an open issue. It is now generally believed that the atomic scale structure related to the reconstruction of the semiconductor surface is responsible for this uniaxial anisotropy. However, the precise role of the atomic scale structure of the substrate surface is unclear. Two distinctly different mechanisms associated with ''unidirectional chemical bonding'' and ''anisotropic lattice relaxation,'' respectively, can be considered.

In this paper, we report the results of *in situ* structural and magnetic studies of an epitaxial ferromagnetic metal/III-V semiconductor system, Fe/InAs(100). We have shown in our preliminary work[24] that epitaxial bcc Fe can be grown on InAs(100) at 175 °C and that Fe/InAs is a very promising system for the fabrication of magnetoelectronic devices due to the Ohmic contact of the Fe/InAs interface. As the lattice mismatch of 5.4% between Fe and InAs is reasonably large, the Fe/InAs system is a better system than Fe/GaAs in which to study the correlation of surface atomic scale structure, anisotropic lattice relaxation, and magnetic uniaxial anisotropy. Furthermore, a comparative study of the magnetic anisotropy in Fe/InAs and Fe/GaAs, as undertaken here, may help to reveal the role surface morphology and possible anisotropic magnetoelastic coupling since the lattice constant of Fe is larger than half the lattice constant of GaAs but smaller than half that of InAs, and equivalent surface reconstructions can be stabilized on the GaAs(100) and InAs(100) surfaces.

This study was carried out in a ''multitechnique'' molecular-beam epitaxy system, which includes *in situ* magneto-optical Kerr effect (MOKE) and Brillouin light scattering (BLS) to probe the static and dynamic magnetic properties of samples, and scanning tunneling microscopy (STM), low-energy electron diffraction (LEED), reflection high-energy electron diffraction (RHEED), and Auger spectroscopy to provide structural, morphological, and compositional information. The Fe films were grown on InAs(100) substrates at a rate of approximately 1 ML per minute using an *e*-beam evaporator. The pressure was around $7-8 \times 10^{-10}$ mbar during growth and the substrate was held at room temperature. The deposition rate was monitored by a quartz microbalance which was calibrated using RHEED oscillations of Fe on a Ag(100) single crystal. The InAs(100) substrates were cleaned using a combination of oxygen

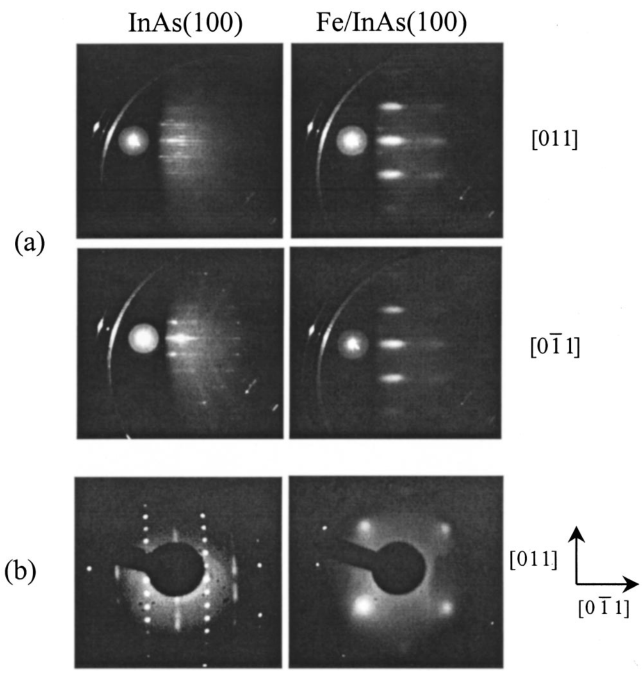

FIG. 1. (a) RHEED patterns of the InAs(100)-4×2 substrate, and after 30 ML of Fe, operated at 15 keV, and (b) LEED patterns of the InAs(100)-4×2 substrate, 135 eV, and after 50 ML of Fe, 120 eV.

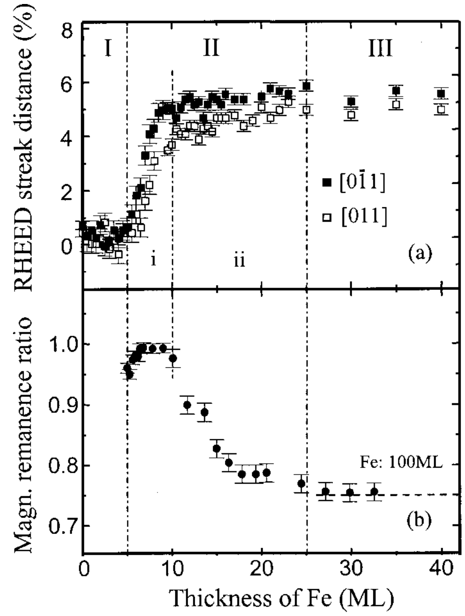

FIG. 2. (a) Relative changes of the RHEED strip distances compared with that of InAs(100) substrate and (b) magnetization remanence ratios measured along [011] direction as a function of Fe coverage.

plasma etching and wet etching (HC:H_2O=1:4) before loading into the UHV system and annealing in the chamber at 510 °C for 0.5 h before growth.

The RHEED and LEED pictures of the substrate after annealing are presented in Figs. 1(a), and 1(b) respectively, which show an In-terminated 4×2 reconstruction of the InAs(100) surface.[7] These clearly reconstructed diffraction patterns indicate that the InAs substrate surface has a high degree of crystallographic order with a long coherence length. Auger spectroscopy measurements show that the substrate is free of O, but has a tiny C peak, after the annealing. The epitaxial growth of the Fe films has been confirmed with both LEED and RHEED measurements. Typical RHEED and LEED patterns from the Fe films are shown in Fig. 1. These clear diffraction patterns demonstrate that single-crystal bcc Fe films have been stabilized on InAs(100)-4×2 at room growth temperature, despite the large lattice mismatch. The epitaxial relationship is Fe(100)⟨001⟩‖InAs(100)⟨001⟩, i.e., the same as that for the Fe/GaAs(100) system.[16,18,22]

The strain relaxation during epitaxial growth has been studied with dynamic RHEED measurements for [011] and [0$\bar{1}$1] directions. A detailed RHEED study along these two directions is crucial in understanding the magnetic properties, since as shown in Fig. 3, a uniaxial magnetic anisotropy develops in the ultrathin region with the easy axis along the [011] direction and the hard axis along the [0$\bar{1}$1] direction. The growth conditions and RHEED setup were kept unchanged throughout to enable direct comparison of the results obtained for the two different directions. Figure 2(a) shows the relative changes of the peak separations compared to that of the InAs(100) substrate as a function of Fe coverage. The growth could be divided into three stages as suggested by the figure. Region I (0–5 ML): in this pseudomorphic growth stage the films have the same lattice constant as that of the substrate and are highly strained. Region II (5–25 ML): this is a transition region between pseudomorphic growth and full relaxation. The films begin to relax after about 5 ML along both directions. However, the relaxation along the [0$\bar{1}$1] direction is *significantly* faster than that along the [011] direction. Region II could then roughly be divided into two subregions, (i) 5–10 ML and (ii) 15–25 ML. In subregion (i), the lattice constant along the [0$\bar{1}$1] direction changes rather sharply with increasing thickness and approaches the bulk value around 10 ML, while the lattice constant along the [011] direction changes much more slowly and levels off around 25 ML in region (ii), i.e., ''anisotropic lattice relaxation'' is clearly observed.

The evolution of the magnetic properties has been probed using *in situ* MOKE measurements. Figure 3 shows the magnetic hysteresis loops of Fe/InAs(100)-4×2 of different thicknesses with the magnetic field applied along four major axes. The thickness dependence of the remanence ratio of the hysteresis loops along [011] direction is included in Fig. 2(b). Figures 3 and 2(b) show that the UMA dominates in the ultrathin region of about 5–10 ML, which corresponds exactly to subregion II-i in Fig. 2(a). The easy axis of this uniaxial anisotropy is along the [011] direction, as shown clearly by the perfect square loops along this direction in Fig. 3. Beyond about 25 ML the magnetic hysteresis loops along four major axes were found to remain almost unchanged with increasing thickness. The films display a cubic anisotropy with the magnetic easy axes along the ⟨001⟩ directions, the easy axes of bulk bcc Fe. This is in good agreement with the thickness range of region III in Fig. 2(a), which shows that bulklike bcc Fe has been established above about 25 ML. The remanence ratio decreases above about 10 ML in the thickness range of 10–25 ML due to a competition be-

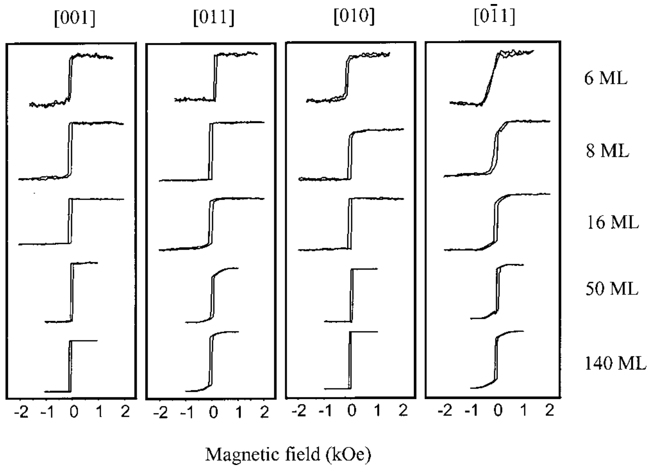

FIG. 3. *In situ* MOKE hysteresis loops of Fe/InAs(100)-4×2 of different thicknesses with the magnetic field applied along four major axes.

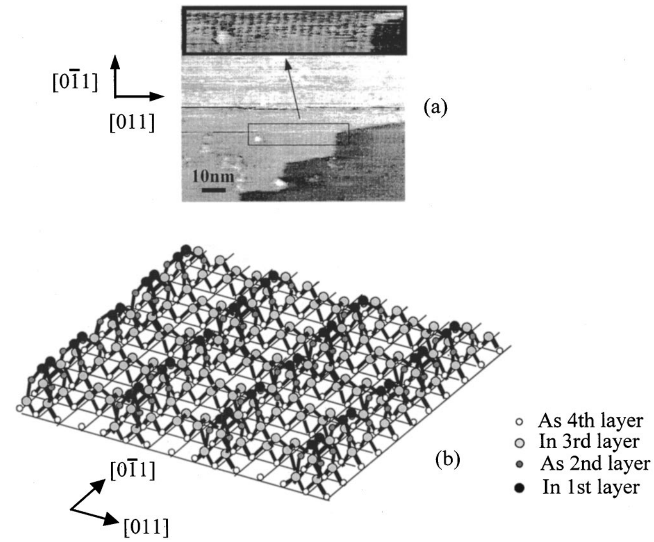

FIG. 4. (a) STM image of the InAs(100)-4×2 substrate, and inset, enlarged image showing clearly the In rows, and (b) a 3D schematic atomic model.

tween the cubic and the uniaxial anisotropies. This region in Fig. 2(b) agrees with the thickness range of the subregion II-ii in Fig. 2(a), where there are still significant differences of the lattice constants along the two directions.

By examining the anisotropy of the Fe films deposited on two different kinds of GaAs substrates showing different reconstructions, Kneeler *et al.*[21] proposed that the unidirectional nature of Fe-As or Fe-Ga bonds is responsible for the UMA. This might be understood as a ''chemical'' effect, in which the electronic structure of the Fe atoms near the interface differ distinctly from ''normal'' bcc Fe. Another picture is that uniaxial magnetoelastic coupling due to the anisotropic lattice relaxation is the origin of the uniaxial magnetic anisotropy. An anisotropic strain relaxation inferred from the island shape of the films was discussed for Co/Cu(110) by Fassbender *et al.*[25] Though the unidirectional ''chemical bond'' cannot be excluded, the present work provides two pieces of key experimental evidence to support the *uniaxial* magnetoelastic coupling picture. First, the easy axis direction of the UMA in Fe/InAs(100)-4×2 differs from that in Fe/GaAs(100)-4×2.[20] The Fe film in the ultrathin region is *compressed* (in-plane) on GaAs whilst *expanded* on InAs. This will lead to opposite strain tensor components for the two systems in accord with the observed anisotropy behavior. However, we would like to note that according to the magnetoelastic energies the sign of the magnetostriction constant in the ultrathin films is opposite to that of bulk Fe, which is in good agreement with a recent study of Fe/W.[26] Second, the uniaxial strain relaxation is observed to occur over the same thickness range in which the uniaxial magnetic anisotropy varies. This may be the first direct experimental evidence, as far as we know, to show that the anisotropic strain contributes to the in-plane uniaxial anisotropy.

We propose here a possible mechanism to explain this unique uniaxial lattice relaxation. The development of a theoretical mode to fit the experimental results remains a challenging issue and goes beyond the scope of this work. Figure 4(a) shows a STM image of an InAs(100)-4×2 surface. The 4× periodicity with a repeat distance of 17 Å along $[0\bar{1}1]$ can be seen clearly from the enlarged inset image. The height of the bright rows is about 3 Å, which agrees with the height of the corrugation between the first and the third In dimers. We would like to note that the twofold periodicity along $[0\bar{1}1]$ has not been resolved by the STM, although this is suggested from the LEED image shown in Fig. 1. Figure 4(b) shows a 3D schematic diagram of the atomic structure of the InAs(100)-4×2 surface, which reveals clearly the In dimer rows along the $[0\bar{1}1]$ direction. The In rows will present an additional energy barrier to the motion of the interfacial dislocations along the [011] direction. This ''anisotropic energy barrier'' will therefore lead to different thickness dependences of the lattice relaxation along two ⟨011⟩ directions. We thus conclude that the atomic scale structure of the reconstructed surface, which breaks the lattice symmetry of the surface atoms, is responsible for the observed uniaxial strain relaxation and uniaxial magnetic anisotropy.

In summary, we have studied the structure and magnetic properties of an epitaxial ferromagnetic metal/III-V semiconductor system, Fe/InAs(100)-4×2 stabilized at room temperature. We report direct observation of an ''anisotropic lattice relaxation,'' which is correlated with the intrinsic atomic scale structure of the reconstructed semiconductor surface. The thickness range of this uniaxial lattice relaxation was found to be in perfect agreement with that over which the uniaxial magnetic anisotropy evolves. The observation of the uniaxial strain relaxation therefore represents direct experimental evidence of the role of the atomic scale structure of the semiconductor surface and associated anisotropic magnetoelastic coupling in giving rise to uniaxial magnetic anisotropy in ferromagnetic metal/semiconductor heterostructures.

We gratefully acknowledge the financial support of the EPSRC, and the ESPRIT MASSDOTS and SUBMAGDEV (FMRX-CT97-0147) projects (EC).

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