# Interplay between Anisotropic Strain Relaxation and Uniaxial Interface Magnetic Anisotropy in Epitaxial Fe Films on (001) GaAs

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Grazing incidence x-ray diffraction study of Fe epitaxial ultrathin films (1.5–13 nm) on GaAs (001) reveals an anisotropy of both domain shape and strain, with [110] and [1-10] as the principal directions. It is shown that the observed thickness-dependent strain anisotropy, together with a uniaxial interface term, can provide an unambiguous explanation to the usual in-plane magnetic anisotropy and its thickness dependence observed in this magnetic thin-film system.

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Ferromagnetic thin films on single-crystal semiconductor substrates have been of great interest in recent years because of the possible future development of "spintronics," e.g., spin injection through the ferromagnetic thin-film semiconductor interfaces [1–4]. Among these ferromagnetic hybrid structures, single-crystal Fe on GaAs (001) is one of the most promising of such heterostructures, and a rather large number of studies have been devoted to this system in recent years [5,6,8–14]. Fe grows epitaxially on GaAs (001) with a cube-on-cube orientation [5,6] due to a relatively small mismatch $m = -1.4\%$ in lattice parameters between Fe ($a_{\mathrm{Fe}} = 0.28664$ nm) and GaAs ($a_{\mathrm{GaAs}} = 0.56537$ nm) provided one considers half the lattice constant of GaAs. The thin films of Fe/GaAs (001), with thickness less than about 60 ML or 9 nm, exhibit a remarkable and unexpected in-plane uniaxial magnetic anisotropy (UMA) with an [110] easy axis [7]. This is dramatically different from the cubic magnetic anisotropy of bulk bcc Fe with $\langle 100 \rangle$ easy axes. This result has been observed by several different research groups [7,9–13] in recent years but its origin remains to be one of the unanswered fundamental questions in ferromagnetic thin-film studies.

Magnetic anisotropy is determined by minimizing the free energy per unit volume with respect to the magnetization direction cosines [15,16]. Different contributions to the total energy have to be added: magnetocrystalline anisotropy, demagnetizing field energy (shape anisotropy), magnetoelastic (ME) coupling energy,.... In thin films additional surface and interface terms [15,16] or strain dependent corrections [17–19] are often included in order to take into account deviations from the bulk behavior. However, none of these terms provide quantitative and definitive results for the Fe/GaAs (001) system. It has also been known that the UMA does not seem to depend on any particular surface reconstruction which the Fe layer is grown on: As terminated $(2 \times 4)$ and $c$-$(4 \times 4)$ [9,12], or Ga terminated $(4 \times 6)$ [10–12,14].

Qualitative and quantitative explanations of the UMA in Fe/GaAs (001) systems require detailed knowledge of the structural properties of the film that include strain field and interfacial morphology. Here in this Letter we present a detailed synchrotron x-ray diffraction study of the Fe/GaAs (001) heterostructures with Fe thicknesses ranging between 1.5 and 13 nm, i.e., crossing the magnetic anisotropy transition. Our results clearly indicate that (a) considerable strain and shape anisotropies do exist in these Fe thin films, (b) the interfacial effect is the principal contributor to the observed UMA for thin Fe films, and (c) the strain anisotropy is the main factor responsible for the reversal of UMA at high Fe thickness.

The Fe/GaAs samples were prepared at room temperature by molecular beam epitaxy [12] on As-rich $(2 \times 4)$-reconstructed surfaces and capped with a 3 nm thick Al layer. The [110] and [1-10] directions are unambiguously identified from the reflection high-energy electron diffraction (RHEED) pattern of the reconstructed surface. Let us recall that the convention for III-V (001) surfaces is to label [1-10] as the direction of the element V dimers.

The in-plane magnetic anisotropy of the Fe layers was investigated by *ex situ* magnetooptic Kerr effect (MOKE) measurements in the longitudinal geometry in fields up to 0.11 T. $w_m$, a quantity proportional to the energy required to saturate the films, was determined by an integration of the anhysteretic loops of the reduced magnetization $m(H)$

  

($m = M/M_s$ where $M_s$ is the saturation magnetization). The integration of $m(H)$ loops was performed by averaging magnetization curves in ascending and descending branches between 0 and 1. The same calculation was performed for $m < 0$ between 0 and $-1$ and $w_m$ was obtained from an average between integrations in negative and positive half spaces.

The x-ray diffraction experiments were performed *ex situ* at CHESS F3 station using a 7.7 keV monochromatic beam using a double-crystal Si (111) monochromator. The Fe/GaAs (001) sample, typically 5 mm by 10 mm in size, was mounted at the center of a vertical four-circle diffractometer with a double-pass Si (111) analyzer and a NaI detector. For convenience and consistency with literature, we use the substrate GaAs reciprocal space indices to index the reflections. Two types of measurements were performed in our experiment: grazing incidence reciprocal space mapping around the 220 and 400 reflections for in-plane coherent domain (CD) size and strain information, and $22l$ and $40l$ scans along the surface normal $q_z$ for information about film thickness and perpendicular lattice parameters.

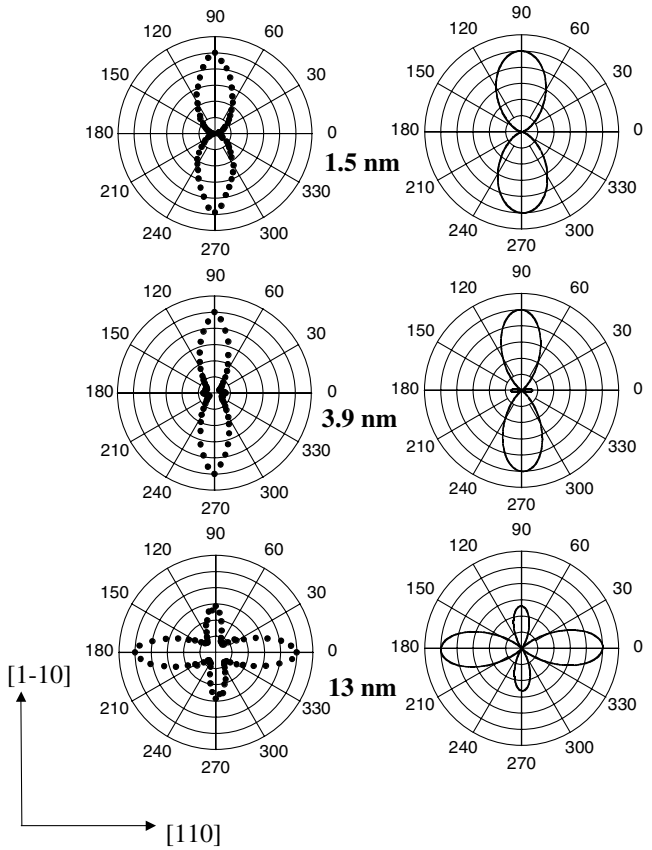


FIG. 1. Right: Experimental polar plots of $w_m$ for Fe thickness of 1.5 nm (top), 3.9 nm (middle), 13 nm (bottom). Left: Calculated magnetic energy density polar plots for the same thickness. The calculation includes the cubic anisotropy of bulk Fe, bulk ME coupling with measured strains, and an interface UMA. $K_u = 1 \times 10^{-4}$ J m$^{-2}$ is a fit parameter. The origin of in-plane azimuth is along [110]. The maximum in energy is always normalized to one.

Selected polar plots of $w_m$ are shown in Fig. 1. For the lowest Fe coverage a clear UMA is evidenced with [110] as the easy axis of magnetization. When increasing the Fe thickness the strength of this UMA decreases and around a thickness of 6 nm (not shown) a fourfold symmetry is observed. Beyond that thickness a reversal of the MA is observed with a tendency for a [1-10] easy axis (Fig. 1) superimposed on the cubic anisotropy.

Characteristic in-plane reciprocal space maps around the $-2$-20 and the $-220$ reflections are shown in Figs. 2 and 3 for Fe thin-film samples of thickness 1.5 and 13 nm, respectively. The thickness was determined from the finite size diffraction fringes in perpendicular $q_z$ scans. The measured reciprocal space maps show some striking structural difference. In Fig. 2 (1.5 nm sample) the reciprocal lattice nodes consist of a very sharp component superimposed on a diffuse streak elongated along the [110] direction. The diffuse component is at the same position as the sharp one, which indicates that Fe is perfectly pseudomorphous with the GaAs substrate. The simplest interpretation for this anisotropic diffraction peak shape is to relate it to an anisotropic domain size. The resulting CD dimensions are $L_{[110]} = 460$ nm and $L_{[1-10]} = 1032$ nm. In Fig. 3 (13 nm sample) the maximum of Fe diffraction peak has clearly moved away from the sharp GaAs one, which indicates that Fe has relaxed at that thickness. A closer look at the {220} maps and the radial $\theta/2\theta$ scans shows that strain relaxation is anisotropic, being greater along [110] than along [1-10]. From the width of Fe diffraction peak the following domain sizes are obtained: $L_{[110]} = 53$ nm and $L_{[1-10]} = 35$ nm. The anisotropy in CD size is thus *reversed* with respect to the 1.5 nm sample. A summary of these results is given in Fig. 4 for all the investigated thickness. The in-plane CD size is anisotropic (Fig. 4): below 3 nm domains are elongated along [1-10], whereas above 3 nm they are elongated along [110]. Below 2 nm Fe is pseudomorphous on GaAs (Fig. 4). Above 2 nm anisotropic relaxation takes place: Fe is more relaxed along [110] than along [1-10].

Several scanning tunneling microscopy studies [9,13,20,21] have reported Fe islands elongated along [1-10], in agreement with what we deduce from the diffraction peak widths for the thinnest sample. The reason for this anisotropic shape may be related to the Fe preferential bonding to As atoms. This could result in anisotropic surface diffusion coefficients and thus different growth rates along these directions. Anisotropy in the bonding of Fe atoms at the interface has been suggested earlier [9]. The recent observation by x-ray absorption [22] of a common type of Fe-As local bonding in different Fe/GaAs interfaces support such a hypothesis.

To our knowledge anisotropic strain relaxation has never been reported before for Fe grown on (001) GaAs.

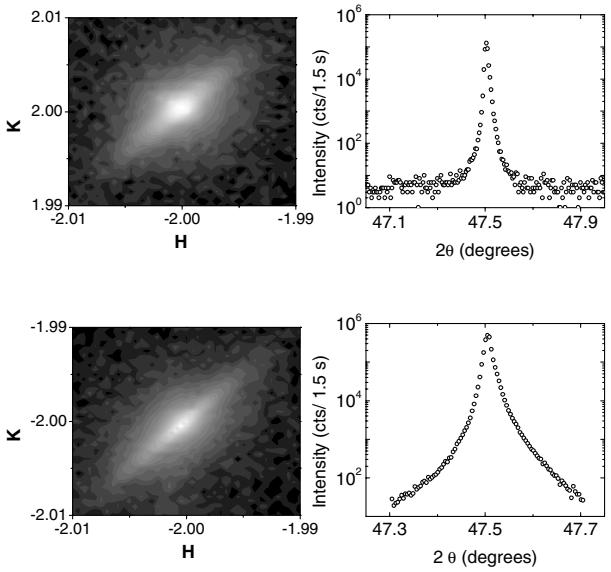


FIG. 2. Left: Reciprocal space maps around the $-220$ and $-2$-20 of GaAs (001). The intensity scale is logarithmic. $H$ and $K$ are the scattering vector components in units of $2\pi/a_{\mathrm{GaAs}}$. Right: Corresponding $\theta/2\theta$ radial scans. Fe deposited thickness is 1.5 nm.

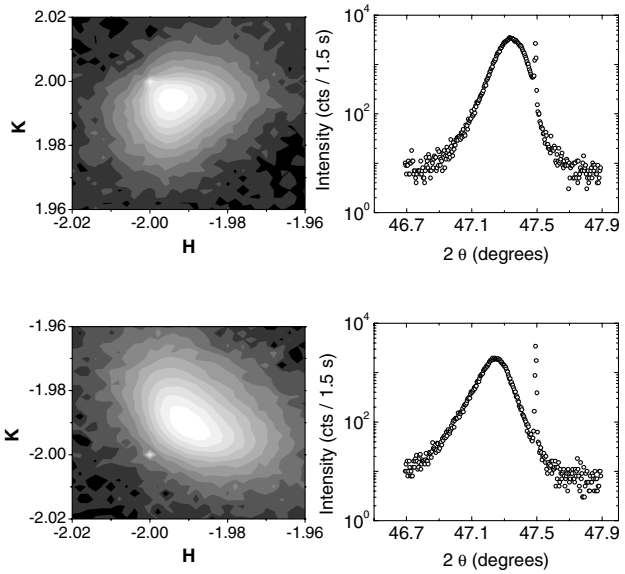


FIG. 3. Left: Reciprocal space maps around the $-220$ and $-2$-20 of GaAs (001). The intensity scale is logarithmic. $H$ and $K$ are the scattering vector components in units of $2\pi/a_{\mathrm{GaAs}}$. Right: Corresponding $\theta/2\theta$ radial scans. The broad peak is Fe {110} and the sharp one is GaAs {220}. Fe deposited thickness is 13 nm.

A similar anisotropic relaxation has, however, been reported [23] for Fe grown on (001) InAs. From RHEED it was observed that strain relaxation is larger along the [110] than along the [1-10] direction. We find the same situation in Fe on GaAs (001) with a larger relaxation along [110] as compared to [1-10] (Fig. 4). Fe thin films grown on InAs (001) are under tension (misfit $= +5.4\%$) and exhibit UMA [23] (easy axis along [1-10]). Anisotropic strain relaxation has been reported in InP quantum dots [24] and correlated with the observed anisotropy in the island shape. Indeed, elastic relaxation from the free edges is more efficient along the shortest dimension. Here the anisotropy in strain from Fig. 4 would correspond to islands elongated along [1-10], which is only observed below about 3 nm (Fig. 4). In this thickness range films do not exhibit any detectable (i.e., less than $10^{-4}$) elastic relaxation because of the very large island dimensions (Fig. 4). What may happen, however, is that plastic strain relaxation mediated by misfit dislocations occurs above 2 nm. In such an interpretation the anisotropy in relaxation could be interpreted as a consequence of an initial difference in the driving force for dislocation motion along the two principal directions.

It is worth noting that the investigated Fe films do exhibit structural anisotropies in CD size and strain relaxation with [110] and [1-10] being the principal axes. As shown in Fig. 4 these anisotropies are the most important for the thickest (13 nm) film investigated. For the thinnest films, the in-plane strain anisotropy vanishes as the films become completely pseudomorphic. UMA due to strain anisotropy mediated by ME coupling can thus be clearly ruled out as a cause for the observed UMA in the thinner samples. The strain anisotropy that develops in thicker films should be, however, taken into account for a quantitative modeling of magnetic anisotropy as a function of the Fe thickness. The resultant shear strain induces a ME coupling term which favors [1-10] as an easy direction. Introducing a uniaxial interface anisotropy term which favors alignment of the magnetic moment along [110] the magnetic free energy density writes

$$f_m = \frac{K_1}{4}\sin^2(2\phi) + \frac{K_u}{h}\sin^2\left(\phi - \frac{\pi}{4}\right) + \frac{B_2\varepsilon_6}{2}\sin(2\phi), \tag{1}$$

where $\phi$ is the angle between the magnetization and [100]. $K_1$ is the cubic anisotropy energy of Fe: $K_1 = 48\ \mathrm{kJ\,m^{-3}}$ and $B_2$ is the second ME coupling coefficient of Fe: $B_2 = 7620\ \mathrm{kJ\,m^{-3}}$ [25]. $\varepsilon_6$ is the shear strain in the crystallographic reference frame as calculated from the in-plane diffraction measurements. The $B_1$ term has been omitted since it does not depend on the angle $\phi$. $K_u$ is the interface uniaxial magnetic anisotropy and $h$ is the film thickness. A series of $f_m$ polar plots calculated according to (1) is shown in Fig. 1 for different film thickness and for $K_u = 1 \times 10^{-4}\ \mathrm{J\,m^{-2}}$. For the thinnest films there are no shear strains and magnetic anisotropy is driven by the interface UMA. As the film thickens anisotropy is the result of an interplay between interface UMA which favors an easy [110] direction and ME coupling which favors an easy [1-10] direction. For larger thickness it is the ME terms that

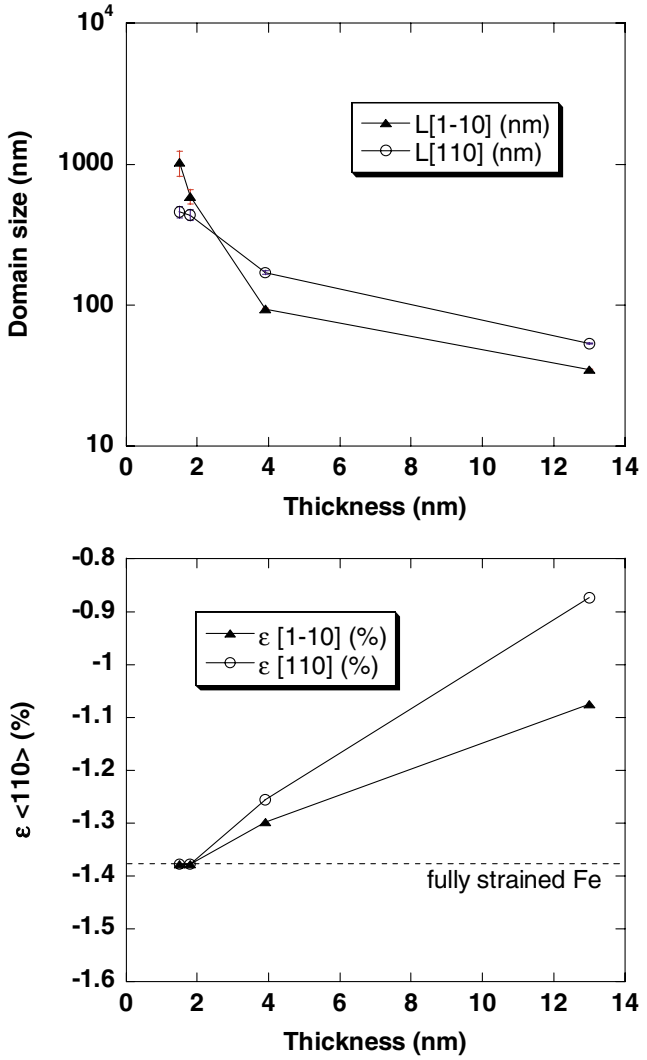


FIG. 4 (color online). Top: In-plane domain size along [110] and [1-10] as a function of Fe thickness. Bottom: In-plane strain along [110] and [1-10] as a function of Fe thickness.

takes over. The comparison with experimental MOKE measurements (Fig. 1) shows a very good agreement, taking into account that there is only one free parameter: $K_u$. The value of $K_u$ that gives the best agreement ($K_u = 1 \times 10^{-4}$ J m$^{-2}$) is remarkably close to the one reported in [8]. In our analysis no shape anisotropy has been included. The small anisotropy in domain size observed in the thinnest films cannot explain the UMA either qualitatively (the easy axis should be along the long axis, i.e., along [1-10]) nor quantitatively. One notes that the calculated magnetic energy density relies on the bulk value of $B_2$, which may be altered [17–19] by strain. In the case of Fe [25] measurements performed on films under tension indicate a strong strain dependence. We are not aware of any such measurements on Fe films under compression.

In conclusion, our x-ray diffraction study of a series of epitaxial Fe films on GaAs (001) clearly rules out ME coupling as well as shape anisotropy as the origin of the observed UMA in the thinnest films. Our investigation leads to a conclusion that the UMA in Fe/GaAs (001) is caused by an interface anisotropy. On the other hand, the evolution of the magnetic anisotropy as a function of Fe deposited thickness is very well described as a result of competition between ME coupling and interface UMA.

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[1] S. Datta and B. Das, Appl. Phys. Lett. **56**, 665 (1990).
[2] R. Sato and K. Mizushima, Appl. Phys. Lett. **79**, 1157 (2001).
[3] H. J. Zhu *et al.*, Phys. Rev. Lett. **87**, 016601 (2001).
[4] A. T. Hanbicki *et al.*, Appl. Phys. Lett. **80**, 1240 (2002).
[5] J. Waldrop and R. Grant, Appl. Phys. Lett. **34**, 630 (1979).
[6] J. Krebs, B. Jonker, and G. Prinz, J. Appl. Phys. **61** , 2596 (1987).
[7] There is some confusion in the literature on the exact orientation of the easy axis, i.e., [110] or [1-10]. After careful determination [110] appears, however, to be the easy axis in all cases. See, e.g., [8] where earlier results from the same group were reevaluated.
[8] M. Brockmann *et al.*, J. Magn. Magn. Mater. **198–199**, 384 (1999).
[9] E. Kneedler *et al.*, Phys. Rev. B **56**, 8163 (1997).
[10] M. Zolfl *et al.*, J. Magn. Magn. Mater. **175**, 16 (1997).
[11] M. Gester *et al.*, J. Appl. Phys. **80**, 347 (1996).
[12] B. Lépine *et al.*, J. Cryst. Growth **201**, 702–706 (1999).
[13] Y. Chye *et al.*, Appl. Phys. Lett. **80**, 449 (2002).
[14] R. Moosbühler *et al.*, J. Appl. Phys. **91**, 8757 (2002).
[15] *Ultrathin Magnetic Structures*, edited by B. Heinrich and J. Bland (Springer, Berlin, 1994), Vol. I, Chap. 2.
[16] M. T. Johnson *et al.*, Rep. Prog. Phys. **59**, 1409 (1996).
[17] D. Sander *et al.*, J. Phys. D **31**, 663 (1998).
[18] K. Ha and R. O'Handley, J. Appl. Phys. **85**, 5282 (1999).
[19] R. Koch *et al.*, J. Magn. Magn. Mater. **159**, L11 (1996).
[20] E. Kneedler *et al.*, J. Vac. Sci. Technol. B **14**, 3193 (1996).
[21] P. Thibado *et al.*, Phys. Rev. B **53**, R10481 (1996).
[22] J. Freeland *et al.*, Phys. Rev. B **63**, 193301 (2001).
[23] Y. B. Xu *et al.*, Phys. Rev. B **62**, 1167 (2000). A careful examination of LEED patterns indicates that the orientation given in this article should be rotated by 90°. This is what we have done in the text.
[24] M. Schmidbauer *et al.*, Phys. Rev. B **65**, 125320 (2002).
[25] G. Wedler *et al.*, Phys. Rev. B **60**, R11 313 (1999).