Reversal of uniaxial magnetic anisotropy in Fe/GaAs (110) films driven by surface relaxation: An *in situ* ferromagnetic resonance study

Cite as: AIP Advances 10, 075219 (2020); doi: 10.1063/5.0004261
Submitted: 2 April 2020 • Accepted: 2 July 2020 •
Published Online: 24 July 2020

Babli Bhagat,[1,a)] **Anna Semisalova,**[1] **Ralf Meckenstock,**[1] **and Michael Farle**[1,2]

AFFILIATIONS

[1] Center for Nanointegration and Faculty of Physics, University of Duisburg-Esssen, 47057 Duisburg, Germany
[2] Kirensky Institute of Physics, Federal Research Center KSC SB RAS, 660036 Krasnoyarsk, Russia

[a)] **Author to whom correspondence should be addressed:** babli.bhagat@uni-due.de

ABSTRACT

We report an *in situ* study of the time evolution of magnetic anisotropy constants of an uncapped 4 nm [~27 monolayers (ML)] Fe film epitaxially grown on a GaAs (110) substrate at room temperature under ultra-high vacuum (UHV) conditions. The structural and chemical properties are monitored by low energy electron diffraction and Auger spectroscopy with a sensitivity of 0.01 ML. The *in situ* UHV ferromagnetic resonance (FMR) study over a period of 6 days in $<10^{-9}$ Pa reveals that there is a slow magneto-morphological transition of the Fe film surface at room temperature. The resonance field measured *in situ* in the $[1\bar{1}0]$ direction initially changes at a rate of 0.3 mT/h within 30 h after deposition and later at 0.1 mT/h over 80 h. We determine the time-dependent changes in the in-plane and out-of-plane anisotropy constants and find a sign change in the uniaxial in-plane anisotropy in the first 24 h due to morphological changes at the surface. The *in situ* FMR measurements and the Auger analysis allow us to exclude changes in the magnetization and anisotropy due to the contamination and oxidation of the Fe film.

Fe/GaAs heterostructures have been extensively studied over the last three decades (for example, Ref. 1 and references therein). High Curie temperature, spin polarization, and the formation of a Schottky barrier at the interface[2] make it a promising system for spin injection devices. A small lattice mismatch of 1.6%[3] allows a highly epitaxial growth of bcc Fe on GaAs, providing a hybrid ferromagnetic (FM)/semiconductor (SC) junction system. Tetragonal distortion (c/a >1) is observed due to the compressive strain of Fe grown on GaAs (100) and (110).[4] This strain, due to the lattice mismatch, can be effectively reduced by growing Fe at an elevated temperature (250 °C), but with an intermixing of Fe and GaAs at the interface.[5] Several theoretical[6,7] and experimental[8–10] studies have been carried out for characterizing the structural (morphology and crystal structure) and magnetic (magnetization and magnetic anisotropy) properties of the Fe film on GaAs (100) and (110) substrates. A well-known property of the Fe/GaAs (100) system is the superimposed uniaxial anisotropy on a fourfold cubic anisotropy in the ultrathin limit.[11,12] The easy axis of the Fe film changes to the [110] direction from the [100] direction due to a dominating uniaxial anisotropy at a film thickness below 2 nm–3 nm in Fe/GaAs (100).[13,14] A similar reorientation of the easy axis from [001] to $[\bar{1}10]$ at a thickness below 24 ML Fe was reported[15] previously for Fe/GaAs (110).

GaAs (100) exhibits a range of surface reconstructions (e.g., 4 × 6, 2 × 4, 2 × 6) depending on the preparation and growth conditions[16] responsible for the uniaxial anisotropy in the deposited Fe film, whereas GaAs (110) with either Ga or As termination does not show any surface reconstruction.[17] Moosbühler *et al.*[18] claimed that Fe (001) grown on a Ga-rich GaAs (001) substrate with either (4 × 2) or (2 × 6) surface reconstruction has no effect on the resulting uniaxial magnetic anisotropy in the thickness range of 4 ML–14 ML. The surface termination of the substrate (either Ga or As) and reconstruction, however, usually affects the uniaxial magnetic anisotropy in Fe (001) films. Gradmann[19] determined the in-plane surface anisotropy at the critical thickness, where the spontaneous magnetization switches from [001] to $[1\bar{1}0]$ in Fe (110) films by assuming homogeneous magnetization along the surface normal.

Notably, very few time-dependent studies on Fe films have been reported.[20,21] Excellent time stability of capped 200 nm thick Fe films grown on GaAs (001) was shown by Weissman *et al.*,[21] whereas Gillingham *et al.*[20] reported smoothening of uncapped Fe islands grown on GaAs (100) over a period of 30 h. On the contrary, Godde[22] did not find a change in the surface morphology of either (001) or (110) uncapped Fe films grown at room temperature over 40 h after deposition.

The structural and magnetic stability of the *uncapped* Fe film over time, however, remains an open question. Previous studies were mainly focused on the structure and surface morphology of ultrathin Fe films with time and temperature. How this affects the magnetic properties of the uncapped Fe films over the time scale of several hours or days is still an open question for Fe grown on GaAs (110).

In this paper, we present the magnetic relaxation study of 4 nm Fe film grown on GaAs (110) using *in situ* ferromagnetic resonance (FMR) over the time scale of more than 100 h. Films were grown and characterized at room temperature under ultra-high vacuum (UHV) conditions. In the time-dependent study of magnetic properties, we find evidence that the initial morphology of the film is metastable. The film relaxes over time, which causes characteristic changes in the magnetic anisotropy energy (MAE).

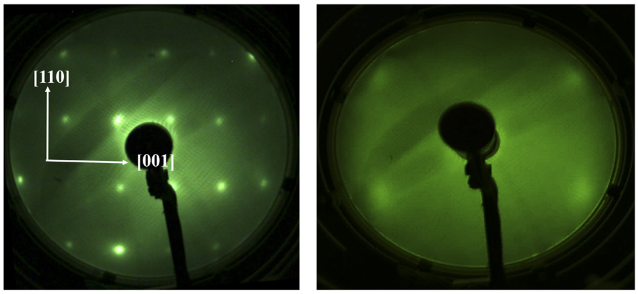
The synthesis and structural, chemical, and magnetic characterizations were carried out *in situ* in a UHV chamber (base pressure $\leq 5 \times 10^{-9}$ Pa). Thin films were grown by molecular beam epitaxy (MBE) using an electron beam evaporator. The films were structurally characterized by low energy electron diffraction (LEED) and Auger electron spectroscopy (AES). Subsequent magnetic characterization was performed using *in situ* ferromagnetic resonance. The setup has been described elsewhere.[23]

A commercial 4×4 mm^2 and 0.5 mm thick GaAs (110) wafer was used in these experiments. The substrate was pre-cleaned with ethanol in an ultrasonic bath before transferring to the UHV chamber. Subsequently, the GaAs surface was ion etched with argon at a partial pressure of 1×10^{-4} Pa for 1 h along with heating up to 960 K. Afterward, it was annealed at 960 K for 30 min at $\leq 8 \times 10^{-7}$ Pa.

A 4 nm Fe film named "Film I" was deposited at room temperature at a rate of 0.1 nm/min ±10%. The pressure during evaporation was maintained at $\leq 7 \times 10^{-8}$ Pa. The thickness of the film was monitored with a quartz thickness monitor. The surface structure and the chemical composition of the substrate and the film were measured by AES (3 keV) and LEED (50 eV–150 eV, 0.1 μA–1.6 μA). The FMR measurements were carried out using a microwave probe with an external magnetic field applied perpendicular to the microwave magnetic field component[23] and parallel to the Fe film plane.

The time-dependent FMR spectra were studied at room temperature under an ultra-high vacuum of $\leq 1 \times 10^{-8}$ Pa. After the time-dependent *in situ* studies, the surface morphology was analyzed by *ex situ* atomic force microscopy (AFM). This meant that these uncapped films were surface oxidized *ex situ*.

Figures 1(a) and 1(b) show the LEED patterns for the clean GaAs (110) substrate and the Fe film (4 nm) at 117 eV and 186 eV, respectively. The diffraction pattern of the Ga-rich (110) GaAs surface was observed with no surface reconstruction. AES spectra confirm an oxygen- and carbon-free GaAs substrate after sputtering and annealing. The LEED pattern of the Fe film, as shown in Fig. 1(b), indicates the epitaxial growth of bcc Fe on GaAs. The ratio between the diffraction spots along [001] and $[1\bar{1}0]$ directions is $\sqrt{2}$, typical for bcc Fe (110). AES measurements show no peaks of Ga and As (see the supplementary material, S1), indicating no outdiffusion to the surface of the Fe film. Auger peaks of Fe at 47 eV and 703 eV are visible in the AES spectra (see the supplementary material, S2).

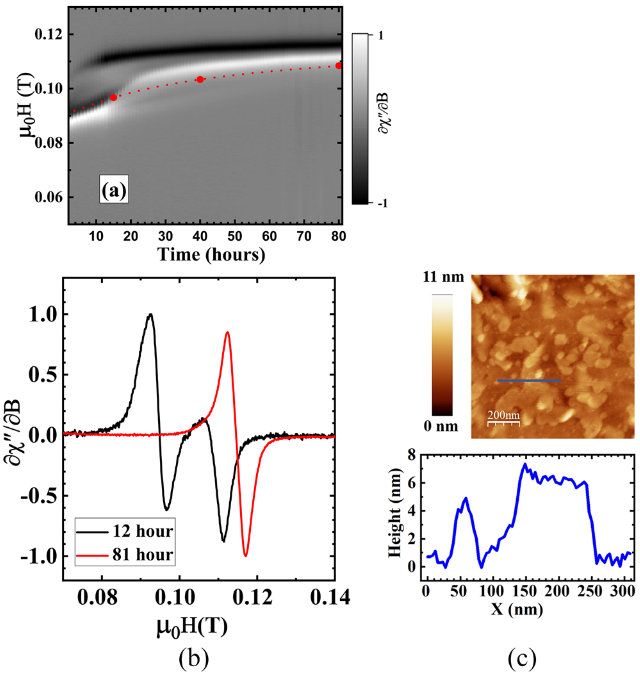


(a) (b)

FIG. 1. LEED images of (a) sputter-cleaned GaAs (110) substrate at 117 eV and (b) 4 nm Fe film on GaAs (110) at 186 eV.

The Auger peak of Fe at 47 eV, measured after 6 days, shows no change in shape (see the supplementary material, S2), confirming no oxidation on the Fe film surface. One should note that changes in the Fe Auger peak were observed by Kebe[24] and Smentkowski[25] even for the sub-monolayer oxygen adsorption. The *ex situ* AFM [Fig. 2(c)] scan of the oxidized Fe "Film I" in air shows a rough surface morphology with terraces of height 4 nm–6 nm. Figure 2(a) shows the result of *in situ* FMR measurements at 12.93 GHz of the 4 nm Fe "Film I" with the magnetic field applied parallel to the $[1\bar{1}0]$ direction in the film plane. The gray scale reflects the normalized amplitude of the microwave absorption derivative, which is proportional to $\partial\chi''/\partial B$.

The FMR spectra were measured every hour at room temperature under UHV conditions. The initial FMR signal after 2 h shows a single resonance line at 0.09 T. A second resonance line appears at a higher magnetic field after 4 h [Fig. 2(a)], which is more clearly seen in the "12-h" spectrum in Fig. 2(b). The two resonances are due to the change from a rough surface with larger areas of thinner Fe to a smoother surface with a quasi-uniform thickness of 4 nm. Directly after preparation, the anisotropy field is given by the roughness of the island-like morphology only. After 4 h and beyond, the final magnetic surface state forms with the narrower resonance at higher fields as expected for an almost flat film. During the time-dependent evolution, both resonances can be observed because both morphological conditions coexist across the few mm^2 large area of the film. As the time goes on, the intensity of the second line increases. The strong shift of the lower resonance field (0.3 mT/h–0.4 mT/h) in the first 24 h indicates a rapid morphological change of the Fe film. In this time period (10 h–35 h), these two areas with different roughnesses are large enough to act as separate magnetic regions that are only weakly coupled to each other. This is also the reason why the upper line is asymmetric. After 30 h, the rate of the resonance field



(b) (c)

FIG. 2. (a) Time-dependent FMR of 4 nm Fe/GaAs (110), named "Film I" 2 h after deposition with the magnetic field applied parallel to the $[\underline{1}10]$ direction. Red dots show the resonance field of another 4 nm Fe/GaAs (110), named "film II" at different time intervals. (b) FMR spectra of "Film I" at 12 h and 81 h after deposition. (c) AFM scan of "Film I" ($1 \times 1\ \mu\mathrm{m}^2$ area) and the line profile of the blue line in the AFM scan.

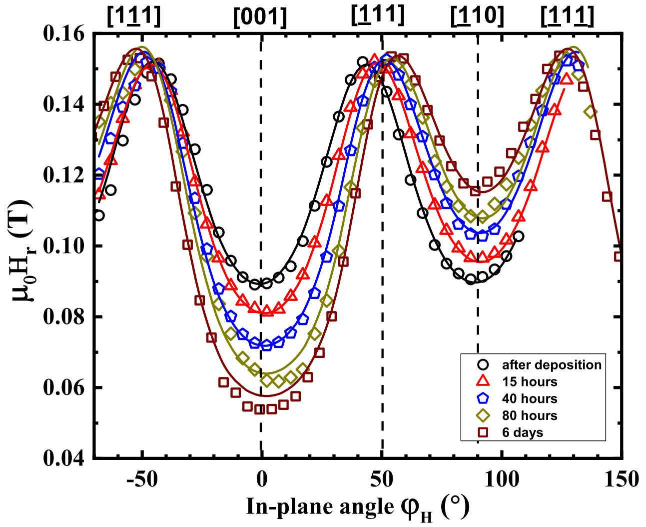


FIG. 3. Angular dependence of the resonance field of 4 nm Fe "film II" on GaAs (110) at different times. Open symbols show the experimental data, and the solid lines are the fit according to Eq. (1). The direction $[\underline{1}10]$ corresponds to that of Fig. 2.

shift decreases to 0.1 mT/h, and the two lines merge as seen by the red line [Fig. 2(b)]. The formation of larger terraces [Fig. 2(c)] is also evidenced by a decrease in the linewidth over time. The apparent change in the resonance field is caused mainly by the change in the anisotropy constants. To confirm this, we performed angular-dependent FMR measurements at different times on another 4 nm Fe film on GaAs (110) grown with the same deposition conditions. This film is called "film II", henceforth.

The *in situ* angular-dependent resonance field ($\mu_0 H_r$) of the lower resonance of 4 nm Fe "film II" taken at 13.054 GHz is shown in Fig. 3. To compare the two samples, resonance fields at different time intervals obtained from Fig. 3 in the $[\underline{1}10]$ direction (marked with red line) are plotted as red dots in Fig. 2(a), matching well qualitatively and quantitatively. The angular dependence of the resonance field 30 min after deposition (Fig. 3, black circles) depicts a pure fourfold symmetry that actually is not expected for this orientation.[26] This indicates the existence of an additional uniaxial anisotropy $K_{2\|}$, which changes over time. The expected [110] crystal symmetry behavior of Fe/GaAs (110) is observed 40 h after deposition.

In order to determine the crystalline cubic anisotropy K_4, uniaxial in-plane anisotropy $K_{2\|}$, and uniaxial out-of-plane anisotropy $K_{2\perp}$ constants, we analyzed the angular dependence of the resonance field according to the free energy density "F" for Fe (110),

$$\begin{aligned} F = {} & \frac{1}{4}K_{4\|}\Big(\cos^4\theta + \sin^2 2\theta\Big(\cos^2\varphi - \frac{1}{2}\sin^2\varphi\Big) \\ & + \sin^4\theta\big(\sin^4\varphi + \sin^2 2\varphi\big)\Big) + \Big(\frac{1}{2}\mu_0 M^2 - K_{2\perp}\Big)\cos^2\theta \\ & + K_{2\|}\sin^2\theta\cos^2(\varphi - \delta) - MB(\cos\theta\cos\theta_B \\ & + \cos(\varphi - \varphi_B)\sin\theta\sin\theta_B), \end{aligned} \tag{1}$$

where M is magnetization, B is the external magnetic field, φ is the in-plane angle of M, φ_B is the in-plane angle of external field B, θ and θ_B are out-of-plane angles of M and B, respectively, and δ is the angle between cubic and uniaxial in-plane directions. The fitting of the resonance field using the Smit–Beljers approach[27,28] to Eq. (1) yields anisotropy constants. All the fits were performed, assuming magnetization $M = 1600$ kA/m and g-factor g = 2.09 (bulk Fe). We obtain $\delta = (90 \pm 3)°$ for all the fits.

The anisotropy parameters obtained from the angular-dependent resonance field at different time periods are shown in Fig. 4. The in-plane cubic anisotropy $K_{4\|}$ is found to be $(3.6 \pm 0.1) \times 10^4$ J/m^3 close to[31] that of 30 ML thin Fe film. This is lower than the cubic anisotropy constant for bulk Fe (4.7×10^4 J/m^3).[32] We find that the cubic anisotropy (K_4) does not change significantly over time. The out-of-plane uniaxial anisotropy ($K_{2\perp}$) decreases by 10% after 6 days. Strikingly, there is a dramatic change in the uniaxial anisotropy ($K_{2\|}$) over time, along with a sign change after 30 h. Such a unique time-dependent change in the in-plane uniaxial anisotropy is very unusual for a Fe film at a constant thickness and temperature. The change in anisotropy constants can be attributed to the process of the surface relaxation, i.e., morphological changes of the Fe film at room temperature, while the crystal structure is maintained and no intermixing at the surface or interface occurs as confirmed by LEED and AES. Similarly, we assume constant M value since there is no oxidation of the Fe film, which would result in the reduction in M.[24]

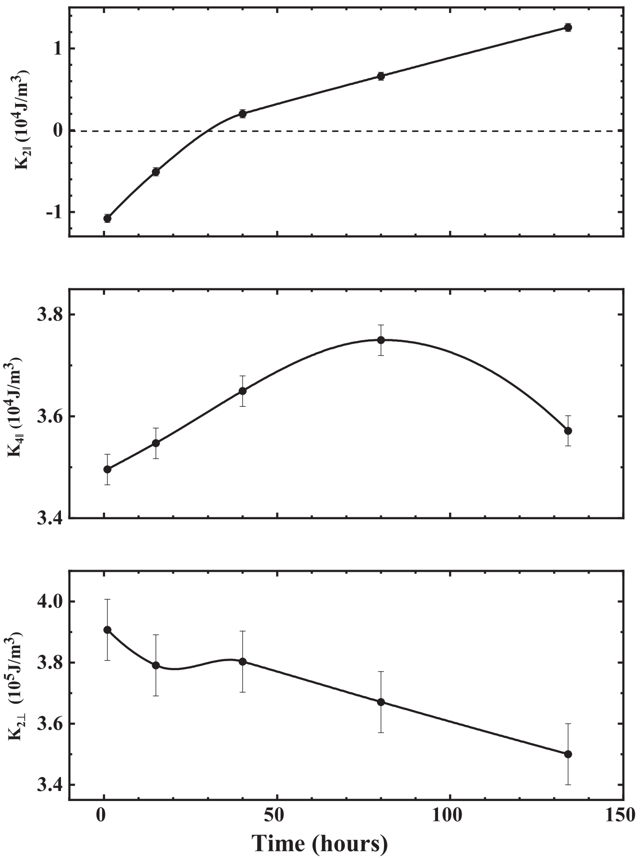


FIG. 4. Anisotropy constants of 4 nm Fe "film II" on GaAs (110) as a function of time after the deposition. In-plane uniaxial anisotropy constant $K_{2\|}$ shows a sign reversal. The error bar for all $K_{2\|}$ value is smaller than the symbol size ($\pm 0.05 \times 10^4$ J/m^3). Cubic anisotropy constant $K_{4\|}$ does not change significantly, whereas the out-of-plane uniaxial anisotropy constant $K_{2\perp}$ decreases by 10%.

Our FMR, LEED, and AES data analysis under UHV conditions reveal the magnetic relaxation of Fe films over time. We assume that the smoothening of the film surface takes place and the formation of the flat terraces within the initially rough film similar to that mentioned in Ref. 20 causes the change in the in-plane surface anisotropy[19] and continous increase in the in-plane uniaxial anisotropy with a sign change.[15,19] The free energy density behavior in Fe (110)[19] matches qualitatively with our resonance line fit (Fig. 3, solid lines). The chosen thickness 4 nm is in the critical thickness range, where the spin reorientation of the easy axis occurs[15] resulting in the sign change of the uniaxial anisotropy.[14] The out-of-plane anisotropy $K_{2\perp}$ is mainly due to the morphology changes at the Fe/vacuum interface since layers at the Fe/GaAs interface and in the volume are not expected to relax over time. The relaxation time here is several days since the magnetic system is kept in UHV at room temperature, which results in a slow morphological relaxation of the film. This slow relaxation at room temperature cannot be compared to the effects of annealing of the sample. Annealing not only results in faster relaxation but also introduces various surface and interface (intermixing) effects and change in the crystallinity.[29,30] Moreover, LEED and AES results show no evidence for oxidation or intermixing of the Fe film with Ga or As from the substrate even after 6 days in UHV (see the supplementary material, S1 and S2). Thus, the magnetism change is solely the result of the interplay of anisotropy contributions acting on the film surface. The significant change in the uniaxial in-plane and out-of-plane anisotropies is due to changes in the surface morphology, i.e., surface relaxation of the film leading to the reorientation of the in-plane easy axis. The surface relaxation is confirmed by the AFM scan of "Film I" taken after more than 3 days in air [Fig. 2(c)], which clearly shows the formation of flat terraces in contrast to the known roughness just after film deposition on GaAs (110). The change in roughness changes the effective surface and shape anisotropy contributions resulting in the change in the overall anisotropy that is seen in the shift of the resonance field.

In summary, we studied the time dependence of magnetic properties of a 4 nm Fe film on GaAs (110) using *in situ* FMR. The uncapped Fe (110) film is magnetically unstable over a period of more than 100 h. The interface/surface anisotropy rapidly changes due to morphological relaxation within 24 h after deposition leading to the unusual sign reversal of the in-plane uniaxial anisotropy.

See the supplementary material for the Auger spectroscopy of 4 nm Fe film on GaAs (110) before and after deposition.

This work was supported by Deutsche Forschungsgemeinschaft (DFG, German Research Foundation): Project Nos. RO 5030/2-1 and 405553726-TRR 270, and by the Government of the Russian Federation (Research Grant No. 075-15-2019-1886). The authors acknowledge Florian Römer for initial funding and training and are thankful to Benjamin Zingsem for helping in data fitting and to Ulf Wiedwald for AFM measurements.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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DOI: 10.1063/5.0004261
URN: urn:nbn:de:hbz:464-20210113-104706-4