In-plane magnetic anisotropies in Fe films on vicinal Ag(001) and Au(001) surfaces

T. Leeb, M. Brockmann, F. Bensch, S. Miethaner, and G. Bayreuther*1)
Institut für Experimentelle und Angewandte Physik, Universität Regensburg, 93040 Regensburg, Germany

In-plane fourfold and uniaxial magnetic anisotropies were studied in Fe films epitaxially grown on vicinal Ag(001) and Au(001) surfaces, which were prepared by molecular beam epitaxy on miscut GaAs(001) substrates. The effective fourfold and uniaxial anisotropy constants, $K_4^\text{eff}$ and $K_u^\text{eff}$, which are determined from magnetisation curves measured with the magneto-optic Kerr effect, are linear functions of the inverse Fe layer thickness. The fourfold anisotropy shows a rotation of the easy and hard axes by 45° below a critical thickness of 6–7 ML. The uniaxial term is mainly an interface contribution. We find that the orientation of the uniaxial easy axis depends on the Fe thickness. In thinner films ($t_{Fe} \leq 20$ ML) it is oriented perpendicular to the step edges, i.e., parallel to Fe[010]. This excludes shape anisotropy as the main mechanism. Above a critical thickness the uniaxial easy axis is aligned parallel to the step edges, i.e., along Fe[010]. This step-induced uniaxial anisotropy may be due to modified electronic states and to strain from the large vertical misfit at the steps.

© 1999 American Institute of Physics. [S0021-8979(99)64408-5]

The magnetic properties of ferromagnetic thin films grown on surfaces with reduced symmetry is of great technological importance and of fundamental scientific interest. Several research groups have shown that the specific symmetry of a ferromagnetic film epitaxially grown on a vicinal substrate surface, i.e., a quasiperiodic sequence of monoatomic steps, will usually produce a uniaxial magnetic anisotropy which is not expected in a ferromagnetic film with fourfold symmetry like Fe(001). This has been found in Fe/W(001),1,2 Co/Cu(001),3–8 and Fe/Ag(001).9 In these studies bulk metal single crystals miscut at a small angle against a low indexed surface were used as substrates. The intention of the present work was to use Ag and Au films epitaxially grown on miscut GaAs wafers to produce vicinal surfaces for the epitaxy of ultrathin Fe films. The obvious advantage of this approach compared to the use of bulk single crystals of Ag and Au is the much lower cost and the advantage of this approach compared to the use of bulk single crystals of Ag and Au.

All films were grown on a GaAs(001) single-crystal wafer with the surface tilted by 2° with the [010] direction as the axis of rotation. During film deposition by molecular beam epitaxy (MBE) the residual pressure in the chamber remained below $p = 5 \times 10^{-10}$ mbar. The surface structure of the films was studied in situ by reflection high energy electron diffraction (RHEED), low energy electron diffraction (LEED), and scanning tunneling microscopy (STM). Film thickness was monitored by four quartz microbalances calibrated by x-ray fluorescence spectroscopy and RHEED intensity oscillations with an error of <10%.

First, the substrate surface was cleaned in UHV by combined annealing and Ar ion etching at 850 K with an ion beam energy of 500 eV.10 As a result, a sixfold reconstruction of the GaAs surface was observed by STM. Two different buffer layers were grown at room temperature on the same substrate. After deposition of a 10 ML Fe seed layer, 750 ML of Ag were deposited on one half of the substrate, 450 ML of Au were grown on the second half at a deposition rate of 1–3 ML/min. After annealing the system at 600 K for 1 h the surface was cleaned from segregating Ga and As atoms by Ar ion etching. Further annealing at 400 K lead to uniformly stepped Ag and Au surfaces with step edges along the Ag and Au [110] direction, which is parallel to the [010] direction of the GaAs substrate and the Fe layers. Typical STM images of these surfaces are shown in Fig. 1. The separation of the monoatomic steps is usually 5–8 nm, but annealing at elevated temperature leads to some step bunching, i.e., the formation of some terraces wider than 30 nm with steps over three or more vertical lattice spacings. For the vicinal Au(001) buffer layer LEED patterns and STM images clearly show the fivefold periodicity of the (5 x 20) surface reconstruction with rows of atoms oriented along the step edges uniformly over the whole surface in contrast to nonvicinal Au(001) surfaces where always two kinds of reconstructed domains are present with the atomic rows at right angles. The reconstruction ridges are slightly rotated with respect to the Au[110] direction leading to the periodic formation of kinks along the step edges [see Fig. 1(d)].

Fe films were grown on both buffer layers in multiple square patches of constant Fe thickness between 3 and 50 ML at room temperature and a rate of 0.8–1 ML/min. STM images show small mounds of about 5 nm in diameter on the Fe surface similar to observations on homoepitaxy of Fe.11

---

*Electronic mail: Guenther.Bayreuther@physik.uni-regensburg.de
The mound height of about 0.3–0.5 nm suggests that these islands consist of 2–3 atomic Fe(001) layers.

From the fact that both the average mound diameter and height remain practically unchanged over a wide thickness range we assume an approximate layer-by-layer growth mode of the Fe films on vicinal Ag and Au surfaces at the given growth conditions.

The Fe film was finally covered with a protective layer: 14 ML Ag +10 ML Au on the sample part with the Ag buffer and 20 ML Au on the other half. The periodic step structure of the vicinal substrate surface is well reproduced as indicated by the STM images. The Au covering layer again shows a single domain (5 ×20) surface reconstruction in the LEED and STM data indicating a very low contamination level and a continuous Au film with atomically flat terraces.

Local magnetization loops, \( m(H) \), normalized to the saturation moment, were measured at room temperature with the longitudinal magneto-optic Kerr effect (MOKE) along different directions. The external field, \( H \), was always applied within the (001) film plane. No indication for an out-of-plane magnetization component was observed.

The angular dependence of the \( m(H) \) loops showed the presence of the fourfold anisotropy expected for cubic Fe and an additional uniaxial in-plane anisotropy with its easy axis parallel to the Fe[100] for films of 20 ML or thinner.

The effective anisotropy constants of the uniaxial and the fourfold anisotropies, \( K_u^{\text{eff}} \) and \( K_i^{\text{eff}} \), were determined by applying the variable external field \( H \) along the Fe[010] axis with an additional constant bias field \( H_{\text{bias}} \) oriented along Fe[100], perpendicular to the sweep field direction according to Weber, Allenbach, and Bischof.\(^{12}\) Within a single domain model the following expression was used for the energy per unit volume, \( e(\varphi) \):

\[
e(\varphi, H) = K_0 + \frac{K_i^{\text{eff}}}{4} \sin^2(2\varphi) + K_u^{\text{eff}} \sin^2(\varphi) - M_s H_{\text{bias}} \cos(\varphi) - M_s H \sin(\varphi),
\]

where \( \varphi \) is the angle of the magnetization relative to the Fe[100] direction. \( K_0 \) accounts for an isotropic contribution, the second term corresponds to the cubic and the third one to the uniaxial contribution within the film plane. The last two terms account for the Zeeman energies due to the applied magnetic fields.

Both the uniaxial and the fourfold in-plane anisotropy constants are determined from one single hysteresis loop, as shown in Fig. 2, by measuring the shift of the minor loop \( H_{\text{split}} \) (‘‘split field’’) and the slope \( \chi \) around \( H=0 \), respectively.\(^{12}\) Minimizing the energy in Eq. (1) with respect to \( \varphi \) yields

\[
\chi = (2K_i^{\text{eff}} + 2K_u^{\text{eff}} + H_{\text{bias}}) M_s^{-1},
\]

where \( K_u^{\text{eff}} \) is determined from the split field, \( H_{\text{split}} \) according to\(^{12}\)

\[
K_u^{\text{eff}} = (H_{\text{split}} - H_{\text{bias}}) M_s.
\]

Figure 3 shows the resulting values for \( K_u^{\text{eff}} \) and \( K_i^{\text{eff}} \) plotted versus the reciprocal Fe thickness both for Ag/Fe/Ag (a) and Au/Fe/Au (b). Both anisotropy constants are linear functions of the inverse Fe thickness and can be fitted with the following expression \((i=1,u)\):

\[
K_i^{\text{eff}} = K_i^V + \frac{2K_i^S}{Nd_{001}},
\]

where the Fe thickness is given in multiples, \( N \), of the interlayer distance, \( d_{001} \), of the Fe(001) film. \( K_i^V \) are the volume and \( K_i^S \) the interface contributions to the effective uniaxial or cubic anisotropy constants, \( K_i^{\text{eff}} \). The slope of the linear fit is positive for \( K_u^{\text{eff}} \) and negative for \( K_i^{\text{eff}} \). The linear fit of the effective cubic anisotropy constants, \( K_i^{\text{eff}} \), for Ag/Fe/Ag yields a volume contribution of \( K_i^V = (4.7 \pm 0.1) \times 10^5 \text{ erg/cm}^3 \) in agreement with the cubic anisotropy constant for bulk bcc Fe of \( K_i^V = 4.7 \times 10^5 \text{ erg/cm}^3 \). The negative interface term, \( K_i^S = -(2.1 \pm 0.1) \times 10^5 \text{ erg/cm}^2 \), is larger than the value of \( -1.5 \times 10^5 \text{ erg/cm}^2 \) found by Heinrich and Cochran\(^{13}\) for singular Fe/Ag(001). \( K_u^{\text{eff}} \) changes sign at a critical Fe thickness \( t_i^{\text{crit}} = (6.2 \pm 0.4) \text{ ML} \), indicating an in-
plane spin reorientation process of the fourfold anisotropy. This value for $t_{\text{crit}}^{\text{ag}}$ is somewhat larger than the value of 4.5 ML expected for singular Fe/Ag(001) by extrapolating ferromagnetic resonance (FMR) data of thicker films.\(^\text{13}\)

For the Fe/Au/Fe system we find a volume contribution of the cubic anisotropy of $K_u^S = (3.7 \pm 0.1) \times 10^5$ erg/cm\(^3\), which is smaller than the cubic anisotropy constant of bulk bcc Fe, but equals the result for nonvicinal Fe/Au(001). $K_u^S = 3.7 \times 10^5$ erg/cm\(^3\), found earlier.\(^\text{14}\) Also, the interface contribution $K_u^S = -(1.8 \pm 0.1) \times 10^{-2}$ erg/cm\(^2\) and as a consequence the critical thickness for the expected spin reorientation of the fourfold anisotropy $t_{\text{crit}}^{\text{ag}} = (6.9 \pm 0.2)$ ML agree well with the previous results.\(^\text{14}\) $K_u^S = -(1.95 \pm 0.2) \times 10^{-2}$ erg/cm\(^2\) and $t_{\text{crit}}^{\text{ag}} = (7.3 \pm 0.7)$ ML, respectively.

Concerning the uniaxial anisotropy, Fig. 3 indicates that this is mainly an interface contribution. For Ag/Fe/Ag we obtain $K_u^S = (4.7 \pm 0.2) \times 10^{-3}$ erg/cm\(^2\), while for Au/Fe/Au we get a larger value of $K_u^S = (6.5 \pm 0.3) \times 10^{-3}$ erg/cm\(^2\). The positive sign corresponds to an easy axis perpendicular to the steps, i.e., parallel to Fe [100]. However, the fit also gives finite negative volume terms of $K_u^V = -(2.3 \pm 0.2) \times 10^4$ erg/cm\(^3\) for Ag/Fe/Ag and $K_u^V = -(1.6 \pm 0.2) \times 10^4$ erg/cm\(^3\) for Au/Fe/Au. The corresponding critical thickness for a rotation of the uniaxial easy axis by 90° is $t_{\text{crit}}^{\text{ag}} = (29 \pm 4)$ ML for Ag/Fe/Ag and $t_{\text{crit}}^{\text{ag}} = (56 \pm 11)$ ML for Au/Fe/Au. This is roughly compatible with the results of Kawakami, Escorcia-Aparicio, and Qiù \(^\text{10}\) who report a uniaxially easy axis parallel to the step edges for a 25 ML Fe film on a vicinal Ag single crystal with a miscut of 2.6°–10° with respect to Ag(001).

In ultrathin Fe films epitaxially grown on vicinal Ag(001) and Au(001) surfaces we have observed a fourfold and a uniaxial in-plane magnetic anisotropy which vary linearly with the inverse film thickness. The origin of the fourfold interface anisotropy seems to be the modification of the electronic structure at the interface, in particular, enhanced orbital moments at the interface compared to the bulk crystal with cubic symmetry.

The origin of the step induced uniaxial anisotropy is poorly understood at present. Dipolar interaction would cause the easy axis to be parallel to the steps in contrast to all but our thickest films ($t_{\text{Fe}} = 50$ ML). Instead, symmetry breaking at the steps and the related change in electronic states could be responsible for the uniaxial anisotropy, but also strain might be a major source of the uniaxial term. Unlike the small in-plane lattice mismatch between Fe(001) and Ag(001)/Au(001) (taking into account the rotation by 45° of the Fe lattice relative to Ag/Au), a large vertical mismatch of nominally 30% exists which gives rise to strong lattice strain fields in the neighborhood of monoatomic steps and hence to a uniaxial magnetic anisotropy. Such a contribution should vanish with increasing thickness because of the local strain is confined to the interface. To clarify the origin of an apparent uniaxial volume term the measurements will be extended to even thicker Fe films ($t_{\text{Fe}} = 20–200$ ML). Experiments are under way to check whether the linear dependence of $K_u^S$ on the inverse Fe thickness breaks down above 50 ML.

---

**FIG. 3.** The effective anisotropy constants, $K_u^{\text{eff}}$ and $K_u^{\text{eff}}$, as a function of the inverse Fe thickness for the system (a) Ag/Fe/Ag(001) and (b) Au/Fe/Au(001). The critical thickness values of the reorientation transition of both the uniaxial and the fourfold anisotropy were taken from linear fits and are marked by arrows.

---