ATOMIC STRUCTURE AND MAGNETIC ANISOTROPY IN ULTRATHIN
Fe FILMS ON Au(001)

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Abstract - We have investigated the growth and magnetic properties of ultrathin Fe films on Au(001). The films used for magnetometry have been prepared by MBE on pre-annealed MgO(001) single crystals. Epitaxial growth of the 300-500 monolayer Au buffer layer as well as of the Fe films in the monolayer range could be achieved by use of a Cr seed layer. Epitaxial quality and surface structure was controlled in situ by electron diffraction (LEED, RHEED) and ex situ by STM and AFM. Comparative growth studies were performed for Fe-films on a Au single crystal substrate by in situ STM and ion scattering spectroscopy. We have investigated the magnetic anisotropy of Fe films in the range 3-50 monolayers with alternating gradient magnetometry (AGM). We find a uniaxial out-of-plane contribution of the surface anisotropy which promotes a spontaneous out-of-plane magnetization for films < 2.1 monolayers. Additionally, we report on a fourfold in-plane surface anisotropy, which leads to a switching of the in-plane easy axes into the [110] and [111] directions for the 3 and 5 monolayer Fe films. We discuss these anisotropy phenomena in context with the breaking of translational symmetry at the interface.

KEYWORDS: MAGNETIC ANISOTROPY, IN-PLANE ANISOTROPY, STRUCTURE, ULTRATHIN FILMS, Fe/Au(001).

INTRODUCTION

Magnetic anisotropy is a key property of magnetic films used for magneto-optic recording. In magnetic ultrathin films and multilayers the anisotropy is a result of volume and interface contributions of dipolar, magneto-crystalline and magneto-elastic origin. This variety of influencing factors opens the way to create new magnetic materials with desired anisotropies by choosing a proper combination of magnetic and nonmagnetic layers of appropriate thicknesses.

However, the interpretation of experimental data using a phenomenological classification as given above is somewhat doubtful because the related anisotropy or magnetoelastic constants at interfaces and in ultrathin films in general will be substantially different from the corresponding bulk values and are not known a priori.

On the other hand, considerable progress has been made in recent years regarding ab initio band theory of magnetic materials. The inclusion of spin-orbit interaction in density functional theory using the local density approximation has made it possible to predict magnetic anisotropies of ultrathin films with surprising success (e.g.[1]-[4]). This is valid for the uniaxial magnetic anisotropy with respect to the film normal which arises from broken cubic symmetry at the interfaces in 3d ferromagnetic metals. It is of second order in spin-orbit coupling and hence considerably larger than bulk anisotropy in cubic 3d ferromagnets. From pure symmetry arguments, however, we can expect both a perpendicular interface anisotropy as mentioned before and an interface anisotropy term within the film plane. Such a contribution has been found experimentally (e.g. [5], [6]) but it is out of reach of present day's ab initio band calculations because the corresponding energies are very small. Therefore, we have to restrict the discussion of these anisotropies to the phenomenological scheme used above.

Heinrich et al. [5] implicitly assume that magnetic surface anisotropies are generated by lattice distortions at the interfaces, either by surface relaxation which produces a tetragonal distortion perpendicular to the plane and a correlated perpendicular uniaxial anisotropy, or by a network of misfit dislocations with the appropriate in-plane fourfold symmetry in order to explain the fourfold interface anisotropy observed in Fe(001)/Ag(001) epitaxial films. According to their argument, no interface anisotropy would exist for a magnetic film grown on a substrate with perfect lattice match in the absence of surface relaxation. This interpretation ignores the fact that, even in a film without any distortion of the atomic structure, the breaking of the translational symmetry at the interface will distort the electronic structure which, in turn, is the source of magnetic order and anisotropy via spin-orbit interaction. This means that a purely electronic contribution to magnetic interface anisotropies has to be expected; this interpretation is in agreement with the original concept of magnetic surface anisotropy proposed by Néel [7].

Obviously, in reality the purely electronic effect cannot be separated from the other mechanisms because lattice distortions themselves are a consequence of altered electronic interactions at interfaces. This means that electronic and atomic structure are intimately connected with each other and will have to be treated selfconsistently in a future unified solid state theory of magnetic materials.

In the present work we attempt to study the in-plane interface anisotropy of Fe(001) ultrathin films grown on Au(001) with the intention to correlate magnetic anisotropy with the growth mode and atomic structure of the films. In particular, we want to investigate the question if the fourfold interface anisotropy deduced from previous experimental data [5] is an effect of a specific dislocation network or if it is an intrinsic property of Fe(001) interfaces which also occurs in films with highly perfect structure.

Fe(001)/Au(001) films were grown on MgO single crystal substrates. Film structure was characterized by several methods in situ and ex situ. The effect of Fe deposition on the (5x5') reconstruction of the Au(001) surface was also studied on a bulk Au single crystal for comparison. Magnetic properties were determined by an alternating gradient magnetometer and torque magnetometry at room temperature. A change of sign of the in-plane cubic anisotropy constant was observed for 5 and 3 monolayer (ML) thick Fe(001) films which shows up as a rotation of the easy axes from the [100], [010] and equivalent directions to the [110] etc. directions.
GROWTH AND STRUCTURE

The growth of Fe on the clean (001) surface of a Au single crystal substrate at room temperature (RT) was studied in situ by STM, LEED and ion scattering spectroscopy (ISS). The clean (001) surface shows a reconstruction which, depending on the resolution of the method, has been described by (5x1), (5x20), c(26x68) or even more complicated superstructures (e.g. [8],[9]). In its initial stage the Fe growth is influenced by the Au reconstruction. The Fe nucleation takes place preferentially along ridges of the reconstruction which is destroyed in the vicinity of the iron deposit.

At an iron coverage of approximately 0.2 Monolayers (ML) the Au reconstruction is no longer visible in STM and LEED. ISS spectra measured at several Fe coverages show that 1...2 ML of Au are floating on top of the Fe film, apparently promoting a two dimensional growth mode. The atoms of this floating Au layer are gradually incorporated into the growing iron film.

The films used for magnetometry where prepared on MgO(001) substrates by MBE at \( p < 5 \cdot 10^{-10} \) Torr during evaporation. The MgO substrates were previously annealed under oxygen flow at 1050°C which results in a clean and atomically flat surface with terrace widths of several hundred nm. The substrate morphology was checked by AFM. A gold buffer layer was grown on the MgO using a thin Cr seed layer of 3 ML. The growth mode and the surface structure were studied in situ by RHEED, LEED and AES. The 10keV-RHEED images (as in fig. 1a) show a clear streak pattern, indicating a not perfect but still very smooth surface. Furthermore RHEED and LEED patterns (fig. 1a and 1b, respectively) include streaks and spots of fractional order with a spacing of 1/5th of the regular (01)-streaks/spots as a result of the (5x1') surface reconstruction growth mode.

Upon deposition of 0.2 ML Fe the reconstruction streaks and spots vanish and the diffraction patterns of the unreconstructed iron lattice emerge (fig. 2). At iron coverages between 2 and 10 ML, the patterns are rather diffuse (higher background intensity), which can be explained by an increasing disorder of the Au layer floating on top of the Fe film. At coverages above 10 ML the background intensity and the spot widths decrease, probably because atoms of the disordered Au surface layer get incorporated into the Fe film at 300 K. All films were finally covered by a protective layer of Au (at least 20 ML). During the growth of this layer, the (5x1') reconstruction re-appeared after only 2-3 ML of Au (fig. 3). This indicates a nearly perfect two-dimensional growth.

Fig. 1a: RHEED pattern of a 400 ML Au buffer layer. The arrows indicate the diffraction streaks corresponding to the bulk lattice. The additional streaks are due to the surface reconstruction.

Fig. 1b: LEED image of the Au(001) surface at 58 eV, also showing the (5x1') reconstruction.

Fig. 2a: RHEED pattern of 0.2 ML Fe deposited on the Au(001) surface. The (5x1') reconstruction has almost disappeared.

Fig. 2b: LEED image of 0.2 ML Fe deposited on the Au(001) surface at 58 eV.
MAGNETIC ANISOTROPY

Magnetic anisotropy of several samples has been investigated by alternating gradient magnetometry and torque magnetometry. For film thicknesses of 3 and 5 ML Fe a decrease of the saturation field parallel to the film normal was found. From extrapolation to lower film thickness the critical thickness \( t_e \) for spontaneous out-of-plane magnetization could be determined as \( t_e = 2.1 \) ML at room temperature. This is not discussed further in this communication.

In addition, we investigated the in-plane anisotropy for various Fe thicknesses. First, the anhysteretic magnetization curves were determined by averaging both branches of the \( M(H) \) loop in the \( \langle 100 \rangle \) axis. The energy required to saturate the film was calculated and identified with the anisotropy energy according to

\[
E_a = \int_0^M_s H dM
\]

The in-plane anisotropy energy as a function of the angle \( \varphi \) between the applied field and the [100] crystal axis was fitted with the following expression:

\[
E_a = K_0 + K_{eff} \sin^2(2\varphi + \omega) + K_{s} \sin^2(\varphi + \gamma)
\]

where \( K_{eff} = K_{||} + \frac{1}{N \cdot d_{001}} K_{s} \) is an effective cubic anisotropy constant consisting of a volume term \( K_{||} \) and a surface term \( K_{s} \) (\( N \): number of monolayers).

A 50 ML Fe film showed bulk behaviour with easy axes parallel to [100] and [010], corresponding to a positive \( K_{||} \) (fig. 4).

For Fe thicknesses of 3 and 5 ML we observed a switching of the easy axes to the [110] and [110] directions (fig. 5). This means that \( K_{s} \) is negative and overcomes the bulk cubic anisotropy.

Our results are in good agreement with the experiments of Heinrich et al. (e.g. [5],[6]) on Fe/Ag(001) samples. By extrapolating their FMR data these authors predicted the rotation of the easy axes for an Fe thickness < 4.6 ML.
Fig. 6 shows the anisotropy energy as a function of the angle between the [100] crystal axis and the applied field. In addition to the fourfold anisotropy a uniaxial contribution resulting in an asymmetric anisotropy energy in [100] and [010] directions is observed. This twofold anisotropy may be attributed to the step structure of the sample. This is suggested by STM images of the Au covering layer. A quantitative check will be made possible by in situ STM measurements on a larger scale of the substrate surface.

An additional rotation of the fourfold term by approx. 7° with respect to the cubic axes is not understood at present. It might be connected with the nucleation of Fe on the reconstructed Au(001) surface which can be described as a hexagonal layer slightly rotated with respect to the underlying square lattice.

CONCLUSIONS

We have investigated the magnetic anisotropy in Fe(001) ultrathin films epitaxially grown on reconstructed Au(001) surfaces. LEED, RHEED and STM data suggest that the films grow essentially in a two-dimensional way with flat interfaces. Magnetic measurements reveal a pronounced fourfold magnetic anisotropy in the film plane which is essentially like in bulk Fe for films of 50 ML or thicker. For an Fe layer thickness of 5 and 3 ML we observe a rotation of the easy axes by 45° which is equivalent to a change of sign of the effective cubic anisotropy constant in the plane of the film. This result together with the structural data suggest that the sign reversal is not an effect of a special defect structure but an intrinsic property of Fe(001) interfaces. This can be understood on the ground of the altered electronic band structure at the interface, in particular due to strongly enhanced orbital moments first predicted theoretically [10] and observed experimentally in recent XMCD measurements [11].

These findings emphasize the potential of interface effects in magnetic multilayers to facilitate the creation of new materials e.g. for future ultrahigh density magnetic and magneto-optic information storage media.

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REFERENCES