Perpendicular Magnetic Anisotropy Induced by Tetragonal Distortion of FeCo Alloy Films Grown on Pd(001)

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We grew tetragonally distorted Fe_xCo_{1-x} alloy films on Pd(001). Theoretical first-principles calculations for such films predicted a high saturation magnetization and a high uniaxial magnetic anisotropy energy for specific values of the lattice distortion c/a and the alloy composition x. The magnetic anisotropy was investigated using the magneto-optical Kerr effect. An out-of-plane easy axis of magnetization was observed for $Fe_{0.5}Co_{0.5}$ films in the thickness range of 4 to 14 monolayers. The magnetic anisotropy energy induced by the tetragonal distortion is estimated to be almost 2 orders of magnitude larger than the value for bulk FeCo alloys. Using LEED Kikuchi patterns, a change of the easy axis of magnetization can be related to a decrease of the tetragonal distortion with thickness.

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The increase of the recording density in hard disk drives requires new media materials with both a large uniaxial magnetic anisotropy energy (MAE) K_u and a large saturation magnetization M_s. To avoid the superparamagnetic limit, the stored magnetic energy $K_u V$ in the grain volume V has to be high compared to the thermal energy. At the same time, a large saturation magnetization ensures that the available writing fields H_W can switch the magnetization of the grains: $H_W \approx K_u/M_s$. Based on first-principles theory, it was predicted that structural distortions of FeCo alloys result in a combination of large uniaxial MAE and saturation magnetization which is yet unrivaled [1]. The MAE of the cubic forms of Fe and Co which is in the order of only few μeV/atom [2] should be increased by several orders of magnitude in tetragonally distorted alloys. They also retain high values of the saturation magnetization as is well known from their cubic forms [3].

The calculations of Burkert *et al.* predict an unusually large magnetic anisotropy energy for the $Fe_{0.4}Co_{0.6}$ alloy films at a c/a ratio of about 1.2 [1], the bcc structure having a c/a ratio of 1 and fcc of $\sqrt{2}$. The maximum magnetic anisotropy energy is predicted to be between 700 and 800 μeV per atom and the average magnetization to be about $2.1\mu_B$/atom. Tetragonally distorted Fe and Co films grown on Pd(001) have been observed previously [4,5], with c/a ratios of 1.11 for Fe [4] and 1.15 for Co [5]. This suggests that Fe_xCo_{1-x} alloy films grown on Pd(001) could have a similar distortion, while theory predicts an increased anisotropy energy of about 200 μeV/atom at an average magnetization of $2.1\mu_B$/atom in this distortion range. Accordingly, the maximum anisotropy should be reached at a chemical composition around $x = 0.5$.

In this Letter we demonstrate that it is possible to grow such tetragonally distorted Fe_xCo_{1-x} films on Pd(001). Kikuchi patterns of quasielastically backscattered electrons recorded in our low energy electron diffraction (LEED) apparatus are used to quantitatively assess the amount of distortion. Magnetic properties are analyzed utilizing the *in-situ* magneto-optical Kerr effect (MOKE).

The experiments were carried out in a multichamber ultrahigh vacuum (UHV) system with a base pressure better than 5×10^{-11} mbar and less than 2×10^{-10} mbar during deposition. The Pd(001) substrates were prepared using sputter-anneal cycles. The quality of the sample was checked by Auger electron spectroscopy (AES) and by scanning tunneling microscopy (STM) until a clean surface with nearly equidistant, parallel monoatomic steps was obtained. The films were grown at 295 K by molecular beam epitaxy (MBE) using two effusion cells. To obtain a specified alloy composition, prior to deposition the corresponding rates were calibrated as a function of cell temperature. Thickness calibration was performed using reflection high energy electron diffraction (RHEED) oscillations and AES, alloy composition was checked using x-ray photoelectron spectroscopy (XPS). Polar and longitudinal MOKE were used to detect the out-of-plane and in-plane magnetization, respectively [6].

Both Fe and Co films are subject to lattice misfit when deposited on Pd(001). Namely, while the in-plane lattice constant of Pd(001) amounts to $a_{Pd} = 2.75$ Å, the corresponding lattice constants of Fe and Co are $a_{Fe} = 2.54$ Å and $a_{Co} = 2.51$ Å, respectively, resulting in large misfit values for fcc Fe(−7.7%) and fcc Co(−8.8%) [7]. As the Pd in-plane lattice constant is larger, the overlayer lattice has to expand in-plane for pseudomorphic growth. To keep the atomic volume constant, the film contracts in the direction of the c axis which leads to a c/a ratio less than $\sqrt{2}$. Because of the large lattice misfit, a perfect layer-by-layer growth is not expected for the Fe_xCo_{1-x} alloy films on Pd(001). Correspondingly, the LEED patterns show an increased background intensity. However, the spot analysis of the LEED patterns proves a still pseudomorphic growth of at least part of the Fe_xCo_{1-x} films because the LEED spots of the films stay at the same positions as the substrate spots. From this observation, the in-plane lattice constant of the well-ordered part of the film is thus determined to be $a_{Pd} = 2.75$ Å. Damped RHEED oscillations were observed, indicating initial layer-by-layer growth which rapidly decays with increasing film thickness. This was confirmed by STM observations.

Kikuchi patterns obtained in the LEED apparatus at 1000 eV allowed us to estimate the c/a ratio of the films, because they are providing instantaneous real-space information on the local order of the crystal structure. On the other hand, to obtain similar information from LEED patterns long-range order in the sample would be required, which is not sufficiently fulfilled in our case. The quasi-elastically backscattered electrons can be thought to be emitted from pointlike sources inside the crystal and are then predominantly scattered in the forward direction by the surrounding atoms on their way out of the sample [8]. In this way, interatomic scattering directions show up as intensity maxima. Together with the information about the in-plane lattice constant drawn from the LEED pattern, it is possible to determine the dimension of the unit cell. We used a multiple scattering cluster program [9] to simulate the observed changes in the Kikuchi patterns and to estimate the c/a ratio of the grown film. Clusters of about 500 atoms we used for the calculation, we assumed an inelastic mean free path of 10 Å at 1000 eV and took into account 7 layers of scattering atoms, which were assumed to emit the backscattered electrons spherically isotropic.

In Fig. 1, a clear change in the Kikuchi patterns from the Pd(001) substrate [Fig. 1(a)] to a grown film [Fig. 1(c)] is visible, directly indicating an alteration of the crystal structure. The sample shown is a $Fe_{0.4}Co_{0.6}$ film. A slight deviation from the ideal symmetry in the experimental patterns is caused by inhomogeneous sensitivity of our LEED system across the screen. A fit of simulated to the experimental Kikuchi patterns was leading to a c/a ratio of about 1.13, as judged by the position of the forward-scattering maxima. From the dashed lines in Fig. 1 it can be seen that the simulations reproduce the positions of the experimentally observed intensity maxima and their relative shifts very well. The value obtained is consistent with quasikinematical LEED intensity measurements, which however could be done only in a limited energy range due to the large background intensity. Because of the limited accuracy of the whole image recording system we have to assume an error of about 0.02 in the c/a ratio. Within the experimental accuracy, no change in distortion is detectable for alloy compositions between $x = 0.4$ and $x = 0.6$ for the same film thickness. The values thus obtained are fully consistent with the observations for pure Fe and Co films on Pd.

We observe a changing tetragonal distortion with increasing film thickness. The c/a ratio decreases by the appearance of lattice defects and the transition to a more

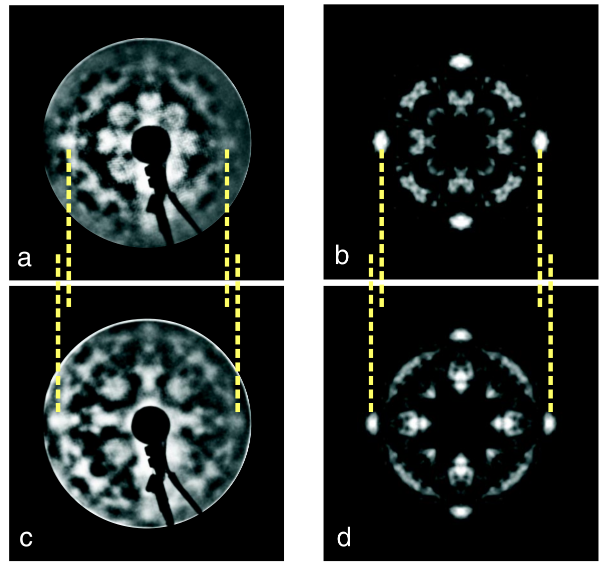

FIG. 1 (color online). Kikuchi patterns obtained (a) experimentally in the LEED apparatus at 1 keV from the clean Pd(001) substrate, (b) by multiple scattering simulation of clean Pd(001), (c) experimentally from 15 ML $Fe_{0.4}Co_{0.6}$, and (d) by multiple scattering simulation of 15 ML $Fe_{0.4}Co_{0.6}$ with $c/a = 1.13$. Intensity maxima used for the calculations are marked with dashed lines. The central areas in the simulations have been rendered black to account for the shadowing by the LEED gun.

bcc-like structure which reduces the strain originating from the large lattice misfit with respect to the Pd(001) surface. To show this, we deposited a "two-step" sample of 8 ML and 38 ML $Fe_{0.5}Co_{0.5}$ in order to take Kikuchi patterns from both parts of the sample in a single experiment. Also, we can simultaneously apply the structural and magnetic analysis safely for such samples, because the limited lateral resolution of the electron beam has to be taken into account. Finer stepped wedge samples are used for MOKE analysis alone. In Fig. 2 we show how the change in the pattern between 8 ML (A) and 38 ML (B) can be made visible by calculating an asymmetry $(B - A)/(B + A)$. In the asymmetry pattern, denoted "change" in Fig. 2, the regions with positive values are shown light; the regions with negative values are dark. As can be seen from the Kikuchi bands in the outer regions of the patterns, there are light areas at higher angles and the corresponding dark areas at lower angles. This means that the forward-scattering directions move to higher angles in the 38 ML film as compared to the 8 ML film. Correspondingly, the c/a ratio decreases with increasing thickness as expected. From the Kikuchi patterns we estimate the c/a ratio to be below 1.10 for the 38 ML film. As will be seen below, there are corresponding changes in the magnetic behavior of the films.

To investigate the magnetic properties of the samples, we measured thickness dependent hysteresis loops at 70 K using MOKE on ultrathin films grown as wedges. At first we checked the magnetic anisotropy of the Co/Pd(001)

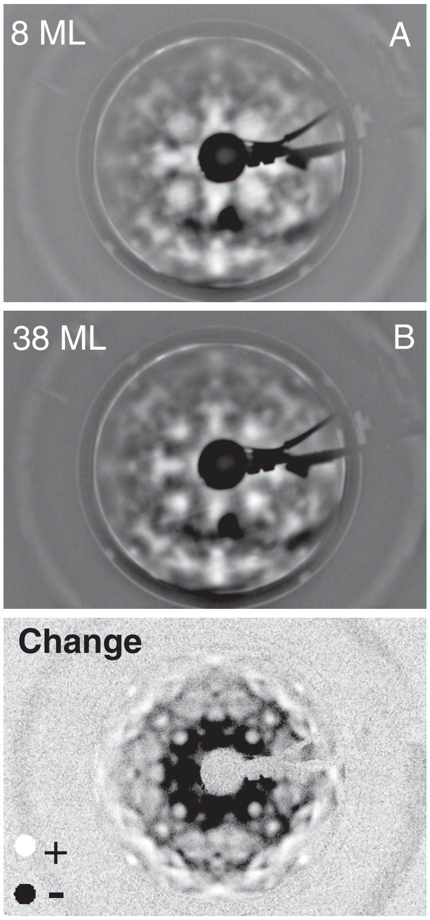

FIG. 2. LEED Kikuchi patterns of an $Fe_{0.5}Co_{0.5}$ alloy film at a beam energy of 1000 eV. (A) 8 ML thick sample area, (B) 38 ML thick sample area, (change) normalized difference $(B - A)/(B + A)$ of both patterns indicating a decreasing c/a ratio.

and Fe/Pd(001) systems. No polar MOKE signal was detected for pure Co and Fe films in the thickness range investigated (up to 14 ML). The in-plane [110] direction was found to be the easy axis of magnetization. As has been shown elsewhere [6], an out-of-plane easy axis of magnetization can be obtained in the Co/Pd(001) system under very specific, different experimental conditions than applied here.

Results of MOKE measurements for three different Fe_xCo_{1-x} alloys are shown in Fig. 3. For the discussion of the effects of interest in this Letter, we limit ourselves to the thicknesses above 4 ML because deviations from a linear increase of the saturated MOKE signal are observed below this thickness. We ascribe this to interface effects. It is known that the Co/Pd(001) and Fe/Pd(001) interfaces can have considerable influence on the observed MOKE signal [6]. Also, the problem of interface alloying in these systems has been extensively discussed [10–12]. Considering these previous studies, the formation of the interface can be assumed to be finished above 4 ML in our system.

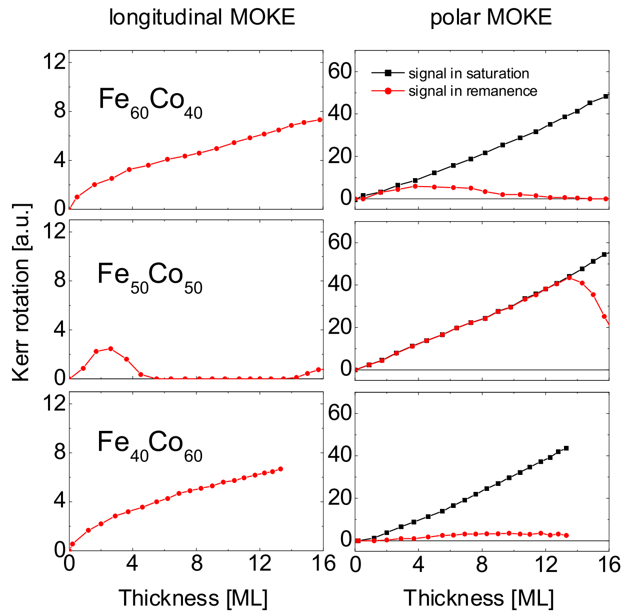

FIG. 3 (color online). Thickness dependent measurements of the magneto-optical Kerr rotation in remanence (gray circles) and saturation (black squares) for the grown Fe_xCo_{1-x} alloy films, for incident p-polarized light.

In the MOKE results shown in Fig. 3, the easy axis of magnetization is characterized by equal remanent and saturation signals. We illustrate for two examples of $x > 0.5$ (upper panels) and $x < 0.5$ (lower panels) that an in-plane easy-axis of magnetization was detected for all Fe_xCo_{1-x} alloy films in which the composition differs by more than about 10% from equiatomic. Here, the remanent *longitudinal* MOKE signal is equal to the saturation signal (not shown). In contrast to this, the nominal equiatomic alloy $x = 0.5$ showed a clear out-of-plane easy axis of magnetization. This is seen in the middle part of Fig. 3 by the disappearing longitudinal signal and the equal *polar* remanent and saturation signals. The increase of the perpendicular anisotropy proceeds smoothly as we could already saturate our samples at the chemical compositions between $Fe_{0.6}Co_{0.4}$ and $Fe_{0.4}Co_{0.6}$ perpendicular to the sample plane with the available magnetic field. A small remanent polar signal in these films is also a sign of the increasing perpendicular anisotropy (Fig. 3). These observations indicate a spin-reorientation transition from in plane to out of plane and back to in plane with increasing cobalt content. This clearly illustrates the influence of the chemical composition on the magnetic anisotropy.

As a function of film thickness, the easy axis of magnetization of the $Fe_{0.5}Co_{0.5}$ alloy films begins to gradually change back to in plane for thicknesses larger than about 14 ML (Fig. 3). To analyze this further, we used the sample of 8 ML and 38 ML $Fe_{0.5}Co_{0.5}$ mentioned earlier. The measured MOKE hysteresis loops clearly indicated an out-of-plane easy axis for the 8 ML and an in-plane easy axis for the 38 ML thick part of the film. This correlates

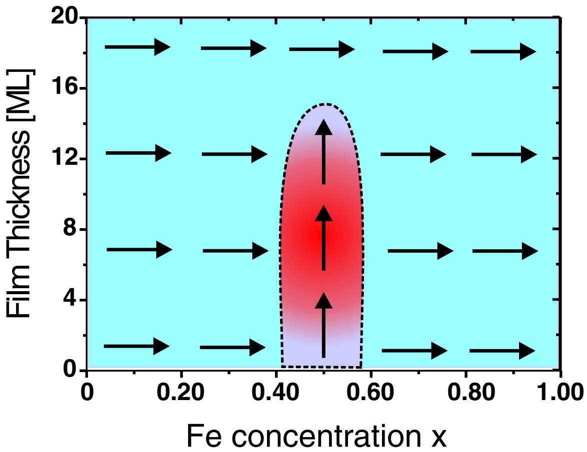

FIG. 4 (color online). Schematic diagram showing regions of perpendicular and in-plane easy axis of magnetization for Fe_xCo_{1-x} alloy films grown on Pd(001). Free parameters of the system are Fe concentration (horizontal axis) and total film thickness (vertical axis).

with the clearly decreasing tetragonal distortion which was found in the structural analysis for this sample, and thus strongly supports the interpretation that the change in tetragonal distortion is responsible for the change in magnetic anisotropy with thickness.

Because the calculations of Burkert *et al.* were done for bulk alloys, their results cannot be directly compared to the results we obtained for ultrathin films. In addition to the MAE calculated from the total energy differences between the [001] (the distortion axis) and the [100] directions of the magnetization, there is the magnetostatic energy $E_{dip} = -\frac{1}{2}\mu_0 M_s^2 t$ which tends to keep the magnetization of a thin film in the film plane. Assuming an average magnetization of $2.0\mu_B$ for the $Fe_{0.5}Co_{0.5}$ alloy, and the lattice parameters obtained from the Kikuchi pattern analysis, a shape anisotropy energy of about 120 μeV/atom has to be overcome to cause the magnetization to point out of the sample. Further important contributions to the overall anisotropy come from the film-substrate interface and the film surface. A perpendicular interface anisotropy energy of about 300 μeV/atom has been observed in Co/Pd multilayers [13], which however can only support perpendicular magnetization for Co thicknesses near 1 ML in the multilayer system of Co/Pd(001). With this in mind, and our observation that none of the considered Fe_xCo_{1-x} films with $x \neq 0.5$ showed an out-of-plane easy axis of magnetization, we have a reasonable indication that also for $x = 0.5$ interface plus surface anisotropies are probably not strong enough to cause a different behavior, especially for thicknesses above 4 ML. For the case of the Fe/Pd(001) interface, an *in-plane* easy axis is reported [10] (and references therein) with the anisotropy energy estimated to be about 80 μeV/atom. These estimations show that in any case there must be out-of-plane MAE which overcomes at least the shape anisotropy above 4 ML. The magnetostatic energy E_{dip} of 120 μeV/atom gives a lower bound for the amount of this energy. This value is almost 2 orders of magnitude larger than the MAE of only a few μeV/atom measured in the corresponding bulk alloys [2]. In addition, it is of the same order as the theoretically predicted MAE values for the c/a ratio measured in our films.

To summarize our results, we show in Fig. 4 a schematic phase diagram of Fe_xCo_{1-x} films grown tetragonally distorted on Pd(001). It illustrates the interplay between thickness (and related to that the tetragonal distortion), chemical composition, and magnetic anisotropy in the films studied. Near the equiatomic composition, these films showed an out-of-plane easy axis of magnetization. This is in agreement with the theoretical prediction that both a tetragonal distortion and a specific composition are necessary to result in the strong perpendicular anisotropy. We estimated a magnetic anisotropy energy which is almost 2 orders of magnitude larger than the bulk value. The c/a value of about 1.13 ± 0.02 obtained for the tetragonal distortion is consistent with the results formerly obtained for pure Fe and Co films on Pd(001). The perpendicular MAE reduces with film thickness correlated with a decrease in c/a ratio.

Note added.—Recently published work of G. Andersson *et al.* is related to our results [14].

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