

Spin reorientations induced by morphology changes in Fe/Ag(001)

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By means of magneto-optical Kerr effect we observe spin reorientations from in-plane to out-of-plane and vice versa upon annealing thin Fe films on Ag(001) at increasing temperatures. Scanning-tunneling microscopy images of the different Fe films are used to quantify the surface roughness. The observed spin reorientations can be explained with the experimentally acquired roughness parameters by taking into account the effect of roughness on both the magnetic dipolar and the magnetocrystalline anisotropy. [S0163-1829(99)01521-0]

Ultrathin ferromagnetic films have attracted enormous interest in recent years. Due to the broken symmetry at the surface their anisotropies are strongly altered compared to bulk values. In a phenomenological description one can distinguish between bulk and surface contributions to the effective anisotropy¹

$$K^{eff} = K_v + \frac{2K_s}{d}, \quad (1)$$

where K^{eff} denotes the effective, K_v the volume, and K_s the surface anisotropy. In the Fe/Ag(001) system, it has been found that the out-of-plane surface anisotropy can dominate the volume anisotropy in the ultrathin range and align the magnetization perpendicular to the surface.²⁻⁶ Recently, the temperature and thickness dependent spin reorientation transition has been further investigated on wedge-shaped samples.⁷⁻¹⁰ In spite of this much better understanding of the magnetic phase transition, there is still considerable disagreement in literature concerning the exact thickness and temperature where the transition occurs, which makes it difficult to compare experimental data with theory. As an example, we mention the discrepancy between the results recently obtained by Berger and Hopster¹⁰ and the magnetic phase diagram constructed by Qiu *et al.* in Ref. 8: For a 4.3-monolayer (ML)-thick Fe film on Ag(001), Berger and Hopster measure a temperature dependent spin reorientation from out-of-plane to in-plane at $T \approx 220$ K, whereas Qiu *et al.* observe that the out-of-plane configuration is stable up to 400 K at this thickness.

The importance of morphology in thin film magnetism has been realized from the very beginning of this field and quite some discussion arose about the growth mode of Fe on Ag(001),¹¹⁻¹⁵ but there are hardly any *direct* measurements of the influence of structure on magnetism. Without quantifying the morphology, Heinrich *et al.*¹⁶ observe for example an increased uniaxial anisotropy of Au/Fe/Ag sandwiches grown on Fe whiskers compared to those sandwiches grown on bulk Ag(001). They speculate that a significantly smaller density of steps on the Fe whisker substrates may be a reason for this observation.

In this study, we use scanning-tunneling microscopy (STM) to get direct space information of the morphology of our samples. Direct comparison of the structural results with magneto-optical Kerr effect (MOKE) measurements allows

us to observe spin reorientations induced solely by morphology changes. Morphology dependent spin reorientations have an influence on the magnetic phase diagram and might thus explain some of the discrepancies reported in literature.^{8,10}

Sample preparation and characterization with the exception of MOKE measurements are performed in an ultrahigh vacuum system with a base pressure of 5×10^{-11} mbar which is equipped with molecular-beam epitaxy, STM, low-energy electron diffraction (LEED), and x-ray photoemission electron spectroscopy (XPS). The magnetic measurements are performed *ex situ* with a MOKE setup that allows the detection of longitudinal and polar Kerr effect. The diameter of the laser spot is 100 μm . All measurements are carried out at room temperature (RT). As substrates, we use 150-nm-thick Ag films which are grown on Fe precovered GaAs(001) wafers. After growth at 100 °C and postannealing at 300 °C for one hour, we get high quality single crystalline Ag(001) films. More details about the substrate preparation can be found in Ref. 17. The Fe films are grown at RT onto the Ag(001) substrates. The evaporation rate is monitored by a quartz thickness monitor and is typically 0.5 ML/min. Wedge-shaped Fe films with a slope of 2 ML/mm are grown by linearly moving a shutter in front of the substrate during deposition. After deposition, the Fe films are either investigated as grown or after postannealing for half an hour at elevated temperatures. The temperature accuracy is estimated to be ± 10 °C. Before the magnetic measurements are performed, the samples are coated with 10 nm Ag in order to have two chemically identical interfaces of the Fe film and to protect the samples from air exposure.

Longitudinal Kerr effect is used to measure the in-plane component of the magnetization. For this purpose, the external magnetic field is applied in the plane of the film parallel to a [100] easy axis of the Fe film. In Fig. 1(a), the remanent magnetization divided by the saturation magnetization is plotted for Fe films directly after growth at RT and after postannealing for half an hour at 150, 200, and 250 °C, respectively. The Fe thickness corresponds to the nominal deposited film thickness, which is used throughout this text. The data points basically lie on two different curves: The sample annealed at 200 °C shows full remanence down to a thickness of ~ 5.5 ML and the remanence disappears for smaller thicknesses within a monolayer. All the other

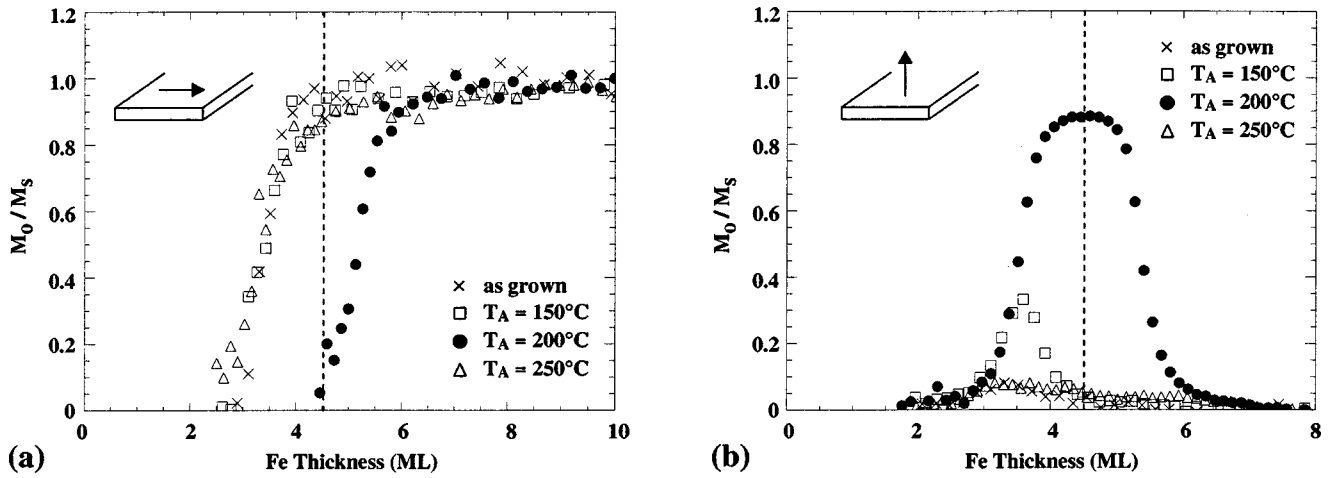


FIG. 1. Remanent magnetization measured at RT for Fe films grown at RT on Ag(001) and annealed at various temperatures T_A : (a) longitudinal Kerr signal, (b) polar Kerr signal.

samples show full remanence down to ~ 3.5 ML and again the remanence fully vanishes for smaller thicknesses within a monolayer.

In Fig. 1(b), the remanent magnetization is plotted for the different samples in polar geometry, which is sensitive to out-of-plane magnetization. Again, the sample annealed at 200°C differs substantially from all the others: It shows almost full remanence around 4.5 ML, whereas all the other samples show no or only very little out-of-plane remanence. This means that for Fe films with a thickness of about 4.5 ML, which is highlighted in the plots by a dashed vertical line, we observe a spin reorientation from in-plane to out-of-plane upon annealing at 200°C . A second spin reorientation back to in-plane is observed after annealing at 250°C . Because the Curie temperature T_C is falling below RT at 2–3 ML,⁸ neither in-plane nor out-of-plane remanent magnetization is observed below this thickness.

We perform structural analysis of 4.5-ML-thick Fe films by LEED and STM. By LEED, we observe a sharpening of

the spots upon annealing, in agreement with previous reports,^{8,10} but it is difficult to get quantitative information about the morphology. Therefore we characterize the surfaces also by STM (Fig. 2). To quantify the morphology, we determine the *vertical roughness* σ as the average deviation from the mean flat surface, $\sigma = \langle |\delta z| \rangle$, and the *lateral roughness* L as the average lateral size of terraces. After growth at RT [Fig. 2(a)], we observe an irregular arrangement of growth hillocks, in accordance with measurements of thicker Fe films on Ag(001).¹⁵ The vertical roughness amounts to $\sigma_{RT} = 0.105$ nm. The lateral roughness is too small to be determined from the STM image, but it must be on the order of the Fe lattice constant, hence $L_{RT} \approx 0.3$ nm. The upper part of the figure displays the histogram of the image, which has a Gaussian shape. There cannot be detected any peaks corresponding to Fe terraces. After annealing at 150°C , the STM image (not shown) looks very similar to Fig. 2(a) and also the vertical roughness is only slightly reduced to $\sigma_{150^\circ\text{C}} = 0.103$ nm. Evident morphology changes appear after

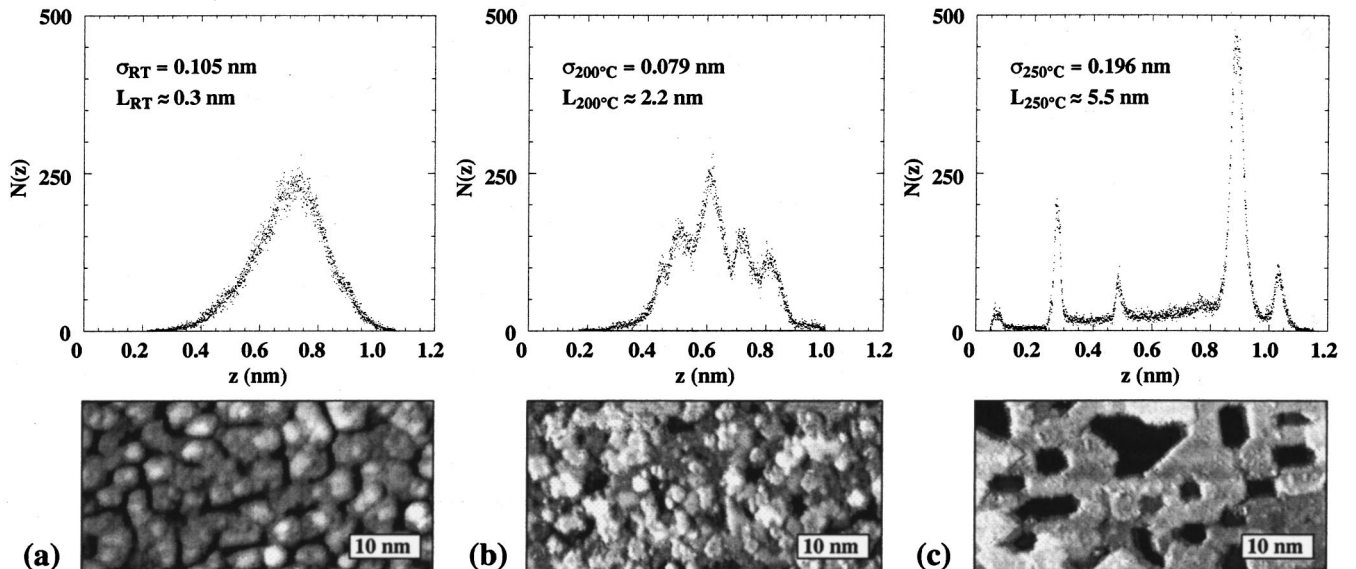


FIG. 2. STM images ($50 \times 25 \text{ nm}^2$) and the corresponding histograms of a 4.5-ML-thick Fe film grown at RT on Ag(001) and annealed at various temperatures: (a) as grown, (b) after post annealing at 200°C , (c) after post annealing at 250°C . The vertical roughness σ and the lateral roughness L extracted from the images are displayed.

a further annealing at 200 °C: The hillocks have transformed into islands with single atomic steps [Fig. 2(b)]. This can be concluded also from the clear peaks in the histogram. The distances between the histogram peaks correspond to a combination of Fe steps on different Ag terraces. $\sigma_{200\text{ °C}}$ is further reduced and amounts to 0.079 nm. Because the lateral period is now determined by terraces rather than growth hillocks, L can be evaluated by calculating the position of the first maximum R in the radial height-height correlation function. For morphologies with locally only two levels, L corresponds to half the period in the height-height correlation function, hence $L \approx R/2$. With this statistical method we obtain $L_{200\text{ °C}} \approx 2.2$ nm. Further annealing of the Fe film at 250 °C again changes the surface drastically [Fig. 2(c)]: The terraces are much larger, $L_{250\text{ °C}} \approx 5.5$ nm, and holes with a depth of 4–5 ML between them expose the Ag substrate. These holes cover 25% of the surface. The appearance of the holes increases the vertical roughness to $\sigma_{250\text{ °C}} = 0.196$ nm. This value is probably underestimated because the STM tip cannot reach the bottom of the smaller holes. The histogram exhibits much sharper peaks corresponding to Ag (left three peaks) and Fe step heights.

The morphological changes detected by STM coincide with the chemical analysis by XPS measurements: The ratio of the Ag 3d peak area to the Fe 2p peak area is constant or even slightly reduced after postannealing at 150 and 200 °C, in correspondence with a reduced surface roughness, and it is increased by 15% after postannealing at 250 °C, which is consistent with the appearance of holes exposing the Ag substrate and covering 25% of the area.

The appearance of these holes upon annealing at high enough temperatures can be explained by the twice as large surface free energy of Fe(001) compared to the value of Ag(001).¹⁸ If the diffusion length is large enough and the coverage is low, the system can minimize its energy by dewetting the substrate and forming an Ag surface. The influence of the surface free energy on the growth mode of Fe on Ag(001) has been discussed in more detail in Ref. 15.

Roughness changes both the magnetocrystalline and the magnetic dipolar anisotropy. Using Néel's model,¹⁹ several authors investigated the additional symmetry breaking at steps giving rise to a magnetic step anisotropy.^{20–22} The Néel model fails to predict a surface anisotropy for a bcc(001) surface in first-nearest-neighbor approximation.²³ Because second-nearest neighbors are only 15% more distant than first-nearest neighbors in a bcc crystal, they must not be neglected and the Néel model predicts indeed an out-of-plane surface anisotropy in this approximation. Second-nearest neighbors have simple cubic coordination. For this symmetry, Bruno calculated in Ref. 20 a decrease of the out-of-plane surface anisotropy by 50% for each step atom. This reduction needs to be multiplied by the percentage of step atoms at the surface, which can be counted to be $4\sigma/L$.²⁰ This results in a decrease of the surface anisotropy by $\Delta K_s/K_s = -2\sigma/L$.

Using the experimentally acquired roughness parameters, we calculate a decrease of K_s by more than 50% for the sample prepared at RT, whereas after annealing at 200 and 250 °C, the reduction amounts only to 7.2 and 7.1%, respectively. Consequently, the reduction of K_s induced by roughness can explain the spin reorientation from in-plane to out-

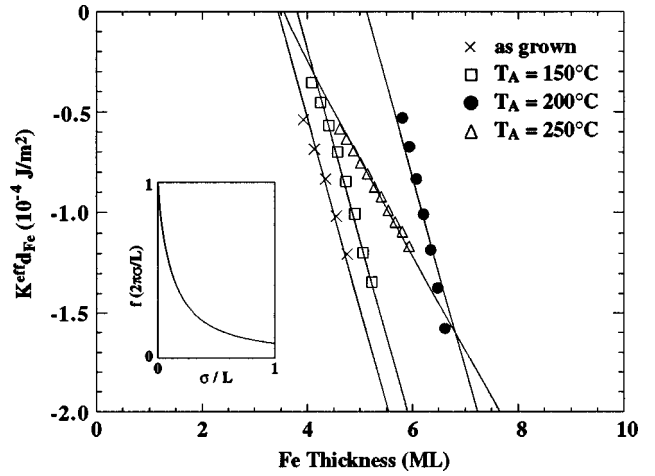


FIG. 3. The product of the effective anisotropy and the Fe thickness is plotted as a function of the Fe thickness for Fe films grown at RT and annealed at various temperatures T_A . The solid lines are a simultaneous fit of the data to Eq. (2). The inset shows the graph of the function f used in Eq. (2).

of-plane upon annealing the Fe film at 200 °C, but it cannot account for the second spin reorientation back to in-plane after further annealing at 250 °C. This second spin reorientation is caused by the reduction of the interface area by 25% due to the holes which results in a reduction of the surface energy contribution by the same amount. Additionally, we have to take into account the effect of roughness on the magnetic dipolar anisotropy. Bruno calculated in Ref. 24 that roughness also gives rise to a purely dipolar surface anisotropy. If the vertical roughness σ is dependent on thickness, this anisotropy reduces the absolute value of K_v . A strong thickness dependence of the vertical roughness is expected for the sample annealed at 250 °C, because the hole depth and thereby also σ linearly increase with thickness.

Summarizing the effect of roughness on magnetic anisotropy, we can rewrite Eq. (1), taking into account the effect of roughness at one of the interfaces:

$$K^{eff} = -\frac{1}{2}\mu_0 M_s^2 + \frac{2K_s}{d} \left(1 - 2\alpha \frac{\sigma}{L} \right) + \frac{3}{8}\mu_0 M_s^2 \frac{\sigma}{d} \left[1 - f\left(2\pi \frac{\sigma}{L} \right) \right]. \quad (2)$$

The last term is the roughness induced dipolar surface anisotropy calculated in Ref. 24. The graph of the function f which is given explicitly in Ref. 24 is plotted in the inset of Fig. 3. The parameter α denotes the reduction of the surface anisotropy per step atom, which is predicted to be 50% in the Néel model.

To quantify our observations, we determine the effective anisotropy of the different samples. Elementary electromagnetic considerations show that the area enclosed between polar and longitudinal hysteresis loops is proportional to K^{eff} .¹ In the simple case where we measure square longitudinal loops, the polar saturation field, the so-called anisotropy field H_A , is proportional to the effective anisotropy and $K^{eff} = -\mu_0 H_A M_s / 2$. We use the bulk value for the saturation magnetization of Fe, $M_s = 1.714 \times 10^6$ A/m.

In Fig. 3, the product of the effective anisotropy and the Fe thickness is plotted as a function of the Fe thickness for the differently prepared samples. The solid lines are the result of a simultaneous fit of the data to Eq. (2). The roughness parameters determined from the STM images are used and the vertical roughness of the sample annealed at 250 °C is assumed to be linearly dependent on thickness, $\sigma_{250\text{ °C}}(d) = \sigma_0 d$, in agreement with the linear increase of the hole depth with thickness. For the sample annealed at 250 °C, the decrease of the surface energy contribution by 25% due to the reduction of the interface area is taken into account.

The best fit is achieved with the following parameters: $\alpha = 64\%$, $M_s = 1.03 \times 10^6$ A/m, $K_s = 0.27$ mJ/m², $\sigma_{250\text{ °C}}(d) = 0.81d$, and $L_{150\text{ °C}} = 1.16L_{RT}$. The reduction factor α is indeed fitted very close to 50% which is what is expected from the Néel model in second-nearest-neighbor approximation. The other parameters are in reasonable agreement with the expected values: $L_{150\text{ °C}}$ is only slightly larger than L_{RT} , and $\sigma_{250\text{ °C}}$ has a thickness dependence close to $\sigma(d) = 0.5d$ valid for a flat film perturbed by holes making up 25% of the area. The values of M_s and K_s are fitted rather small (see, e.g., Ref. 1), but similarly small values have been measured on 3-ML-thick Fe films on Ag(001) substrates with a miscut of 1.8° by Heinrich *et al.*¹⁶ Larger values of K_s have been obtained by the same authors on substrates with a

lower miscut.²⁵ Thus the roughness of the bottom interface might account for the reduced surface anisotropy. The lower interface is not considered in the above analysis since it is equal for all samples and can therefore not explain the observed spin reorientations.

In conclusion, we have shown by a combined STM and MOKE study that the magnetic anisotropy of thin Fe films can considerably be altered by morphology. Spin reorientations induced solely by morphology changes are observed which can explain some of the discrepancies reported in literature concerning the magnetic phase diagram of Fe/Ag(001). The results are explained by a change of both the magnetic dipolar and the magnetocrystalline anisotropy due to roughness. The change of the magnetocrystalline anisotropy cannot be predicted within the Néel model in first-nearest-neighbor approximation, even if strain is taken into account. In second-nearest-neighbor approximation, which may be important in a bcc crystal with small strain, the Néel model predicts a reduction of the out-of-plane surface anisotropy which is in close agreement with our experiments.

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²³The symmetry breaking due to strain gives rise to an out-of-plane surface anisotropy proportional to the misfit (Ref. 22), which is very small in the well lattice-matched Fe/Ag(001) system. Strain effects do not change our conclusions and are therefore neglected.

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