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Effect of substrate inclination on the magnetic anisotropy of ultrathin Fe films grown on $Al_2O_3(0001)$

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We have investigated the effect of substrate inclination on the in-plane magnetic anisotropy of ultrathin Fe films grown on $Al_2O_3(0001)$. For Fe films grown on flat Al_2O_3 substrates, no preferred direction of magnetization exists in the film plane due to the three equivalent epitaxial orientations of Fe(110) that form. However, for Fe grown on an inclined Al_2O_3 substrate, a uniaxial anisotropy appears that is parallel to the step edges. The anisotropy increases in magnitude with decreasing Fe thickness and growth temperature, and as the surface morphology changes from being rough to being smooth. We attribute the uniaxial anisotropy to the effective demagnetization field caused by the surface corrugation. [DOI: 10.1063/1.1850075]

Controlling the magnetic anisotropy of ultrathin films has been an area of active pursuit. The magnetic anisotropy in ultrathin films predominantly originates from two different effects. One is the spin-orbit interaction, which gives rise to the magnetocrystalline anisotropy. At a surface or interface this interaction can yield enhanced anisotropies, including perpendicular magnetic anisotropy[1,2] and step-induced magnetic anisotropies.[3–5] A characteristic spin-orbit interaction occurs at the surface/interface, compared to that in the bulk, due to the reduction of crystalline symmetry and/or to the magnetoelastic effects. In order to explore further, it is necessary to understand the surface electronic structure. However, in real system, disorder, such as surface roughness, complicates the analysis. Another origin of magnetic anisotropy is the long-range dipole interaction, which is controllable by altering the shape of the film. Hence, it is known as the shape anisotropy. In the present study we focus on tailoring the dipole interactions to control the anisotropy and the easy-axis orientation. We utilize the surface corrugation created by regularly aligned steps on an inclined substrate as a means to alter the film morphology and the resultant magnetic dipole interactions.[6–11] We choose Fe on an inclined $Al_2O_3(0001)$ substrate, since Fe itself has only a small magnetic anisotropy, and $Al_2O_3(0001)$ forms an atomically flat surface with regularly aligned, straight steps. These two factors make our system suitable to investigate the magnetic anisotropy in ultrathin films. We report on the measurements of the magnetic anisotropy induced by substrate inclination, and we clarify the origin of the induced anisotropy. In the following, we consider only the dipole interactions as an origin of magnetic anisotropy since the spin-orbit interactions at the surface should be canceled out due to the epitaxial Fe orientations on $Al_2O_3(0001)$, as mentioned below.

Ultrathin Fe films were prepared by molecular-beam epitaxy (MBE) using a VG-80M MBE system. The pressure before and during the deposition was typically $<4\times10^{-9}$ and 5×10^{-8} Pa, respectively. The nominal thickness of Fe was varied from 10 to 25 monolayers (ML) and the growth temperature was varied in the range of 323–773 K. The surface structure of the Fe films was investigated *in situ* by means of noncontact atomic force microscopy (NC-AFM) and reflection high-energy electron diffraction (RHEED). The magnetic anisotropy energy is estimated from the magnetization curves and from the angular dependence of the resonance field. Each measurement was performed *ex situ* by means of vibrating-sample magnetometry (VSM) and ferromagnetic resonance (FMR) operated with *X*-band (9.4 GHz)

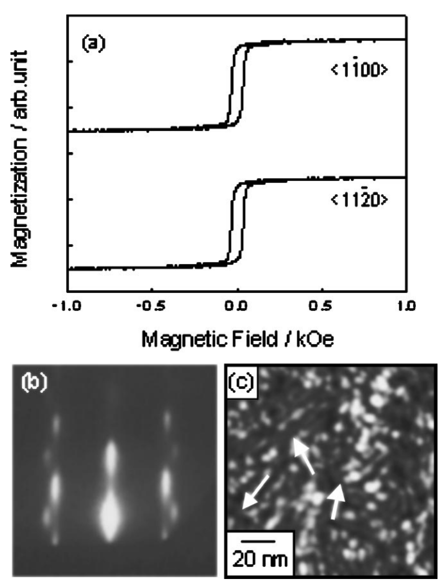

FIG. 1. (a) Magnetization curves, (b) RHEED pattern, and (c) AFM image of a 20-ML Fe film grown on the flat substrate. The applied-field direction for the magnetization measurements is along the $\langle 1\bar{1}00\rangle$ and $\langle 11\bar{2}0\rangle$ of the $Al_2O_3(0001)$ substrate in the upper and lower curves, respectively.

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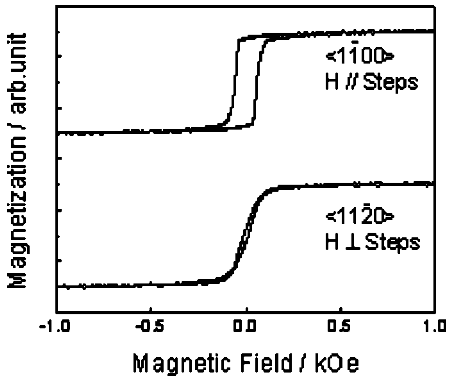

FIG. 2. Magnetization curves of a 20-ML Fe film grown on the inclined substrate. The applied-field direction is along the $\langle 1\bar{1}00\rangle$ and $\langle 11\bar{2}0\rangle$ of $Al_2O_3(0001)$ in the upper and lower curves, respectively.

microwaves. To avoid oxidation, a 10-nm-thick Au capping layer was deposited at room temperature. We confirmed the lack of oxidation even for 5-ML Fe films indirectly from the fact that the magnetization curves at 10 K showed no bias after cooling in a 10-kOe magnetic field. Two types of α-$Al_2O_3(0001)$ substrates were used. One is nominally flat, having 0.216-nm-high steps and 129.5-nm-wide terraces on average, as reported previously.[7] The other substrate is inclined at an angle of 4° in the $\langle 11\bar{2}0\rangle$ direction. Such inclined substrates have straight steps, and several atomic steps can bunch to form, by suitable thermal treatment, average step heights of 6.06 nm and average terrace widths of 65.5 nm.[7]

Magnetization curves for a 20-ML Fe film are shown in Fig. 1(a) for Fe on the flat substrate. The curves are identical for different applied-field directions, meaning that no preferred magnetization direction exists in the film plane. The isotropic behavior of the magnetization is explained by the epitaxial growth of Fe on $Al_2O_3(0001)$. A RHEED pattern and an AFM image of an Fe film on the flat substrate are shown in Figs. 1(b) and 1(c), respectively. The RHEED pattern shows that the Fe grows epitaxially, forming three in-plane bcc-Fe(110) orientations on the $Al_2O_3(0001)$. The AFM image identifies three kinds of ellipsoidal Fe(110) epitaxial islands, as indicated by the arrows in Fig. 1(c). Given the epitaxial relationship, the in-plane symmetry of the Fe is sixfold, and a uniaxial magnetic anisotropy cannot exist due to the high symmetry.[12]

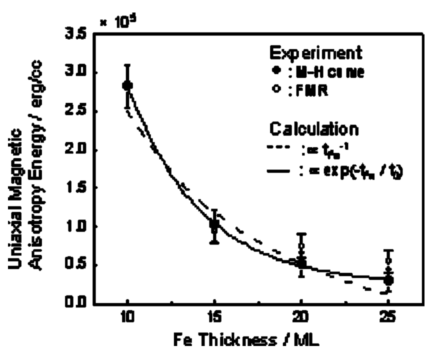

FIG. 3. The Fe thickness dependence of the uniaxial magnetic anisotropy. The open and closed circles represent the experimental values estimated from the magnetization curves and ferromagnetic resonance field, respectively. The dashed and solid lines represent the calculated values, as described in the text.

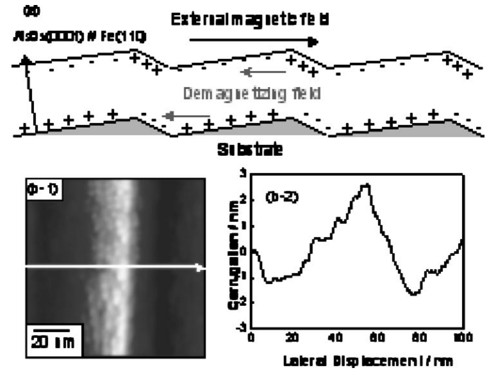

FIG. 4. (a) Schematic representation of the surface structure of an Fe film grown on the inclined substrate and the magnetic charge distribution at the corrugated surface when the magnetization is saturated perpendicular to the steps. The lower panel shows the surface structure of a 10-ML Fe film grown at 323 K on the inclined substrate, where (b) is the AFM image and (c) is a cross-sectional plot of the corrugation corresponding to the arrow in (b).

Fe grown on the inclined substrate, however, does possess a uniaxial magnetic anisotropy that is parallel to step edges, as shown in Fig. 2. Since the uniaxial anisotropy is absent for Fe on the flat substrate, its appearance on the stepped substrate is attributed to the substrate inclination. In ultrathin films, the induced anisotropy could have different origins, including the spin-orbit interaction and/or dipole interactions, as mentioned above. In order to clarify the origin of the observed anisotropy, we investigated its dependence on Fe thickness, growth temperature, and surface morphology. The change in the magnitude of the uniaxial magnetic anisotropy with Fe thickness is shown in Fig. 3. The anisotropy increases with decreasing Fe thickness, which means that it should originate from a surface/interface effect. However, the well-known phenomenological relationship[13] $K_u = K_{uV} + K_{uS}/t_{Fe}$, does not describe the experimental results, as shown by the dashed lined in Fig. 3. It was reported in Ref. 14 that the magnetic anisotropy induced by surface corrugation should increase exponentially with decreasing magnetic layer thickness.[14] The experimental results are reproduced by an exponential dependence, as shown by the solid line in Fig. 3. In such a case, the uniaxial anisotropy is explained by the film structure, as schematically represented in Fig. 4(a).[14] According to this viewpoint, the uniaxial magnetic anisotropy originates from the effective demagnetizing field at the magnetic film surface/interface. Thus, in order for the uniaxial anisotropy to appear, the magnetic film has to grow conformally to the step structure of the inclined substrate. At

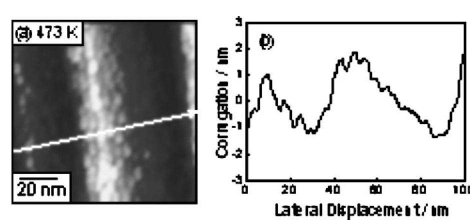

FIG. 5. Surface structure of a 10-ML Fe film grown at 473 K on the inclined substrate.

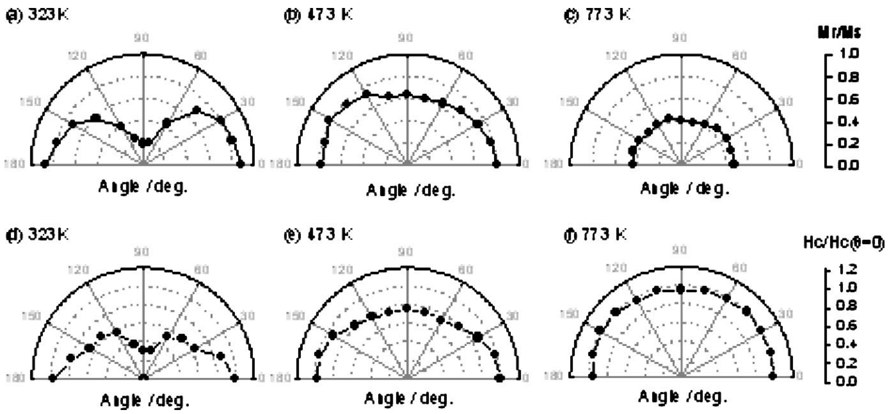

FIG. 6. The dependence on the applied-field direction of the remanence ratio (M_r/M_s) appears in (a)–(c) and the coercivity (H_c) in (d)–(f) for 20-ML Fe films grown on the inclined substrate. The growth temperature is 323 K in (a) and (d), 473 K in (b) and (e), and 773 K in (c) and (f). The angle of 0° corresponds to the direction parallel to the step edges.

low growth temperature, the Fe films do grow in such a fashion. [see Fig. 4(b)]. Moreover, as the surface morphology becomes rough, the effective demagnetizing field decreases; consequently, the uniaxial magnetic anisotropy should decrease. Thus, we can verify the above by investigating the dependence of the anisotropy on the surface morphology, which is altered by changing the growth temperature. Figure 5 shows the roughened surface structure of Fe films grown at high temperature, where Fe particles become visible. As mentioned above, the roughening should cause a reduction in the magnitude of the anisotropy. Thus, the growth temperature dependence of the uniaxial magnetic anisotropy clarifies the validity of the above discussion.

Figure 6 shows the dependences of the remanence ratio M_r/M_s and coercivity H_c on the applied-field direction for 20-ML Fe films grown at various temperatures. At low growth temperature, M_r/M_s and H_c exhibit the twofold symmetry of the uniaxial magnetic anisotropy. But as the growth temperature increases, both M_r/M_s and H_c become more isotropic. Thus, the magnitude of the uniaxial anisotropy decreases as the growth temperature increases and the surface morphology roughens. These observations are consistent with the expectation based on the above-mentioned model. Thus, we conclude that the uniaxial magnetic anisotropy induced by the substrate inclination is derived from the effective demagnetizing field caused by the surface corrugation.

In summary, we have investigated the effect of substrate inclination on the in-plane magnetic anisotropy of ultrathin Fe films. Fe films grown on flat Al_2O_3 substrates have no preferred direction of magnetization in the film plane due to the three equivalent Fe(110) epitaxial orientations that form and that result in a high in-plane symmetry. Whereas, for Fe grown on the inclined Al_2O_3 substrate, a uniaxial anisotropy appears, which is parallel to the step edges. The induced anisotropy can be understood to originate from the effective demagnetizing field caused by the surface corrugation.

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