Symmetry-Induced Magnetic Anisotropy in Fe Films Grown on Stepped Ag(001)

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Symmetry breaking at atomic steps of ultrathin Fe films grown on stepped Ag(001) substrates creates a uniaxial magnetic anisotropy with the easy axis parallel to the step edges. The relation between this induced anisotropy and the step density is systematically investigated by magneto-optically scanning films grown on a curved substrate with a vicinal angle range between 0° and 10°. We find that (a) the uniaxial anisotropy depends quadratically on the step density, and (b) the spin-reorientation transition occurs at a larger thickness for the stepped than for the flat film. [S0031-9007(96)01218-5]

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Magnetocrystalline anisotropy is due to the spin-orbit interaction [1], thus it must obey the symmetry of the lattice. In a lattice with cubic symmetry, quadratic uniaxial anisotropy can exist only at the surface, where the translational symmetry is broken. For some systems [Fe/Ag(100), for example], this surface anisotropy is even stronger than the bulk shape anisotropy and can align the magnetization perpendicular to the film surface below a critical thickness [2–4]. Within the film surface it is known that no uniaxial anisotropy can exist if the surface normal is an n-fold rotation axis with $n > 2$ [5]. In an effort to understand how symmetry breaking induces the uniaxial magnetic anisotropy, a few groups [6–9] have performed experiments on magnetic thin films grown on stepped (100) substrates. Atomic steps on a (100) surface break the four-fold rotational symmetry of the film surface and therefore should induce a uniaxial anisotropy within the film plane. This type of step-induced uniaxial anisotropy has been observed in Ni/Fe and Fe films on Ag(3 1 100) [6], Co films on stepped Cu(100) [7,8], and Fe films on stepped W(100) [9]. In these studies, the step density (determined by the vicinal angle from the [100] direction) is fixed, so the relation between induced anisotropy and step density was not explored. In this Letter, we report a systematic study of this induced anisotropy vs step density for the Fe/Ag(001) system. Fe/Ag(001) has a perpendicular surface anisotropy. The competition between the surface and shape anisotropies results in a spin-reorientation transition (SRT) at an Fe film thickness of ~6 ML (monolayers) [10,11]. We show that the SRT shifts to an ~10% larger critical thickness due to the step-induced anisotropy contribution to the energy competition.

Our experiments were performed using two 10 mm diameter Ag(001) substrates. On the *fixed-angle* substrate, steps were introduced onto half of the surface by polishing at ~6° from the [001] orientation. On the *curved* substrate, half was polished into a curved surface so that the angle between the surface normal and the [001] direction varied continuously from 0° to ~10°. Both crystals were polished under the same conditions: mechanically down to a 0.25 μm diamond-paste finish, followed by chemical polishing [12]. The step edges are parallel to the [110] of the Ag(001) surface so that the Fe(001) overlayer has step edges parallel to its [100]. The substrates are cleaned *in situ* by cycles of Ar^+ sputtering at 2–5 keV and annealing at 500 °C. All Fe films are grown epitaxially on the Ag substrate at room temperature with the growth chamber pressure $< 5 \times 10^{-10}$ Torr. After growth, the films are annealed at 150 °C for ~30 min. Our RHEED and LEED results agree with those reported in Ref. [11]. The RHEED intensity for the growth of Fe on vicinal Ag exhibits regular oscillations after an initial transient period due to the tetragonal difference between the bcc and fcc structures [6]. The magnetic properties of the films are measured *in situ* by the surface magneto-optic Kerr effect (SMOKE) using a He-Ne laser (632.8 nm; beam diameter ≈0.2 mm).

To investigate the step-induced, in-plane magnetic anisotropy, an Fe film of ~25 ML was grown onto the fixed-angle substrate. At this thickness, the magnetization is fully in-plane. Figure 1 shows the longitudinal SMOKE hysteresis loops measured on the stepped surface with the magnetic field H parallel and perpendicular to the step edges, respectively. The shapes of the loops show that the atomic steps induce an in-plane, uniaxial magnetic anisotropy with easy axis parallel to the step edges [8,9].

To explore the relation between this anisotropy and the step density, a 25 ML Fe film was grown onto the curved substrate and investigated via SMOKE. As the laser beam scans across the substrate to measure the hysteresis loop, its reflection angle also simultaneously determines the vicinal angle within the laser spot. Figure 2 shows representative longitudinal SMOKE loops at four vicinal angles for H along the hard axis. The split loops are characterized by a shift field (H_s) which is proportional to the step-induced anisotropy [8]. Figure 3 shows the relation between H_s and α. The error bars in the figure account for the fact that the laser spot covers a finite range of step densities.

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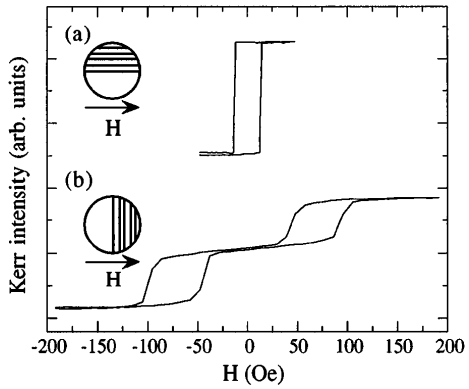

FIG. 1. Longitudinal hysteresis loops of a 25 ML Fe film grown on a stepped Ag(001) substrate (6° vicinal angle). (a) H applied parallel to the step edges, (b) H applied perpendicular to the step edges.

The linear dependence of H_s on α in the log-log plot [Fig. 3(b)] indicates a power-law relation. Fitting $H_s \sim \alpha^n$ yields an exponent $n = 1.97 \pm 0.07$. Thus the step-induced uniaxial anisotropy depends quadratically on step density.

To better understand the step-induced anisotropy we considered a phenomenological model based on the Néel pair-bonding mechanism. In Néel's model, the magnetic anisotropy is determined by the spin-orbit interaction through the nearest-neighbor electronic hybridization; therefore, it should be accurate for localized electrons. In transition-metal magnets, the magnetic $3d$ electrons are not fully localized, so the magnetic anisotropy should be derived with realistic energy-band calculation [1]. If we disregard the numerical values of the anisotropy constants from the Néel model but keep only its functional form, which reflects the symmetry of the lattice, the energy density of a stepped [001] bcc film of thickness d is

$$E = -\frac{K_s}{d}\,u_\zeta^2 + 2\pi M^2 u_z^2 + K_V(u_\xi^2 u_\eta^2 + u_\eta^2 u_\zeta^2 + u_\zeta^2 u_\xi^2) - \frac{K_{sp}}{dL}\,u_\eta u_\zeta\,, \qquad (1)$$

where $\mathbf{u}$ is the unit vector of the magnetization M, and ξ, η, and ζ are the [100], [010], and [001] axes, respectively. The steps are on the (001) surface with edges parallel to the [100]. The average terrace length of the steps is L. The normal direction (z axis) of the stepped surface makes an angle α to the [001] axis, so that $\alpha \approx a/L$ is proportional to the step density (a is the layer spacing in the [001]). The last term in Eq. (1) is the anisotropy generated by the atomic steps in a [001] bcc surface [13]. The effect of strains has been ignored in this model. It should be kept in mind, however, that the lattice distortion could sometimes generate an appreciable amount of volume-type uniaxial anisotropy and also could make the K_V significantly different from its bulk value [14]. Therefore the K_S and K_V in Eq. (1) should be viewed as operationally defined surface and volume anisotropies and should in general be thickness dependent. After a coordinate transformation from the crystal $\xi\eta\zeta$ frame to the film xyz frame with x and y axes in the plane of the film and parallel and perpendicular to the step edges, respectively, the energy density of Eq. (1) transforms to order α^2 into

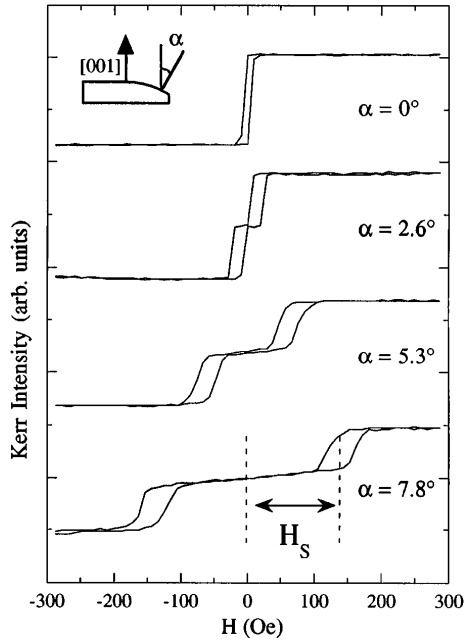

FIG. 2. Longitudinal hysteresis loops of a 25 ML Fe film grown on a curved Ag(001) substrate. The vicinal angle α, ranging from 0° to 10°, is proportional to the step density. H is applied perpendicular to the step edges. The split field H_s is proportional the step-induced, uniaxial anisotropy.

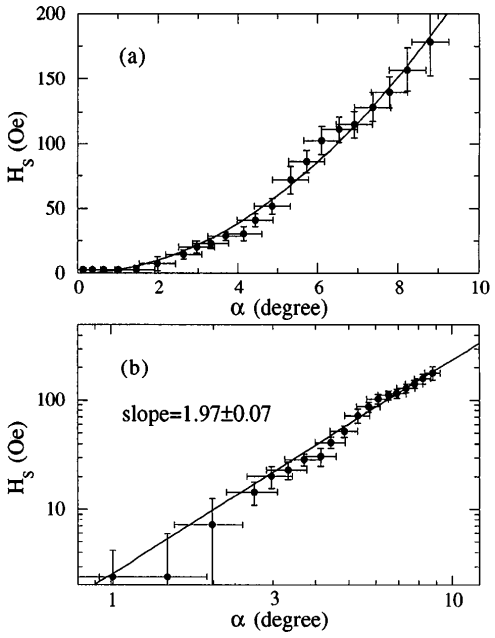

FIG. 3. (a) The split field H_s vs vicinal angle α. (b) Log-log plot of H_s vs α. The solid lines are the result of a power-law fitting of $H_s \sim \alpha^n$, yielding $n = 1.97 \pm 0.07$. Thus the step-induced, uniaxial anisotropy depends quadratically on the step density.

$$E = \left[-\frac{K_s + (K_{sp}/a - K_s)\alpha^2}{d} + 2\pi M^2 \right] u_z^2 + \frac{(K_{sp}/a - K_s)\alpha^2}{d} u_y^2 - \frac{(K_{sp}/a - 2K_s)\alpha}{d} u_y u_z + K_V[u_x^2 u_y^2 + u_y^2 u_z^2 + u_z^2 u_x^2 - 2\alpha u_y u_z (u_y^2 - u_z^2) + \alpha^2 (u_y^4 + u_z^4 - 6u_y^2 u_z^2)]. \quad (2)$$

We first consider the situation of thick films where the strong shape anisotropy forces the magnetization in the plane of the film ($u_z = 0$). The energy density in this case is

$$E = \frac{(K_{sp}/a - K_S)\alpha^2}{d} u_y^2 + K_V(u_x^2 u_y^2 + \alpha^2 u_y^4), \quad (3)$$

which has in-plane, uniaxial anisotropy $(K_{sp}/a - K_S)\alpha^2/d$. The quadratic dependence of this anisotropy on the step density is consistent with our experimental observations. For $K_{sp}/a > K_S$, the hard axis will be along the y axis, i.e., perpendicular to the step edges. For H applied along the hard axis, it is easy to show [8] that the hysteresis loop will split into two loops with an offset field of $H_S = 2(K_{sp}/a - K_S)\alpha^2/Md$. A numerical estimate of K_{sp} can be obtained by fitting the data in Fig. 3(a) with this formula. Taking the values $d = 25$ ML, $a = 1.435$ Å, $M = 1.71 \times 10^3$ G, and $K_S = 1.6$ erg/cm^2 [15], the fit yields $K_{sp} \approx 5.73 \times 10^{-8}$ erg/cm.

We now turn our attention to the thin-film regime. For a flat surface ($\alpha = 0$), the quadratic energy terms in Eq. (2) reduce to the familiar form $(-K_S/d + 2\pi M^2)u_z^2$, which gives rise to a sharp spin-reorientation transition between $u_z = 1$ and $u_z = 0$ at $d_R = K_S/2\pi M^2 \approx 6$ ML. The formation of stripe domains and the effect of the volume anisotropy usually make the transition less abrupt [16,17]. Under certain circumstances, the volume anisotropy can even alter the nature of the SRT from a first-order switching to a continuous rotation [14]. For a stepped surface, it is easy to show that the competition among the three quadratic terms in the anisotropy energy will result in a continuous rotation of the magnetization toward the film plane in the y-z plane upon increasing film thickness. At a critical thickness d_R^*, the magnetization will switch via a first-order transition. The z components of the magnetization and d_R^* are

$$u_z^2 = \frac{1}{2} - \frac{2\pi M^2 d - K_S - 2(K_{sp}/a - K_S)\alpha^2}{2\sqrt{(K_{sp}/a - 2K_S)^2\alpha^2 + [2\pi M^2 d - K_S - 2(K_{sp}/a - K_S)\alpha^2]^2}}$$

$$\text{and} \quad d_R^* = \frac{K_{sp}^2/a^2}{8\pi M^2 (K_{sp}/a - K_S)} + \frac{(K_{sp}/a - K_S)\alpha^2}{2\pi M^2}. \quad (4)$$

For $\alpha = 6°$ (corresponding to our fixed-angle substrate), Eq. (4) gives $d_R^* \approx 6.4$ ML and $u_z \approx 0.5$ (at $d = d_R^*$). Thus a larger SRT thickness or, equivalently, a higher SRT temperature [11] is expected for Fe films grown on the fixed angle vs the flat substrate. To avoid any thickness error in our experiment, the SRT as a function of temperature was first studied by growing a uniform 5.5 ML Fe film on the fixed-angle substrate. In this way, the films on the flat and stepped surfaces of the substrate were grown at the same time, so that they should have exactly the same thickness. Figure 4 shows the polar SMOKE loops at three temperatures. The polar remanence is always higher on the stepped surface than on the flat surface at any given temperature, confirming that the SRT on the stepped surface has the higher transition temperature. To make a quantitative study, the SRT as a function of film thickness was investigated by growing wedged Fe films (slope ~1 ML/mm) on the fixed-angle substrate. The polar and longitudinal remanence on the flat and stepped surfaces were measured at 150 K and are plotted in Fig. 5. For the stepped surface, the longitudinal SMOKE loop was taken along the in-plane easy axis. As expected, Fig. 5 shows that the SRT thickness has shifted from ~6 on the flat surface to ~6.8 ML on the stepped surface. The discrepancy between the calculated 6.4 and the observed 6.8 ML values could be due to several possible reasons. Experimentally, the error in the zero-thickness registration point in our wedged samples may cause a shift in the film thickness (estimated to be within ~0.2 ML). Theoretically, the d_R^* was calculated using the value of $K_{sp} = 5.73 \times 10^{-8}$ erg/cm, which was derived from a 25 ML Fe film. If we allow the K_{sp} to vary as a function of film thickness [18], a value of $K_{sp} = 6.7 \times 10^{-8}$ erg/cm will give the observed 6.8 ML SRT thickness. It is also interesting to note that the polar signal in the SRT region on the stepped surface decreases slower than on the flat surface with increasing film thickness. This may reflect the continuous rotation of the magnetization in the y-z plane on the stepped surface.

The quadratic relation between the step-induced anisotropy and the step density originates from the $\sim u_\eta u_\zeta$ term in Eq. (1). In general, the steps in a (001) surface of a cubic-symmetry lattice should induce both $\sim u_\eta u_\zeta$ and $\sim u_\xi^2$ anisotropies. The $\sim u_\xi^2$ term will result in a linear relation between the induced anisotropy and the step density. For a bcc lattice, only the $\sim u_\eta u_\zeta$ term exists. For an fcc lattice, the steps could induce

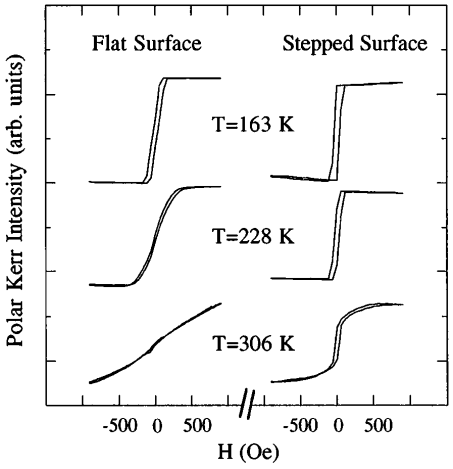

FIG. 4. Polar hysteresis loops at different temperatures for a 5.5 ML Fe film grown onto the fixed-angle substrate (6° vicinal angle). The spin-reorientation transition occurs at a higher temperature on the stepped surface rather than on the flat surface.

both $\sim u_\eta u_\zeta$ and $\sim u_\xi^2$ anisotropies, so a superposition of linear and quadratic dependences of the induced anisotropy on the step density is expected. In a real experimental system, strain and defects at the steps could also give some residual u_ξ^2 anisotropy. Fitting our data by including both a linear and a quadratic term, we find that the linear term comprises only 2.5%, 0.5%, and 0.3% of the overall anisotropy at $\alpha = 1°$, 5°, and 10°, respectively. Hence the dependence of the step-induced anisotropy on the step density in our case is nearly purely quadratic.

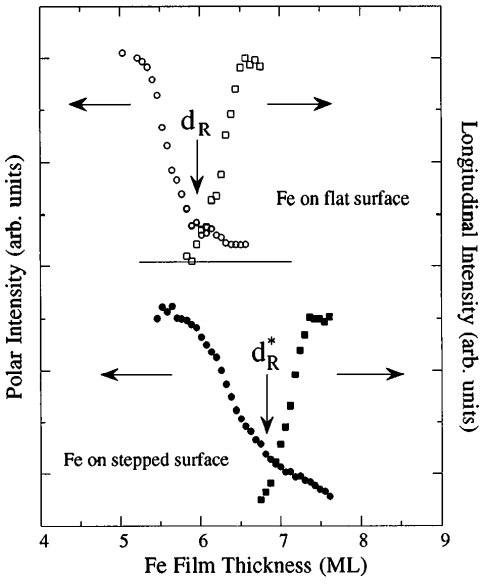

FIG. 5. Magnetic remanence of Fe films grown on the fixed-angle substrate (6° vicinal angle). The measurements are made at 150 K. The Fe films on the stepped surface exhibit a higher spin-reorientation transition thickness than on the flat surface.

In conclusion, stepped Fe/Ag(100) possesses a step-induced, in-plane, uniaxial anisotropy with easy axis parallel to the step edge. The strength of this anisotropy varies quadratically with step density. This result is explained by a Néel-type model. The steps stabilize the perpendicular magnetization to a larger film thickness and a higher temperature than is observed on flat films. Based on our simple model, this behavior is consistent with the observed in-plane, uniaxial anisotropy.

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