

Temperature dependent magnetic surface anisotropy in ultrathin Fe films

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The temperature dependence of the uniaxial surface anisotropy, $K_u(T)$, in ultrathin Fe films (4–10 atomic layers) grown on Cu(100) is determined by comparing the dipole and anisotropy energies at the spin reorientation temperature for films of varying thickness. It is observed that the uniaxial anisotropy has a weak temperature dependence compared to the bulk constants for Fe. The measured exponent of $\Gamma_u=2.6(0.5)$ agrees well with the $l(l+1)/2$ law, which is obtained from a spin fluctuation model where $l=2$ at the surface. This shows that the spin reorientation transition can be understood as being driven by thermal spin fluctuations. © 1996 American Vacuum Society.

Ferromagnetism in films of transition metals a few atomic layers (ALs) thick has become well established since the development of modern vacuum equipment has allowed the preparation of high quality films.¹ The most striking behavior of some of these films, e.g., Fe grown on Cu(100) or Ag(100), is the presence of magnetic anisotropies large enough to overcome the dipole energy of the film and pull the magnetization perpendicular to the plane of the film.^{2,3} The presence of magnetic anisotropy is very important to the magnetic ordering of these systems because it has been shown that the critical behavior of the magnetism of very thin films comes very close to that of a true two-dimensional system.⁴ Since an isotropic two-dimensional system at finite temperature has no long range order,⁵ it is very important to understand how anisotropy enters into the problem. The Hamiltonian for a Heisenberg spin system can be written as a sum of exchange energy, spin–spin (dipole) coupling, and spin–orbit terms (with no external field),

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_D + \mathcal{H}_{\text{so}}. \quad (1)$$

If the exchange term is taken to be isotropic, e.g., using the Heisenberg model, then the anisotropy originates either in the dipole or the spin–orbit terms.

For a constant saturation magnetization M_s , the classical dipole energy/unit area of the film is obtained from

$$E_D = -\frac{1}{2} \mathbf{H}_{\text{loc}} \cdot M_s a_0, \quad (2)$$

where a_0 is the lattice constant. The local field \mathbf{H}_{loc} at the point \mathbf{r}_i exerted by a collection of point dipoles \mathbf{p}_j at $\mathbf{r}_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is given by

$$\mathbf{H}_{\text{loc}} = - \sum_{j \neq i} \left[\frac{\mathbf{p}_j}{r_{ij}^3} - 3 \frac{\mathbf{r}_{ij} \cdot \mathbf{p}_j \mathbf{r}_{ij}}{r_{ij}^5} \right] \dots \quad (3)$$

Evaluating this sum for perpendicular and in-plane magnetizations gives rise to a demagnetizing field $4\pi M_s$ in the plane of the film. Therefore, the total demagnetizing energy of the film (*shape* anisotropy energy) is

$$E_D = 2\pi M_s^2, \quad (4)$$

which always favors magnetization in the plane of the film. It has been shown that only the presence of a dipole term is sufficient to cause spontaneous magnetization.⁶

The spin–orbit term originates from the interaction of the electrons with the crystal lattice. In bulk bcc-Fe, for example, the spin orbit term is anisotropic, favoring magnetization in the [100]-like directions.⁷ The bulk anisotropy energy/unit volume in Fe is about an order of magnitude smaller than the shape anisotropy energy density in a thin film (10^6 vs 10^7 erg/cm³) and, hence, is unlikely to be a dominant contribution in Eq. (1). The breaking of symmetry at a surface can also cause the spin–orbit interaction to be anisotropic. In cubic materials the anisotropy vanishes to fourth order, however, at the surface the broken symmetry brings the second order anisotropy term into play. Neél⁸ has discussed the surface anisotropy with a phenomenological theory. Using the classical form of the pairwise interactions between neighboring atoms, he predicted that a strong anisotropy perpendicular to the plane of the film is a natural consequence of the reduced symmetry at, e.g., the (100) face of a cubic lattice. The contribution of this term can be included into the Hamiltonian as

$$\mathcal{H}_{\text{so}} = -2K_u \sum_i (\mathbf{S}_i \cdot \mathbf{n})^2, \quad (5)$$

where K_u is a uniaxial surface anisotropy energy/unit area and the factor of 2 includes both the top and bottom surfaces. Here, a positive value of K_u will minimize the energy, thus favoring magnetization perpendicular to the film. The energy density associated with this surface anisotropy term then becomes

$$E_{\text{Surf}} = \frac{2K_u}{d}, \quad (6)$$

with d being the film thickness. The first efforts to evaluate the direction and strength of K_u were made by Neél using typical elastic and magneto-striction constants. He predicted that K_u at the (100) face would favor magnetization perpendicular to the film and be on the order of 1 erg/cm². Thus, for a film of thickness less than about 10 Å the surface anisotropy energy/unit area, $E_{\text{Surf}} > 10^7$ erg/cm³, would be large enough to overcome the dipole energy. However, it is neces-

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sary to do a fully relativistic bandstructure calculation in order to predict from first principles the strength and sign of the magneto-crystalline anisotropy in transition metals. *Ab initio* electronic structure calculations which include the spin-orbit interaction correctly predicted that the moments of some thin transition metal films orient themselves perpendicular to the film,⁹ however it is clear that the difference between perpendicular and in-plane energies is small, and the calculations are pushing to the limits of present computational techniques.^{10,11}

These theoretical considerations in combination with experimental studies that confirmed that at low thickness the magnetization is perpendicular to the plane of the film has sparked much work over the past few years on this topic. In particular, it has been observed that films thinner than 5–6 AL of Fe grown on Ag(100)¹² are perpendicularly magnetized, and become magnetized in the plane of the film above this critical thickness. A similar spin reorientation behavior for Fe grown on Cu(100) is observed with a critical thickness of about 6 AL. These observations agree with expectations from ferromagnetic resonance results, in which the uniaxial anisotropy for Fe/Ag(100) was measured to be on the order of 1 erg/cm².¹³ Contributions to K_u may also arise from epitaxial strain,¹⁴ however it is interesting to note that the spin reorientation transition thickness is very close for Fe grown on Ag(100) and Cu(100), even though the structures are entirely different (bcc and fct, respectively).

The first indications of a temperature dependent spin reorientation from perpendicular at low T to in-plane at high T was observed by Jonker *et al.*,¹⁵ using spin resolved photoemission on Fe films grown on Ag(001), and was then verified by Volkening *et al.*¹⁶ using the conversion electron Mössbauer effect. Subsequent studies of Fe/Cu(100) grown at low temperature showed that the temperature dependent spin reorientation transition is reversible and occurs over a narrow temperature and thickness range¹⁷ with an accompanying loss of magnetic signal as the reorientation occurs for both Fe/Ag(100) and Fe/Cu(100).¹⁸ This loss of signal near has been investigated both from an experimental^{19,20} and theoretical perspective,^{21,22} and has been shown to be due to the formation of microscopic stripe domains in the region of thickness and temperature that $E_D = E_{\text{Surf}}$. This is important because it shows that the higher order anisotropy terms are sufficient to maintain short range magnetic order.

Observation of a temperature dependent spin reorientation leads to the consideration of the temperature dependence of the anisotropy constants with the conclusion that the surface anisotropy energy, E_{Surf} , decreases faster than the dipole energy, E_D , as the temperature is increased. This behavior can be expected because the origin of K_{Surf} is the same as that of the bulk magneto-crystalline anisotropy, i.e., the spin-orbit interaction. Callen and Callen have discussed the temperature dependence of the bulk anisotropy constants²³ using a spin fluctuation model. By allowing each spin to sample a local temperature independent anisotropy $K_l(0)$ and then allowing the spins to deviate from their equilibrium positions, the following relation is obtained:

$$\frac{K_l(T)}{K_l(0)} = \left[\frac{M_s(T)}{M_s(0)} \right]^\Gamma, \quad (7)$$

where l is the order of the anisotropy and $\Gamma = l(l+1)/2$. Good agreement with experiment is obtained using this expression for bulk bcc-Fe with exponent of $\Gamma_B = 10$, where $l=4$ for a cubic system. For the surface, however, an exponent of $\Gamma_u = 3$ is expected because the anisotropy comes in at second order, i.e., $l=2$.⁹ Spin fluctuation theory has been used successfully to understand the spin reorientation transition,^{21,22} however most emphasis has been placed on the nature of the domain formation at the reorientation transition rather than the cause of the temperature dependence of the uniaxial anisotropy. It has also never been shown experimentally that spin fluctuations can explain this effect without including thermal stress and bandstructure changes into the problem. In the present work, the exponent Γ is obtained for ultrathin Fe films grown on Cu(100) by comparing the saturation magnetization and thickness at which the spin reorientation occurs. This is achieved by finding the normalized magnetization $[M_s(T)]/[M_s(0)]$ at the spin reorientation temperature T_R for a given thickness,¹⁷ and equating the dipole energy density to the surface energy density at that thickness. Good agreement with the spin fluctuation model is found.

In order to find $[M_s(T)]/[M_s(0)]$ it is necessary to fit the dependence of M_s with respect to temperature in the range where the transition occurs because the long range order is reduced at T_R .¹⁸ Figure 1 illustrates the temperature and thickness dependence of the magnetization in thin Fe films which were grown at 125 K and annealed while the secondary electron polarization was monitored.¹⁷ The reduction of signal at T_R is demonstrated in the center panel for the 5.4 AL film and has been shown to be due to the formation of microscopic domains in this temperature range.^{20–22} The 4 and 10 AL films show only remanent magnetization perpendicular and in-plane, respectively. These results match with magneto-optical Kerr effect (MOKE) studies on Fe/Cu(100), where the border between perpendicular and in-plane magnetization was observed at ≈ 6 AL.^{12,3} The relevant aspect of these curves for this work is the decrease of the magnetization with temperature up to about 300 K. In the spin wave regime, i.e., for $T < T_c/2$, it is well known that the bulk and surface magnetization can be fit using the Bloch law $T^{3/2}$ law.^{7,24,25} For a thin film, however, it can be shown²⁶ that the behavior of the magnetization is more appropriately fit with a function of the form

$$\frac{M_s(T)}{M_s(0)} = 1 - kT \ln T. \quad (8)$$

The low temperature fit of the polarization data of the 5.6 AL film to Eq. (8) is shown in Fig. 2. Since it has been shown that the films are in a single domain state in this temperature region,^{12,19} this fit can be used to determine the magnetization at the spin reorientation temperature. A prefactor of $k=0.00068$ is obtained for the 5.6 AL film. Using this fit, it is now possible to determine the relation between $M_s(T)$ and

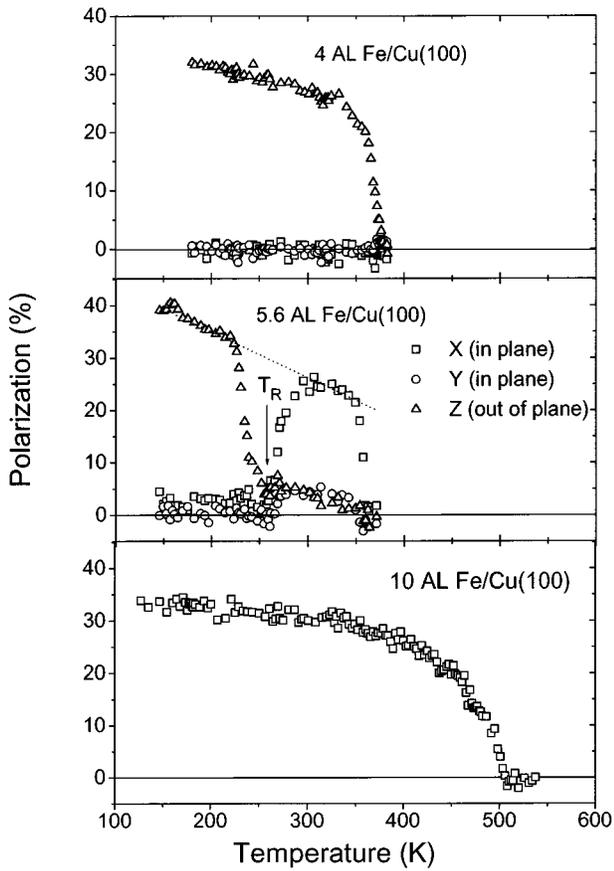


FIG. 1. Temperature dependence of the spin polarization of secondary electrons from a 4 AL (top), 5.6 AL (center), and a 10 AL film (bottom panel).

$K_u(T)$ for the first time from the data of Ref. 17. This can be obtained by observing that at the transition temperature T_R ; the dipole energy/unit volume E_D of the film is equal to the energy due to the uniaxial anisotropy $K_u(T_R)$. From Eq. (4) and Eq. (6) for a given thickness d_R , we obtain

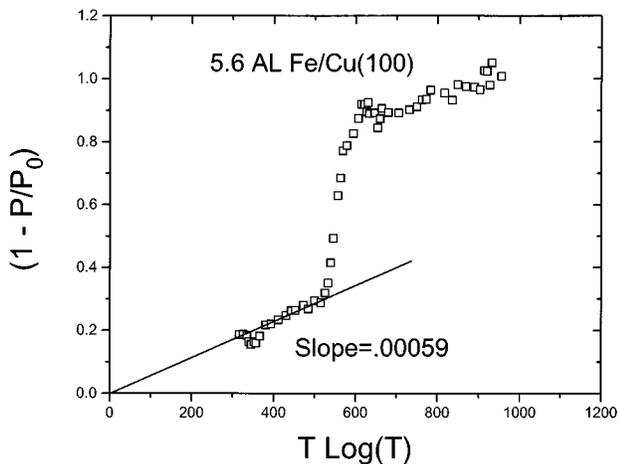


FIG. 2. Fit of the data from Fig. 1 to find the coefficient k of the equation $M_s(T) = M_s(0)[1 - k \times T \ln T]$ in the spin wave regime for 5.6 AL film.

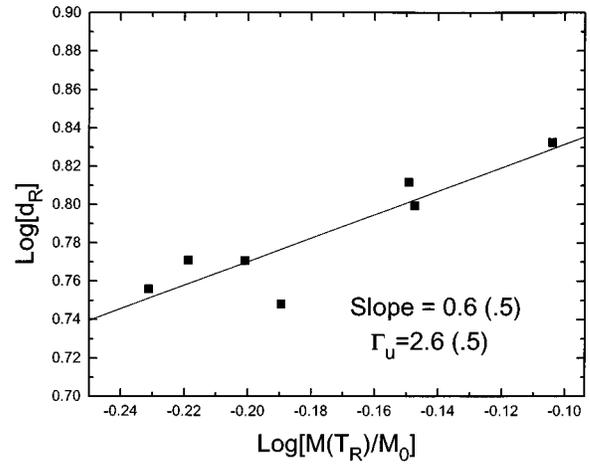


FIG. 3. Log-log plot of the thickness d_R vs magnetization $M_s(T_R)$ at the spin reorientation transition temperature. The data from this plot is obtained from the same data set as Ref. 17, where the corrected thickness calibration (see Ref. 18) is used.

$$\frac{2K_u(T_R)}{d_R} = 2\pi M_s^2(T_R). \quad (9)$$

Here, the normalized anisotropy and magnetization can be used, and the relation of Eq. (7) substituted for the value of $[K_u(T)]/[K_u(0)]$ to find

$$\frac{2}{d_R} \left[\frac{M_s(T_R)}{M_s(0)} \right]^{\Gamma_u} = 2\pi \left[\frac{M_s(T_R)}{M_s(0)} \right]^2 \quad (10)$$

and thus

$$d_R = \frac{1}{\pi} \left[\frac{M_s(T_R)}{M_s(0)} \right]^{\Gamma_u - 2}. \quad (11)$$

A log-log plot of d_R vs $[M_s(T_R)]/[M_s(0)]$ will thus give a slope of $\Gamma_u - 2$. Therefore, for each of seven films that demonstrated a spin reorientation transition the normalized magnetization is determined at T_R using Eq. (8). The results are plotted in Fig. 3. The linear least squares fit shown gives a slope of 0.6 ± 0.5 , and therefore $\Gamma_u = 2.6 \pm 0.5$.

In this work, the magnetic properties of Fe grown on Cu(100) substrates at low temperature are studied. The uniaxial anisotropy is observed to fall off faster than the bulk anisotropy as the magnetization decreases. A reduced exponent relative to the bulk is determined that agrees well with the spin fluctuation model exponent of $l(l+1)/2=3$ where $l=2$ at the surface. This demonstrates that thermal fluctuations have a much smaller effect on the anisotropy at the surface than in the bulk of a cubic material because the next closest energy minimum is 180° away versus only 90° in the bulk. This shows that the magnetic anisotropy can be understood using a thermal spin fluctuation model. However this does not rule out the possibility that thermal stress and/or band structure changes affect the magnetic properties of the films.

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