Static magnetization direction under perpendicular surface anisotropy

R. C. O'Handley and J. P. Woods*
Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
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The problem of depth dependence of the magnetization tilt angle $\theta(z)$ is solved analytically for an exchange-coupled ferromagnet with dipole energy, applied field, and surface anisotropy K_s. For $K_s \leq (\pi M^2 A)^{1/2} = K_c$, the surface magnetization direction remains in the plane, $\theta_s = \theta(0) = \pi/2$. For $K_s \geq K_c$, the magnetization vector moves out of the surface, initially with $d\theta_s/dK_s = -\infty$, then slowly approaches $\theta_s = 0$ as K_s approaches infinity. For Fe, K_c is of order 4 erg/cm^2. The results indicate that a perpendicular component of magnetization at the surface relaxes to its bulk in-plane orientation over a distance that is generally much shorter than the bulk domain-wall width. The surface remanence and initial susceptibility of the surface magnetization for in-plane applied field are calculated and provide useful expressions for interpreting experimental results. The experimentally measured surface magnetization increases linearly with probe sensitivity depth.

I. INTRODUCTION

Significant advances have been made recently in theoretical and experimental aspects of surface magnetism. Self-consistent local orbital calculations indicate anomalously large perpendicular anisotropy for Fe surfaces.[1] These calculations are supported by a host of measurements suggesting a significant perpendicular component of magnetization in Fe films of a few monolayers.[2–7] Recent results[6,7] indicate that the magnetization reverts to in-plane orientation for films greater than approximately four monolayers. Perpendicular surface magnetization is suggested by the spin polarization of electrons emitted from the [100] surface of iron single crystals, while the bulk shows in-plane magnetization.[8] This finding is challenged by the observed degeneration of a bulk Bloch domain wall in single crystal Fe to a Néel wall at the surface.[9] Finally, coupled magnetic layers, often one with a perpendicular easy direction and the other in plane,[10–12] reveal the possibility of extrinsic factors causing a perpendicular component of magnetization at the surface of an otherwise in-plane material.

Given the possibility of a strong perpendicular magnetic anisotropy in the outermost layers of a bulk ferromagnet, it is of interest to know the conditions on the applied field, on the bulk and surface anisotropy energy densities, and on the saturation magnetization M for which the surface magnetic moment would have a component out of plane. Further, it is important to determine how the magnetization orientation varies from the surface to the interior where only the bulk anisotropy operates. Does the change in magnetization orientation from surface to bulk occur over a distance comparable to a bulk domain-wall width, and how do surface magnetic dipole fields play a role? When the surface favors perpendicular magnetization in the outer few atomic layers, we expect that dipole energy will restore in-plane magnetization beneath the surface within a distance shorter than the bulk domain-wall width if the dipole energy cost of an out-of-plane magnetization $2\pi M^2$ exceeds the bulk, in-plane anisotropy. Some of these questions were addressed by Rado[13] who solved a linearized torque equation to deduce, among other properties, a characteristic length for surface magnetization relaxation. In contrast to Rado's treatment, we assume a material that is isotropic except for the surface-induced, perpendicular anisotropy. Our surface anisotropy constant K_s should be compared with Rado's term for a (100) surface; otherwise it is a mixture of his two terms K_s and K_{ss}.

When the surface anisotropy favors in-plane magnetization orthogonal to that in the bulk, dipole energy is not a factor and the magnetization should reorient from its surface direction toward that of the bulk over a distance comparable to the bulk domain-wall width (a few hundred nm in a soft magnetic material). This case is related to the extensively studied[14] problem of exchange coupling across a clean interface between two magnetic media each having a uniaxial anisotropy favoring a magnetization at a different orientation but both in the plane of the interface.

We consider the effect of perpendicular surface anisotropy K_s on the magnetization orientation both at the surface and in the subsurface regions in a classical continuum model. Exchange, dipole, and Zeeman energies are minimized by solving the Euler equation with appropriate boundary conditions. The solution gives the local equilibrium (experimental times are much longer than those required to relax precession of M about H) magnetization orientation everywhere in the material. We use these solutions to calculate the strength of the perpendicular surface anisotropy, which is needed to overcome exchange coupling to the bulk, and the depth to which a given surface anisotropy perturbs the subsurface magnetization. We also calculate the surface remanence and initial response to applied fields and use the results to interpret existing empirical data. The origin of the surface anisotropy is not at issue here.

II. SURFACE ANISOTROPY MODEL

Consider a semi-infinite ($z \geq 0$) magnetic medium of uniform saturation magnetization M and negligible bulk anisotropy. This medium has a perpendicular surface anisotropy K_s (erg/cm^2) acting only at $z=0$. We define the angle θ between the magnetization vectors **M** and the surface normal **n** and assume all the rotation of **M** takes place in the plane containing **n** and the applied field H (see Fig. 1). The total energy over the thickness d of such a sample is expressed[15]

$$E=\int_0^d \left[A\left(\frac{d\theta}{dz}\right)^2 - 2\pi M^2 \sin^2\theta - HM\cos(\theta-\psi)+\delta(z)K_s\sin^2\theta \right] dz \ . \quad (1)$$

A is the exchange stiffness coefficient, assumed to be independent of z. The Zeeman term is for a field applied at an angle ψ to the surface normal. The Dirac δ function gives the surface anisotropy term the appropriate units and localizes it at the free surface, $z=0$. Minimization of the energy using variational methods gives two torque equations. One is the Euler equation,

$$2A\frac{d^2\theta}{dz^2}+2\pi M^2\sin 2\theta - HM\sin(\theta-\psi)=0 \ , \quad (2a)$$

everywhere in the material $0 \leq z \leq \infty$, and the other is a boundary condition at $z=0$,

$$2A\frac{d\theta_s}{dz}-K_s\sin 2\theta_s=0 \ . \quad (2b)$$

This boundary condition is the same as that derived by Rado and Weertman from the Landau-Lifshitz equation.[16] We take the value of $\theta(z)$ deep in the material to be $\pi/2$.

A field applied in plane ($\psi=\pi/2$) does not change the asymptotic value of $\theta(\infty)=\pi/2$ while affecting mainly the magnetization near $z=0$. A perpendicular field ($\psi=0$) will affect the magnetization throughout the sample. In either case $(d\theta/dz)_\infty=0$ will be assumed.

A. Zero-field limit

In the limit where H approaches zero ($H \ll 2\pi M \approx 6$ kOe), Eq. (2a) is identical to that for a Bloch wall in a material with uniaxial anisotropy $K_u=2\pi M^2$. It has a bulk solution[17] $z=b^{-1}\ln[\tan(\theta/2+\pi/4)$, which does not satisfy the boundary condition (2b) for our surface problem. However, inverting it and generalizing it to the form

$$\theta(z)=2\tan^{-1}[\exp(a_0+b_0 z)]-\pi/2$$

or, equivalently,

$$\theta(z)=\tan^{-1}[\sinh(a_0+b_0 z)] \ , \quad (3)$$

exactly solves the surface problem for $H=0$.

Putting Eq. (3) in (2a) with $H=0$ gives the condition

$$b_0^2=2\pi M^2/A \ , \quad (4)$$

and with the boundary condition (2b), we get the additional condition

$$\tanh a_0 = Ab_0/K_s=(2\pi M^2 A)^{1/2}/K_s \ . \quad (5)$$

The parameter b_0 characterizes the distance over which the value $\theta(0)$ due to surface anisotropy relaxes to its bulk value $\pi/2$ and the parameter a_0 displaces $\theta(z)$ along the z axis so that its slope at $z=0$ satisfied Eq. (2b). For $M=10^3$ G and $A=10^{-6}$ erg/cm, we get $b_0^{-1}=4.0$ nm, much shorter than the bulk domain-wall width in a soft magnetic material. (These values of A and M are taken to characterize an arbitrary magnetic material, X, which, together with Fe, is used in Table I to realize the parameters of the present model.) In this case the demagnetization field is acting on **M** as a bulk anisotropy to make b_0^{-1} small. Our b_0^{-1} plays the same role as Rado's exponential decay length[13] but will clearly be shorter because of its definition in terms of the tangent function [Eq. (3)]. For the Fe parameters that Rado uses (Table I) our b^{-1} is of order 3.3 nm, while Rado's exponential relaxation length is 1.28 nm.

Equation (3) for $z=0$ combines with (5) to give

$$\csc\theta_s = K_s(2\pi M^2 A)^{-1/2}=K_s/K_c \ , \quad (6)$$

where $\theta_s=\theta(0)$. Equation (6) has real solutions only for $K_s \geq K_c$, i.e., the magnetization first moves out of the surface when K_s exceeds a critical value K_c ($K_c=2.5$ and 6.1 erg/cm^2 for materials X and Fe, respectively, Table I). For $K_s < K_c$, physical intuition dictates that $\theta_s=\pm\pi/2$. Thus a fairly strong surface anisotropy (3 or 6.5 ergs/cm^2 for X or Fe, respectively) is required to overcome dipole effects. Gay and Richter[1] calculate a surface anisotropy of less than 1 erg/cm^2 for an Fe monolayer. The exact

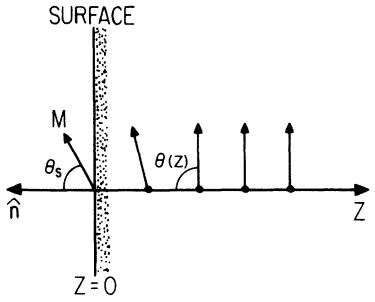

FIG. 1. Model for magnetic surface, $z=0$, showing magnetization tilt angle relative to surface normal at $z=0(\theta_s)$ and in the interior of sample $\theta(z)$.

TABLE I. Assumed magnetic parameters (A and M) and parameters calculated from present theory for Fe and for a hypothetical magnetic material X.

	Fe	X
A (10^{-6} erg/cm)	2	1
M (G)	1 720	1 000
b_0^{-1} (nm) from Eq. (4)	3.3	4.0
K_c (erg/cm^2), Eq. (6)	6.1	2.5
H/H^* (Oe), $K_s=K_c$	43 200	25 100

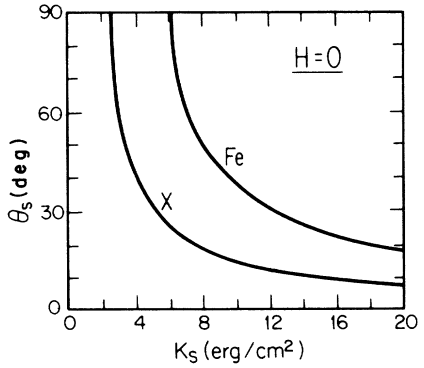

FIG. 2. Variation of surface tilt angle θ_s as a function of surface anisotropy K_s for zero in-plane field [Eq. (6)]. Material X and Fe parameters, Table I, are used.

behavior of θ_s versus K_s from Eq. (6) is shown in Fig. 2, where it is clear that M literally pops out of the surface plane (having a component into or out of the material) as K_s first exceeds its critical value. This critical value is the bulk domain-wall energy density for an in-plane anisotropy given by the demagnetization energy (magnetization at the center of the wall points out of the surface). The total magnetization is never perfectly normal to the surface for weak fields and finite K_s.

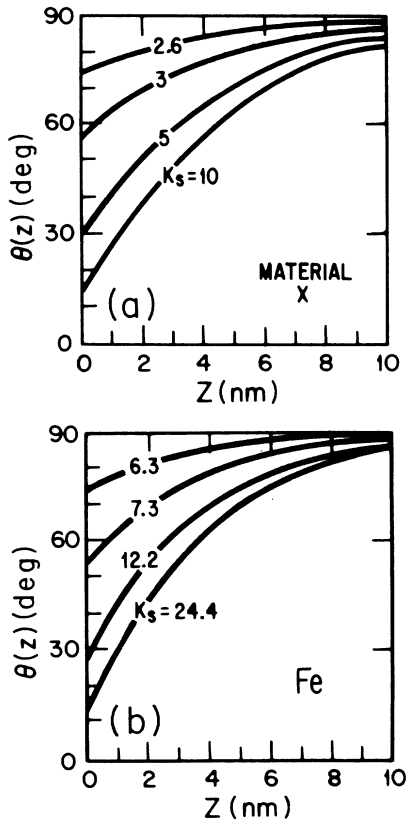

FIG. 3. Surface tilt angle as a function of distance from surface [Eq. (3)] for several values of surface anisotropy and assuming material X parameters (a) or Fe parameters (b) (Table I). The K_s/K_c ratios chosen are the same for Fe as for material X.

With the definitions of a_0 and b_0 from Eqs. (5) and (4), respectively, we can calculate $\theta(z)$ exactly for different values of K_s. These results are shown in Fig. 3(a) for material X parameters ($K_c=2.5$ erg/cm^2) and $K_s=2.6$, 3.0, 5.0, and 10 erg/cm^2. Figure 3(b) shows results for Fe ($K_c=6.1$ erg/cm^2) and $K_s=6.3$, 7.3, 12.2, and 24.4 erg/cm^2 [same ratios K_s/K_c for Figs. 3(a) and 3(b)]. Note that as K_s decreases from very large values toward K_c, θ_s increases from zero toward $\pi/2$ [Eq. (6)] and a_0 increases diverging at $K_s=K_c$ [Eq. (5)]. The set of ratios K_s/K_c are the same for the two materials in Fig. 3 so the sets of θ_s values correspond. However, it is clear that the larger dipole energy of Fe relative to X causes its magnetization to rotate toward in-plane orientation over a shorter range. Compare b_0^{-1} for the two materials.

B. Field dependence

We now consider the dependence of the solutions (3) on in-plane ($\psi=\pi/2$) applied field. Using the zero-field parameters a_0, b_0, and K_c defined above, we rewrite the Euler equation and boundary condition:

$$\theta''+(b_0^2/2)\sin 2\theta+h\cos\theta=0 \tag{7a}$$

and

$$\theta_s'-(b_0K_s/2K_c)\sin 2\theta_s=0\ , \tag{7b}$$

where $\theta_s'=(d\theta/d_z)_{z=0}$, $h=HM/2A$, and b_0 and K_c are defined in (4) and (6).

We assume a solution of the form (3) but now allow a_0 and b_0 to become functions of the applied field, a and b,

$$\theta(z,h)=\tan^{-1}[\sinh(a+bz)]\ , \tag{8}$$

which reduces Eqs. (7) to

$$b^2-b_0^2-h/\sin\theta=0 \tag{9a}$$

and

$$Ab/K_s=\sin\theta_s\ . \tag{9b}$$

Because $\sin\theta$ is a function of z, it is clear from Eq. (9a) that (8) is not a solution because b was taken as independent of z. However, we can seek approximate solutions to Eqs. (9) valid for $bz\ll 1$, which is the region of physical interest for secondary-electron spectroscopy.[18] Moreover, we know the behavior in the limit of $bz\gg 1$, namely $\theta\approx\pi/2$, unchanged by an in-plane field. Using Eq. (9b) we rewrite (9a) *at the surface*

$$b^3-b_0^2b-hK_s/A=0\ . \tag{10}$$

This cubic equation in b may be solved by the usual method for such equations with linear and constant terms that are negative.[19] Defining $b=(2b_0/\sqrt{3})\cos\phi$, Eq. (10) is satisfied for

$$\cos 3\phi=3\sqrt{3}hK_s/(2Ab_0^3)=3\sqrt{3}H^*\ .$$

The solution for the reduced surface magnetization, $\sin\theta_s=bA/K_s=(2K_c/K_s\sqrt{3})\cos\phi$, which is *exact* in H,

is plotted in Fig. 4. The solid lines are for material X, the dotted lines for Fe and the dashed line is the low-field behavior for X (see below). For $H^* \ll 1$ this solution gives

$$b = b_0(1+H^*) , \tag{11a}$$

and the boundary condition becomes

$$\tanh a = Ab/K_s = (K_c/K_s)(1+H^*) \tag{11b}$$

(for material X, $H^* \approx 5\times 10^{-5} H$, so the approximation $H^* \ll 1$ is good for $H < 10^3$ Oe). It is easily verified that (8) and (11) solve (7) for small fields at the surface ($bz \ll a$). They are not valid for arbitrary z. Equation (11b) gives the reduced magnetization at the surface

$$\sin\theta_s = m_0 = (K_c/K_s)(1+H^*) = m_r + H/8\pi M , \tag{12}$$

which defines the low-field magnetic response of the outermost layer of the material. This is shown for material X by the dashed line in low fields in Fig. 4. The arrows indicates the field H for which $H^* = 0.1$, below which the low-field approximation is valid. The susceptibility of the surface for a parallel field is

$$\chi_{\parallel}/M = dm_0/dH = (8\pi M)^{-1} . \tag{13}$$

Thus $\chi_{\parallel} \approx 0.04$ regardless of the material parameters. The initial surface susceptibility is constant even though the $m_0 - H$ curve begins from a different remanent point for different surface anisotropies.

By analogy with Eq. (6) and Fig. 2, Eq. (12) also defines the field dependence of the critical angle in the weak-field approximation. The dotted and solid lines in Fig. 5 show the exact surface magnetization angle for the two materials in $H = 1000$ and 5000 Oe.

The abrupt knee at saturation, visible in Fig. 4 for $K_s = 2.6$ (6.3) erg/cm^2 at $H \approx 1000$ Oe is a consequence of the infinite slope of the θ_s versus K_s curve at $K_s = K_c$. For a given $K_s > K_c$, as H increases, θ_s increases and abruptly reaches $\theta_s = \pi/2$ when the field causes the field-dependent critical surface anisotropy to exceed K_s. The knee will *not* be observed for K_s greater than some limiting value because the full field dependence of the critical anisotropy indicates that it increases slowly with H and is concave downward.

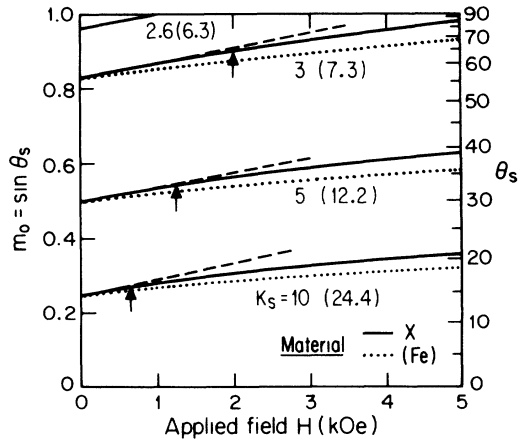

FIG. 4. Reduced surface magnetization (left scale) or tilt angle (right scale) for material X (solid line) and for Fe (dotted line) as functions of applied field [Eq. (10)]. Dashed lines show reduced low-field susceptibility [Eq. (13)] for material X.

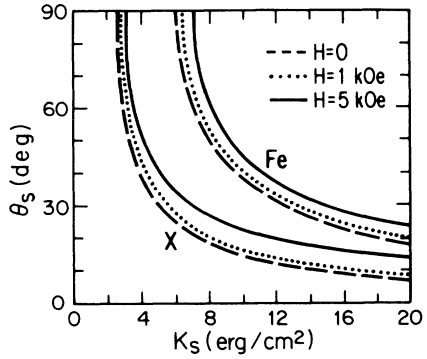

FIG. 5. Dependence of surface tilt angle on surface anisotropy for Fe and material X at 1 and 5 kOe [Eq. (10)]. Zero-field curves from Fig. 2 are shown as dashed lines for reference.

It is interesting to consider the magnetization over a depth ξ near the surface ($bz \ll 1$), where ξ is the characteristic penetration depth of the experimental probe. We write the magnetization measured by such a probe as

$$\langle m(H) \rangle = [\xi(1-e^{-d/\xi})]^{-1} \int_0^d e^{-z/\xi} \sin\theta(H,z) dz , \tag{14}$$

where $\sin\theta(H,z)$ is the reduced magnetization in the material that satisfies Eq. (7). The surface susceptibility [Eq. (13)] is independent of θ_s (or equivalently of m_0) for $H^* \ll 1$. We can, therefore, assume that $\langle m \rangle$ also responds with constant susceptibility $\chi \approx 1/8\pi$ to weak fields. Therefore, the low-field magnetization dependence *at* the surface, Eqs. (9) or Fig. 4, is a good first approximation to that over a shallow depth. That is,

$$\langle m(H) \rangle \approx \sin\theta_s = m_r + H/(8\pi M) . \tag{15}$$

III. DISCUSSION

We apply these results to some recent observations that deal with the orientation of magnetization at a surface and the possibility of magnetic depth profiling to determine $M(z)$ or $\theta(z)$ by varying the experimental probe depth sensitivity ξ. Allenspach *et al.*[8] have analyzed the spin polarization of low-energy secondary electrons emitted from a (100) Fe surface. By varying the primary- and secondary-electron energy they have concluded that a square loop with $m_r \approx 1$ (their measured remanent polarization of 30% is close to that of Fe at saturation) is observed for electrons probing up to 1.0 nm, whereas they find that a slightly rounded M-H loop with $m_r \approx 0.3$ characterizes the outer 0.5 nm of the crystal. (Their magneto-optic Kerr effect (MOKE) loops probing 20 nm also show a square loop.) They interpret their result qualitatively to imply a significant perpendicular moment in the outer 0.5 nm of the crystal with the moment returning to an in-plane orientation by 1.0 nm. Other re-

sults on an amorphous cobalt-based alloy, also using spin-polarization analysis of secondary electrons and magneto-optic Kerr effect, have been reported recently.[20] These cobalt results show a rounded loop with very low remanence ($m_r=0.2$) for the outermost 0.5 nm by secondary-electron-spin polarization and a square loop by MOKE, again indicating a perpendicular surface magnetization relaxing to in-plane orientation well before a depth of 25 nm. The magnitude we calculate for b_0 and the curves shown in Fig. 3 indicate that a relaxation to bulk orientation is possible over 5–10 nm. It is difficult to justify $(b_0)^{-1}$ of order 1 nm unless the magnetization were also reduced in magnitude near the surface.

The value of $\langle m_r \rangle = 0.3$ from the near surface (0.5 nm) data of Allenspach *et al.*[8] for Fe(100) implies $\theta_s \approx 17^0$. Using Eq. (9b) with the Fe values from Table I, this θ_s gives $K_s \approx 20$ erg/cm^2 [cf. Fig. 3(b)]. This surface anisotropy is greater than the total surface spin-orbit energy of Fe (11 erg/cm^2) calculated by Gay and Richter[1] and further exceeds the magnitude of their calculated perpendicular surface anisotropy energy (0.7 erg/cm^2). It also exceeds the values for Fe deduced from ferromagnetic resonance data, 1.0 (Ref. 2), 0.1 (Ref. 5), and 3.0 (Ref. 21) erg/cm^2 and values for amorphous $Fe_{70}B_{30}$ films 0.5 erg/cm^2 (Ref. 22). Thus, interpretation in terms of the present model suggests that the magnetization at the surface of the Fe(100) crystal was reduced in magnitude relative to that in bulk or that a strong, unaccounted for perpendicular anisotropy existed there.

To apply the results of the present model without the approximations $bz \ll 1$ and $H^* < 0.1$, consider an experimental probe whose depth can be varied (energy or incident angle for electrons, wavelength for magneto-optic Kerr effect). The polarization or Kerr rotation measured parallel to the surface is proportional to the magnetization in Eq. (14). We write these expressions for two different experimental probe depths ξ_1 and ξ_2:

$$J_1=[\xi_1(1-e^{-d/\xi_1})]^{-1}\int e^{-z/\xi_1}\sin\theta(H,z)dz\ , \tag{16a}$$

$$J_2=[\xi_2(1-e^{-d/\xi_2})]^{-1}\int e^{-z/\xi_2}\sin\theta(H,z)dz\ . \tag{16b}$$

For assumed values of ξ_1, ξ_2, A, M, and K_s, these integrals can be numerically evaluated. The ratio J_1/J_2 can be calculated for several values of K_s and compared to the ratio of the corresponding empirical polarization data to determine the actual K_s. Our model has assumed that A and M are independent of depth. Variations of these parameters with temperature are expected to alter the surface magnetization orientation.

IV. SUMMARY

The problem of magnetization orientation near a surface with perpendicular anisotropy has an analytic solution

$$\theta(z)=\tan^{-1}[\sinh(a_0+b_0z)]$$

with $b_0=(2\pi M^2/A)^{1/2}$ and $\tanh a_0 = Ab_0/K_s$, independent of z. The magnetization remains in plane unless the surface anisotropy energy density exceeds a critical value $K_s > K_c$ of order 2–6 erg/cm^2.

The characteristic length b_0^{-1} over which the surface orientation of magnetization relaxes to its bulk value is of order 3–4 nm for Fe and other soft ferromagnets. This length scale is of the order of magnitude probed by electrons or photons.

The magnetization at the surface responds linearly to applied fields up to $H^* \approx 0.1$ (H of order 1 kOe) and the in-plane susceptibility is independent of K_s for weak fields: $\chi_\parallel = 1/8\pi$. The surface magnetization measured by an experimental probe of exponential range ξ shows a weak-field dependence similar to that right at the surface.

Recent observations of perpendicular magnetization at the surface of bulk magnetic materials suggest surface anisotropies at least an order of magnitude greater than those measured by magnetic resonance unless the magnetization is considerably reduced at the surface relative to bulk.

The surface anisotropy K_s can be measured by combining experimental probes sensitive to different depths ξ of order $(10b_0)^{-1}$ to $(b_0)^{-1}$.

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*Present address: Department of Physics, University of Nebraska, 260 Behlen, Lincoln, NE 68508.

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