Direction of the magnetization of thin films and sandwiches as a function of temperature

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We present a simple model for the temperature and thickness dependence of the direction of magnetization of ferromagnetic thin films or sandwiches. Above a temperature that is much smaller than the Curie temperature, the magnetization in the presence of a uniaxial anisotropy is aligned completely parallel to the surface plane. The main reason for this is the entropy of the magnetization vector. Also, the direction of magnetization is determined by the competition between uniaxial and shape anisotropy. The decrease of the magnetic ordering for increasing temperature turns out to be not so important.

I. INTRODUCTION

Recently, the magnetic properties of ferromagnetic thin films,[1] sandwiches, and superlattices have been studied intensively. These are, among other reasons, of great interest for new magnetic storage technologies (perpendicular recording). The systems consist of a few layers of $3d$ transition metals (Fe, Co, Ni) on or between nonmagnetic substrates. They can exhibit a relatively strong uniaxial magnetic anisotropy, which in turn is intimately connected with the direction of magnetization. It was shown experimentally for a variety of systems that the anisotropy is extremely sensitive to lattice geometry and thickness, type of substrate and coating layers, adsorbates, and also to lattice imperfections, stress, and induced strain due to growth conditions and other influences. Therefore, a puzzling variety of magnetic surface structures is observed. For example, a thin film smaller than six monolayers (ML) of fcc-Fe on Cu(100) shows perpendicular magnetization,[2-5] whereas bcc-Fe films on Ag(100) exhibit a remanent magnetization along the surface normal only for 3–4 ML.[5-7] For the latter system a vertical remanent magnetization exists for uncovered or Ag- coated films, but does not for Au overlayers.[5] Co films on an Au(111) substrate exhibit a vertical magnetization,[8] but not on Ag(100).[9] At elevated temperatures and for thicker films the magnetization lies always in the surface plane. Due to the strong dependence on the actual film preparation, different experimental results have been obtained for the same system.

In this theoretical study we want to determine, using a simple method, how the direction of magnetization of a thin ferromagnetic film changes with temperature and film thickness. In particular we look for the thermodynamic conditions for a magnetization with vertical components. The anisotropy and exchange coupling parameters are merely taken as given constants. The determination of these quantities is beyond the scope of this work. Its calculation is in fact a very complicated and expensive task because of the above mentioned variety of influences.[10,11] We claim that despite constant anisotropy parameters and a magnetization perpendicular to the surface at $T=0$, the magnetization turns into the surface plane with increasing temperature. The reason for this is the larger entropy of the in-plane magnetization compared to the magnetization perpendicular to the surface.

In Sec. II we set up the Hamiltonian containing the essential interactions and describe the approximate partition function to estimate the temperature dependence of the surface magnetization. In Secs. III and IV results for some special cases are given. These are discussed in Sec. V.

II. THE HAMILTON FUNCTION

The exchange coupling in a ferromagnetic system (only such collinear structures are considered here) generates a parallel alignment of the magnetic moments, but has no influence on the direction of magnetization relative to the crystal lattice. This is rather fixed by the much weaker spin-orbit and (classical) spin-spin interactions. For surface layers and interfaces these interactions are pronouncedly stronger than in bulk.[11] Due to the strong exchange coupling the Curie temperature T_C is still much higher than the interesting temperature range considered here. So we use—also for itinerant electron systems—the picture of classical localized magnetic moments $\boldsymbol{\mu}=\mu\mathbf{S}$ with constant magnitude and only a small decrease of magnetization: $|\langle\mathbf{M}(T)\rangle|\approx|\langle\mathbf{M}(0)\rangle|=M_0$. Visualizing a classical vector picture this means that the magnetic moments are completely aligned or have only a small angle with the average direction of magnetization. For a given lattice geometry we assume completely filled and perfectly ordered layers with the same magnitude of the magnetic moments throughout the film. Only the uniaxial anisotropy is considered here. An azimuthal (in plane) anisotropy may also be present, but is normally one order of magnitude weaker.[12] Therefore, we assume the magnetization vector $\mathbf{M}$ to precess freely around the surface normal $\mathbf{n}$, θ_M being the angle between $\mathbf{M}$ and $\mathbf{n}$. Thus the in-plane component of $\mathbf{M}$ can be aligned by small external fields.

Before dealing with special cases we consider first the total Hamiltonian function:

$$\mathcal{H}'_g=\mathcal{H}'_{ex}+\mathcal{H}'_f+\mathcal{H}'_d+\mathcal{H}'_{so} . \quad (2.1)$$

The exchange interaction (Heisenberg model) and the external field energy are given by

$$\mathcal{H}'_{ex}+\mathcal{H}'_f=-J'\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-\mathbf{H}'\cdot\sum_i\boldsymbol{\mu}_i , \quad (2.2)$$

$\langle i,j\rangle$ denotes summation over nearest-neighbor pairs. In order to calculate hystersis loops we consider an external magnetic field $\mathbf{H}'=(0,0,H')$ perpendicular to the film plane. $\mathcal{H}'_d$ is the demagnetization energy (shape anisotropy) which originates from the classical spin-spin coupling[13] and can readily be written as

$$\mathcal{H}'_d=\frac{\mu_0}{2}(\mathbf{M}\cdot\mathbf{n})\sum_i(\boldsymbol{\mu}_i\cdot\mathbf{n}) , \quad (2.3)$$

μ_0 being the vacuum permeability. For surfaces or layered structures $\mathcal{H}'_d$ prefers an in-plane magnetization ($\theta_M=\pi/2$), but does not depend on azimuthal angle for (100) or (111) faces of cubic lattices. $\mathcal{H}'_d$ is proportional to the total number of magnetic moments and, respectively, to the volume of the sample. The first two terms of the uniaxial anisotropy are given by

$$\mathcal{H}'_{so}=\sum_i[K'_2(\boldsymbol{\mu}_i\cdot\mathbf{n})^2+K'_4(\boldsymbol{\mu}_i\cdot\mathbf{n})^4] , \quad (2.4)$$

where we adopt the sign convention of Gradmann.[12] In many cases $|K'_2|>|K'_4|$, therefore in the following calculations we consider first the K_2 term only. In principle these parameters can be calculated by taking the spin-orbit interaction as a small perturbation of e.g., a Hubbard Hamiltonian.[10] Unfortunately, a reliable calculation is very troublesome because of its strong dependence on details of band structure and a variety of other influences. The effect of a surface or interface are confined practically exclusively to the surface/interface layer itself as shown by theoretical[11] and experimental[5] examinations. For other layers the parameters K'_2 and K'_4 are comparable to the bulk anisotropy constants. Because these are ≈ 100 times smaller than the ones in the surface layers,[11] they will be neglected in the following calculations.

Therefore, the uniaxial anisotropy energy is proportional to the surface area of the sample. Because the demagnetization energy is proportional to the volume, the magnetization of thick films and pure surfaces of bulk systems will lie in the surface plane ($\theta_M=\pi/2$). In contrast, for thin films $\langle\mathbf{M}\rangle$ may be perpendicular to the surface or can have a vertical component ($\theta_M<\pi/2$) *provided that* $K'_2<0$ and if the uniaxial anisotropy is strong enough to overcome the demagnetization energy. Because we are interested in the case of competing uniaxial and shape anisotropy interactions, we assume throughout this work $K'_2<0$ only. In polycrystalline films the uniaxial anisotropy vanishes, the direction of the magnetization is solely determined by the shape anisotropy.[9]

Callen and Callen[14] and later on Levinson[15] examined the temperature dependence of the magnetic anisotropy. They treated Eq. (2.4) as a small perturbation of the exchange coupling which leads to an expression of the free energy with temperature-dependent anisotropy constants A_2 and A_4 (cf. Appendix A):

$$F=F_0(T)+A_2(T)\cos^2\theta_M+A_4(T)\cos^4\theta_M . \quad (2.5)$$

$A_2(T)$ and $A_4(T)$ vary *only* through the decrease of the average magnetization $|\langle\mathbf{M}(T)\rangle|$. A variation of θ_M can happen if both K'_2 and K'_4 terms are present. If only a $K'_2<0$ term is considered, the magnetization will maintain the constant direction $\theta_M=0$ up to T_c.[15] An explicit temperature variation resulting from the entropy of the magnetization vector $\mathbf{M}$ itself was not taken into account.

In the following calculations the exchange coupling is treated within a molecular field approximation (if the complete spin-spin interaction[13] is required, it should also be treated in this way). This means that only one magnetic moment $\boldsymbol{\mu}_1$ is considered, and it interacts with the average magnetization $\langle\mathbf{M}\rangle$. The surface normal $\mathbf{n}$ is set along the z axis, whereas the x axis is chosen to be parallel to the in-plane component of $\mathbf{M}$. Therefore $\mathbf{M}$ is given by $\mathbf{M}=(\mu/v)(\sin\theta_M,0,\cos\theta_M)$, v being the unit-cell volume of the respective lattice. The polar coordinates of $\boldsymbol{\mu}_1$ relative to $\mathbf{M}$ are θ and ϕ, so that $\boldsymbol{\mu}_1$ relative to $\mathbf{n}$ is readily written by

$$\boldsymbol{\mu}_1=\mu\begin{pmatrix}\sin\theta_M\cos\theta+\cos\theta_M\sin\theta\cos\phi\\ \sin\theta\sin\phi\\ \cos\theta_M\cos\theta-\sin\theta_M\sin\theta\cos\phi\end{pmatrix}$$

(cf. Fig. 1). In molecular field theory $\langle\mathbf{M}\rangle$ is given by $\langle\mathbf{M}\rangle=\langle u\rangle\mathbf{M}$ with $u=\cos\theta$. Therefore, the four terms of the Hamiltonian $\mathcal{H}_g$ per area of unit cell of the ferromagnetic film with thickness $d\geq 2$ lead to the expressions[13]

$$\mathcal{H}_{ex}+\mathcal{H}_f=-J(\cos\theta-\tfrac{1}{2}\langle u\rangle)\langle u\rangle-d\mu H\xi , \quad (2.2')$$

$$\mathcal{H}_d=\alpha d\langle u\rangle\xi\cos\theta_M , \quad (2.3')$$

$$\mathcal{H}_{so}=(K_2^{(1)}+K_2^{(2)})\xi^2+(K_4^{(1)}+K_4^{(2)})\xi^4 , \quad (2.4')$$

with

$$\xi=\cos\theta_M\cos\theta-\sin\theta_M\sin\theta\cos\phi , \quad (2.6)$$

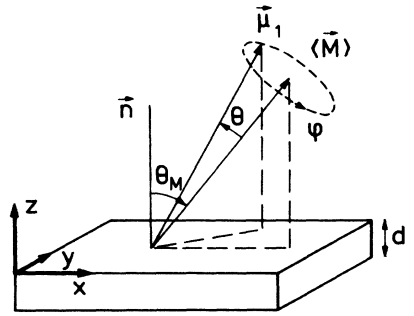

FIG. 1. Illustration of the geometry of the magnetic moment $\boldsymbol{\mu}_1$ and the average direction of magnetization $\langle\mathbf{M}\rangle$; d is the thickness of the film.

and $J=[(d-2)q+2\bar{q}]J'$, $H=H'/v$, $\alpha=\mu_0(\mu/v)^2/2$, and finally $K_i^{(n)}=\mu' K_i'^{(n)}$, $i=1,2$; $n=2,4$. q and $\bar{q}$ are the coordination numbers of the respective (cubic) lattice and its surface/interface. Like the magnitude of moments μ the exchange coupling J' and the angles θ and θ_M assumed to be the same for all film layers. $K_i^{(1,2)}$ are the anisotropy parameters for the two surfaces/interfaces of the film. We put $2K_i=K_i^{(1)}+K_i^{(2)}$ and point out again that $K_i=K_i(d)$ may depend critically on the film thickness d (Refs. 10 and 11). Neglecting a constant the Hamiltonian per unit area is summarized as

$$\mathcal{H}_g=-J\langle u\rangle\cos\theta-d\mu H\xi+2K_2\xi^2+2K_4\xi^4+\alpha d\langle u\rangle\xi\cos\theta_M \ . \tag{2.7}$$

For finite temperatures T the partition function of this Hamiltonian (with $\tilde{\mathcal{H}}=\beta\mathcal{H}=\mathcal{H}/k_BT$, etc., k_B Boltzmann constant) is given by

$$Z=\mathrm{Tr}e^{-\tilde{\mathcal{H}}_g/k_BT}=2\pi\int_0^\pi d\theta_M\sin\theta_M\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi\, e^{-\tilde{\mathcal{H}}_g} \ . \tag{2.8}$$

Although $|J|\gg|K_i|$, we avoid calculating the expectation value $\langle\theta_M\rangle$ using the uniaxial anisotropy as a small perturbation of the exchange coupling. For reasons stated below we rather confine the integration range of Z to a constant angle θ_{M_0} and determine this value by minimizing the free energy $F_0=-k_BT\ln Z_0$:

$$Z_0=2\pi\int_0^\pi d\theta_M\sin\theta_M\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi\, e^{-\tilde{\mathcal{H}}_g}\delta(\theta_{M_0}-\theta_M)$$
$$=2\pi\sin\theta_{M_0}\int_0^\pi d\theta\sin\theta\int_0^{2\pi}d\phi\exp(\tilde{J}\langle u\rangle\cos\theta+d\mu\tilde{H}\xi_0-2\tilde{K}_2\xi_0^2-2\tilde{K}_4\xi_0^4-\tilde{\alpha}d\langle u\rangle\xi_0\cos\theta_{M_0}) \ . \tag{2.9}$$

We point out that the value θ_{M_0} calculated in this way is only an approximation to the statistically correct value $\langle\theta_M\rangle$. For the simple external-field-only term the accuracy of this method is estimated in Appendix B. The expectation value for $u=\cos\theta$ is calculated in the usual way: $Z_0\langle u\rangle=\mathrm{Tr}ue^{-\tilde{\mathcal{H}}_g}\delta(\theta_{M_0}-\theta_M)$. To emphasize the basic features of this model we consider in the following sections some special cases.

III. COMPLETELY ORDERED SYSTEM AND $K_4=0$

First we assume a complete magnetic ordering, i.e., $\langle\theta\rangle=0$ or $\langle u\rangle=1$ or $|\langle\mathbf{M}(T)\rangle|=M_0$, and put $K_4=H=0$. Therefore, the Hamiltonian Eq. (2.7) is reduced to the expression

$$\mathcal{H}=\mathcal{H}_0+2K_2\cos^2\theta_M+\alpha d\cos^2\theta_M \ . \tag{3.1}$$

One sees immediately that at $T=0$ a perpendicular magnetization occurs ($\theta_M=0$), if $2K_2(d)<-\alpha d$. The partition function for this case is

$$Z=2\pi\int_0^\pi d\theta_M\sin\theta_M e^{\tilde{\gamma}\cos^2\theta_M} \ , \tag{3.2}$$

with $\gamma=-2K_2-\alpha d$. The integration argument $f(\theta_M)=\sin\theta_M e^{\tilde{\gamma}\cos^2\theta_M}$ is symmetric with respect to $\theta_M=\pi/2$ (Fig. 2). It has for $\tilde{\gamma}<\frac{1}{2}$ one maximum at $\theta_{M,\max}=\pi/2$, whereas two maxima at $\sin\theta_{M,\max}=\pm\sqrt{k_BT/2\gamma}$ are present for $\tilde{\gamma}>\frac{1}{2}$. Therefore the average value $\langle\theta_M\rangle$ is *always* $\pi/2$ (or $\langle\cos\theta_M\rangle=0$) for all temperatures. This is plausible for the case $\tilde{\gamma}<\frac{1}{2}$. On the other hand for $k_BT<-2K_2-\alpha d$ this is caused by the symmetric positions of the $f(\theta_M)$ maxima around $\pi/2$, while the statistical weight, e.g., for $\theta_{M,\max}$ is evidently larger than for $\theta_M=\pi/2$. This is the reason why we confine the range of θ_M integration [cf. Eqs. (2.8) and (3.2)] to a constant value θ_{M_0} as a first estimation for the temperature dependence of $\langle\theta_M\rangle$. Thus the magnetic moment μ_1 is fixed to the surface of a cone. We arrive at

$$Z_0=2\pi\sin\theta_{M_0}e^{\tilde{\gamma}\cos^2\theta_{M_0}} \ . \tag{3.3}$$

By minimization of the free energy ($\partial F_0/\partial\theta_{M_0}=0$) we reach for the case $\tilde{\gamma}\geq\frac{1}{2}$ at

$$\sin^2\theta_{M_0}=\sin^2\theta_{M,\max}=k_BT/2\gamma \ . \tag{3.4}$$

The vertical component of the magnetization is given by $M_z=M_0\cos\theta_{M_0}=M_0\sqrt{(2\gamma-k_BT)/2\gamma}$, which is shown in Fig. 3. The value of $\theta_{M_0}(T)$ yielded with this approximation is smaller than the correct one because fluctua-

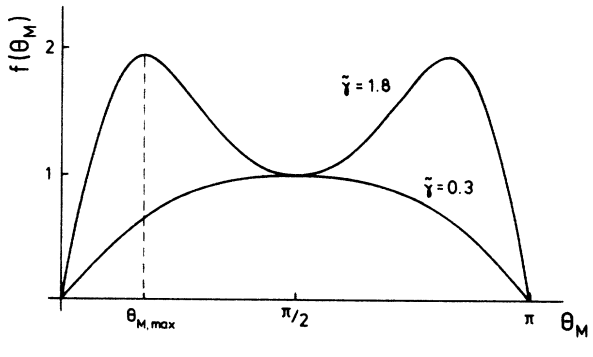

FIG. 2. Statistical weight $f(\theta_M)=\sin\theta_M\, e^{\tilde{\gamma}\cos^2\theta_M}$ of the Hamiltonian Eq. (3.1) for two values of $\gamma=-(2K_2+\alpha d)$, $\tilde{\gamma}=\gamma/k_BT$ below and above $T_\parallel=2\gamma$. Note, at $T_\parallel$ the two maxima of $f(\theta_M)$ shrink to a single one for $T>T_\parallel$. K_2 is the uniaxial anisotropy coefficient, αd refers to the shape anisotropy.

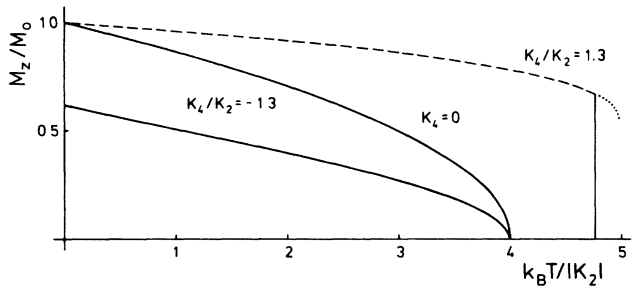

FIG. 3. Vertical component of the film magnetization $M_z/M_0=\cos\theta_{M_0}$ for $K_4/K_2=0.0$, 1.3, and -1.3, assuming $K_2<0$ and $\alpha=0$. The dotted line denotes an unstable solution. Note the discontinuity of the curve for $K_4/K_2=1.3$ in contrast to the other cases.

tions of θ_M are completely neglected. The temperature $T_\parallel$ at which vertical components of the magnetization vanish (i.e., $\theta_{M_0}=\pi/2$) is given by

$$k_BT_\parallel=2\gamma=-4K_2-2\alpha d\ . \tag{3.5}$$

Note that $\theta_{M_0}\propto\sqrt{T}$ or $\cos\theta_{M_0}\approx 1-T/2T_\parallel$ for $T\gtrsim 0$, and $\pi/2-\theta_{M_0}\approx\cos\theta_{M_0}\propto\sqrt{T_\parallel-T}$ for $T\lesssim T_\parallel$.

We emphasize that $T_\parallel$ is in general much smaller than the Curie temperature T_C. Such a temperature does not exist for a system confined to a circle, i.e., in two dimensions. Possibly the estimated value of $T_\parallel$ is too large. In principle one can determine the parameter $K_2(d)$ by measuring this temperature. However, for $T\approx T_\parallel$ the angle θ_{M_0} is subjected to strong fluctuations because the free energy is almost flat in the region $\theta_{M_0}\simeq\pi/2$. A similar property of fluctuations is found, e.g., in the neighborhood of Lifshitz points.[16]

IV. EFFECT OF DIMINISHED MAGNETIZATION AND $K_4\neq 0$

If the decrease of magnetization for finite temperatures is taken into account one has to, in principle, carry out the integrations over ϕ and $u=\cos\theta$ [Eq. (2.9)]. Because $|J|\gg|K_2|$, $\langle u\rangle$ can be taken from the mean field result of the pure exchange interaction. A subsequent expansion of the respective equations to order T/J shows that the correction to the temperature $T_\parallel$ is of order $|K_2|/J$. We do not show here the rather lengthy expression.

We like to emphasize for the temperature behavior of the magnetic anisotropy the important difference between the earlier calculations[14,15] and our results. For a pure K_2 term the average value of θ_M yielded by the former method does not depend on K_2, but takes the values $\langle\theta_M\rangle=0$ or $\langle\theta_M\rangle=\pi/2$ for all $T<T_C$ according to the sign of K_2. Even if the parameter $A_2(T)$ [cf. Eq. (2.5)] is interpreted as a vertical component of the magnetization, it results in a value $\langle\theta_M\rangle<\pi/2$ also for $T\lesssim T_C$ (assuming $K_2<0$). On the contrary our results state that the orientation of magnetization is aligned to the surface plane ($\theta_M=\pi/2$) above a temperature $T_\parallel$ much lower than T_C. This holds also if a decrease of magnetization is taken into account.

We examine now the effect of an additional K_4 term [Eq. (2.4)] on the uniaxial anisotropy. For simplicity we assume $\langle\theta\rangle=0$ and also $\alpha=0$ [cf. Eq. (2.3′)]. Similar to the K_2 term with $K_2<0$ a pure K_4 term with $K_4<0$ results in a perpendicular magnetization at $T=0$, i.e., $\theta_{M_0}(0)=0$. For $T>0$ we use the same approximation of the partition function Eq. (2.9) and determine the direction of magnetization θ_{M_0} through $\partial F/\partial\theta_{M_0}=0$. Therefore θ_{M_0} in this case is given by $\sin^2(2\theta_{M_0})=T/(-2K_4)$, at $T\simeq 1.639(-K_4)$ the value of θ_{M_0} *jumps* from $\theta_{M_0}\simeq 32.25°$ to $\pi/2$. In contrast, for a pure K_2 term the high temperature value $\theta_{M_0}=\pi/2$ is reached *continuously* at $T_\parallel=4(-K_2)$, cf. Eqs. (3.4), (3.5), and Fig. 3.

If both K_2 and K_4 terms are present, the average value of θ_{M_0} is given by

$$\begin{aligned}\sin^2\theta_{M_0}&=\frac{\tilde\gamma+2\tilde\delta}{4\tilde\delta}-\frac{1}{4\tilde\delta}\sqrt{(\tilde\gamma+2\tilde\delta)^2-4\tilde\delta}\\&=\frac{K_2+2K_4}{4K_4}+\frac{1}{4K_4}\sqrt{(K_2+2K_4)^2+2k_BTK_4}\end{aligned} \tag{4.1}$$

or

$$\begin{aligned}\sin^{-2}\theta_{M_0}&=\tilde\gamma+2\tilde\delta+\sqrt{(\tilde\gamma+2\tilde\delta)^2-4\tilde\delta}\\&=-\frac{2}{k_BT}(K_2+2K_4)\\&\quad+\frac{2}{k_BT}\sqrt{(K_2+2K_4)^2+2k_BTK_4}\ ,\end{aligned} \tag{4.1′}$$

where we put $\delta=-2K_4$ and, as usual, $K_2<0$. For $|K_4|<|K_2|/2$ the temperature behavior is very similar to the case $K_4=0$. At low temperatures again $\theta_{M_0}\propto\sqrt{T}$, and the value $\pi/2$ is reached steadily at $T_\parallel=4(-K_2)$, independent of K_4.

On the other hand for $|K_4|>|K_2|/2$ *and* $K_4<0$, the direction of magnetization jumps to $\pi/2$ at a temperature $T>T_\parallel$, the width $\Delta\theta_{M_0}$ and the transition temperature are dependent on $|K_4/K_2|$. In this case the (unstable) end point of the magnetization curve is given by $2\cos^2\theta_{M,\mathrm{end}}=1-1/2\kappa$ and $2k_BT_{\mathrm{end}}/\gamma=(1+1/2\kappa)(1+2\kappa)$, assuming $\kappa=\delta/\gamma\geq\frac{1}{2}$ (cf. Fig. 3). In addition for $|K_4|>|K_2|/2$ *and* $K_4>0$ one has to take into account that for $T=0$ the magnetization is no longer perpendicular to the surface, but has an angle $\cos^2\theta_{M_0}=-K_2/2K_4$. Starting from this value $\theta_{M_0}(T)$ is continuous and reaches $\pi/2$ at $T_\parallel$. Note that the model used by Levinson also exhibits discontinuities if both K_2 and K_4 terms are present.[15]

V. DISCUSSION

Experimental results state that for all systems the magnetization turns into the surface plane with increasing

film thickness and temperature. This is also a property of our model, and by comparing our theoretical ansatz with experiments we can estimate the values of the anisotropy parameters. Many experiments determined the hysteresis curves.[2,3,6,8] Very thin ferromagnetic films are assumed to be in a single domain state in the direction vertical to the film plane.[17] This in turn would produce a hysteresis loop with a discontinuous jump from $\langle \mathbf{M} \rangle$ to $-\langle \mathbf{M} \rangle$. However, experiments show a slightly tilted curve which may indicate the presence of a domain structure along the film surface. In addition, a remanence magnetization close to the saturation magnetization, resulting in an almost rectangular hysteresis loop [cf. Fig. 4(b)], was reported. To examine this we consider the external field term Eq. (2.2′). Our calculations assume inherently a single domain state. For small temperatures compared to $T_{\parallel}$ we obtain also an almost rectangular hysteresis loop with only a minute variation of $|\mathbf{M}(H)|$, see Fig. 4(a). For larger temperatures the hysteresis loops become visibly rounded. The coercive field strength also diminishes with increasing temperature and, more pronounced, with increasing film thickness. All these theoretical results are in close agreement with experimental observations.

It should be mentioned that the system Au/Co/Au(111) [cf. Fig. 4(b)] might not be well suited as an example for our model, since Co might be in an hcp structure.[18] In this case the uniaxial anisotropy is not solely confined to the interface layers, but extends also in the bulk. Despite a thickness dependence of the hysteresis curves is also visible in this experiment,[8] one should merely compare our theoretical results to systems with cubic symmetries such as[2,3,6] Fe on Cu(100) or Ag(100), in which the uniaxial anisotropy exists practically exclusively in the surface or interface layers.

A Mössbauer spectroscopy experiment allows for certain cases a direct estimation of the angle θ_M. For the system bcc-Fe on Ag(100) at $T=15$ K Koon *et al.*[7] obtains $\theta_M \simeq 28°$ for a 2.4-ML film and $\theta_M \simeq 49°$ for a 5.5-ML one. Using Eq. (3.4) and assuming $\mu_{\mathrm{Fe}}=2.2\mu_\beta$ we calculate $K_2 \simeq -4.2$ meV/atom ($T_{\parallel} \simeq 160$ K) for the 2.4-ML and $K_2 \simeq -2.0$ meV/atom ($T_{\parallel} \simeq 65$ K) for the 5.5-ML film. One should remember that K_2 is thickness dependent and Eq. (3.4) is only a rough estimate for K_2. Nevertheless band-structure calculations at $T=0$ obtain the same magnitude for K_2, whereas measurements[4,5] taken at $T=300$ K yield $K_2 \simeq -1$ meV/atom.

It is known that the walls between ferromagnetic domains on a surface are of Néel type.[19] This means that the magnetic moments are aligned to the surface plane in order to avoid a perpendicular stray field. If a strong surface uniaxial anisotropy is present, the behavior of

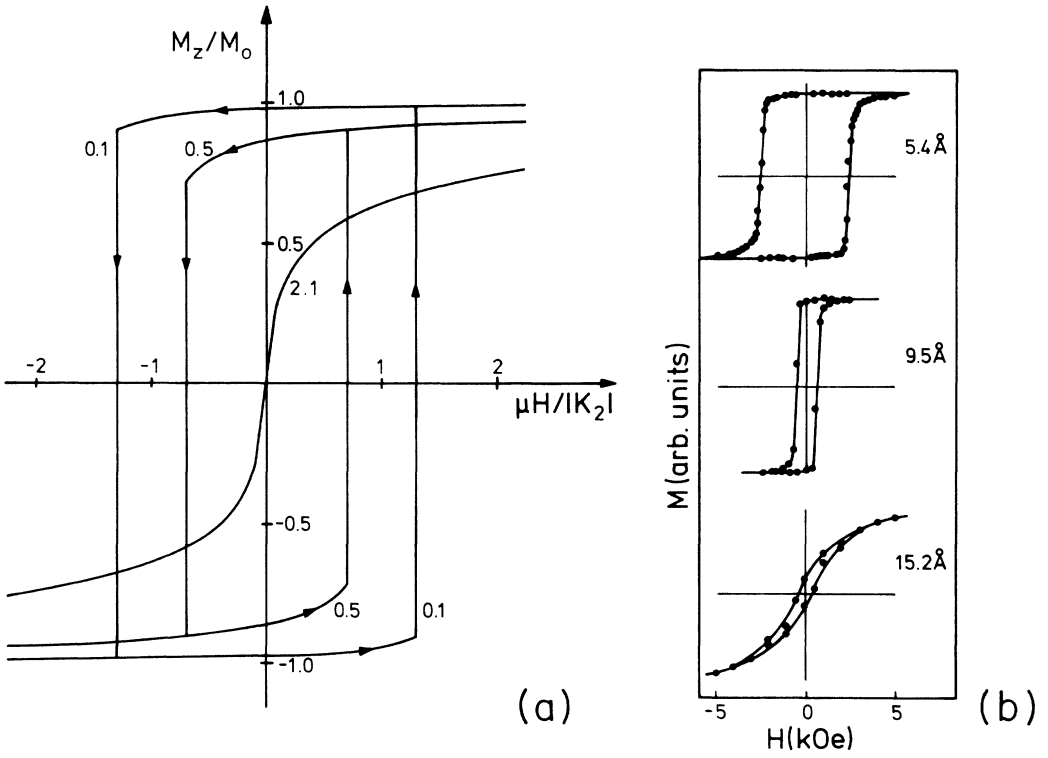

FIG. 4. Comparison of the perpendicular hysteresis loops $M_z(H)$: (a) theoretical results for temperatures $T/\gamma=0.1$, 0.5, and 2.1 obtained by Eq. (3.4); (b) experimental results for the system Au/Co/Au(111) by Chappert and Bruno for different film thicknesses and $T=10$ K (Ref. 8).

domain walls might be different. Therefore, it may be possible that for $T > T_{\parallel}$ the magnetization maintains vertical components in the domain walls because of local weakening of the spin-spin interaction [Eq. (2.3)], i.e., $T_{\parallel}^{\text{wall}} > T_{\parallel}$. In contrast, the magnetization in the interior of domains are oriented in-plane above $T_{\parallel}$.

We considered in our model the direct temperature dependence of the surface magnetic anisotropy, i.e., the decrease of the ferromagnetic ordering (which turns out to be not so decisive) and the variation of its direction. The parameters K_2 and K_4 are taken as constants. However, we emphasize that an indirect temperature dependence of these parameters may still be present, e.g., through the thermal lattice expansion, roughness effects, or segregation. This in turn can seriously affect the values of K_2 and K_4 and may even change their sign.[5] Also collective fluctuations of the magnetization (surface spin waves) are not considered here. These effects have to be taken into account for a complete determination of the temperature dependent magnetic anisotropy. The effect of interface roughness and lattice mismatch on the shape, as well as on the uniaxial anisotropy were treated within a phenomenological model.[8] It produces a contribution to the K_2 term in Eq. (2.4) like the also-not-considered magnetostriction. Surface roughness always diminishes the shape anisotropy.

Besides the thermal dependence the anisotropy parameters depend strongly on changes of the local environment which lead to the confusing variety of surface magnetic structures. Therefore, it is possible for certain systems that their uniaxial anisotropy is strong enough to maintain a magnetization with vertical components also at comparable high temperatures ($T_{\parallel} \lesssim 400$ K) and thick films, e.g., Fe/Cu(100) (Refs. 2 and 3) or Au/Co/Au(111) (Ref. 8).

For very thin films the Curie temperature T_C may become very low[2,6] and comparable with $T_{\parallel}$. In this case the itinerant nature of magnetism becomes important and the picture of localized magnetic moments is no longer valid. In addition, strong fluctuations of the magnetization are present. Therefore, we conclude that in such a case our approach should not be used.

Theoretical calculations predict an enhanced magnetic moment for ferromagnetic surfaces,[20] whereas from the experimental side the situation is not clear.[21] We assumed the same magnetic moment throughout the film and also a uniform exchange coupling constant. It is possible for our model to take into account a variation of the coupling constant and the amount of magnetic moments. However, we emphasize that such a large reduction of coupling constants near surfaces, as recently assumed to predict noncollinear surface magnetic structures,[22] is unlikely for the systems considered here.

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APPENDIX A

Magnetic uniaxial anisotropies are found in hcp lattices or on surfaces or interfaces and layered structures. Callen *et al.*[14,15] considered $\mathcal{H}'_{so}$ [Eq. (2.4)] as a small perturbation of the exchange coupling. For this the polar axis is transformed using spherical harmonics $Y_l^m(\cos\theta_M)$ from the surface normal (or c axis in a hcp lattice) to the direction of magnetization $\mathbf{M}$. Then the Hamiltonian is solved within a molecular field approximation, leading to a free energy with temperature dependent anisotropy coefficients $A_l(T)$:

$$F(T) = F_0(T) + A_2(T)\cos^2\theta_M + A_4(T)\cos^4\theta_M \ . \quad \text{(A1)}$$

The expectation value $\langle \theta_M \rangle$ is determined through $\partial F/\partial\theta_M = 0$ and given by

$$\cos^2\langle \theta_M \rangle = -A_2(T)/2A_4(T) \ . \quad \text{(A2)}$$

The temperature dependence of the $A_l(T)$ enter in *only* through the average magnetization $|\mathbf{M}(T)|$. For low temperatures ($T \ll T_C$) one can show that

$$\frac{A_l(T)}{A_l(T=0)} \propto |\mathbf{M}(T)|^{l(l+1)/2} \ . \quad \text{(A3)}$$

Therefore, $A_2(T) \propto |\mathbf{M}(T)|^3$ and $A_4(T) \propto |\mathbf{M}(T)|^{10}$, $A_l(T) \equiv 0$ for $T > T_C$. The sign of the $A_l(T)$ are determined by the sign of the anisotropy constants K_l: $\operatorname{sgn} A_4(T) = \operatorname{sgn} K_4$, and for $K_4 = 0$: $\operatorname{sgn} A_2(T) = \operatorname{sgn} K_2$. The $A_l(T)$ are only minor affected by external fields.

APPENDIX B

Here we like to examine the validity of the approximation for the partition function Eq. (2.9) used in this work. We restrict the calculation to the case of a pure external field:

$$\mathcal{H}_f = -\boldsymbol{\mu}\mathbf{H} = -\mu H u \ , \quad \text{(B1)}$$

with $u = \cos\theta$. The exact result of the average value $\langle u \rangle$ is the well-known Langevin expression

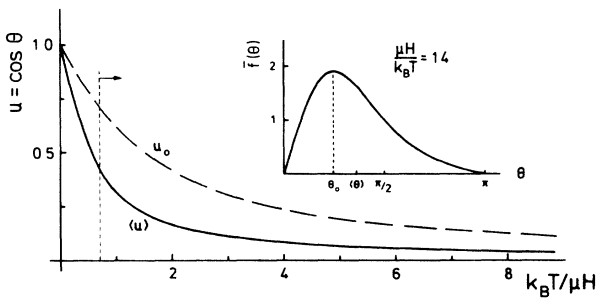

FIG. 5. Expectation values for $u = \cos\theta$ for a classical spin $\boldsymbol{\mu}$ in an external magnetic field $\mathbf{H}$. The value $u_0 = \cos\theta_0$ obtained by the approximation Eq. (2.9) (dashed line) is compared with the exact solution $\langle u \rangle$ (solid line). The insert shows the statistical weight $\bar{f}(\theta) = \sin\theta\, e^{\mu H \cos\theta / k_B T}$ for $\mu H/k_B T = 1.4$, the values θ_0 and $\langle\theta\rangle = \arccos\langle u \rangle$ are indicated.

$$\langle u \rangle = \coth(\mu H/k_B T) - k_B T/\mu H \ . \tag{B2}$$

Using our approximation Eq. (2.9) we obtain for the partition function

$$Z_0 = \mathrm{Tr} e^{-\mathcal{H}_f/k_B T} \delta(\theta_0 - \theta) \simeq 2\pi \sqrt{1-u_0^2}\, e^{\mu H u_0/k_B T} , \tag{B3}$$

$u_0 = \cos\theta_0$ is determined by minimizing the respective free energy:

$$u_0 = \left[\left(\frac{k_B T}{2\mu H} \right)^2 + 1 \right]^{1/2} - \frac{k_B T}{2\mu H} \ . \tag{B4}$$

The two curves for $\langle u \rangle$ and u_0 are depicted in Fig. 5. The behavior for $T \gtrsim 0$ and for $T \to \infty$ is the same besides constant factors. In the intermediate temperature range large quantitative deviations are present. Nevertheless we assume this approximation to be a reliable qualitative estimation for the temperature dependence of $u = \cos\theta$.

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$$\mathcal{H}'_{ss} = \frac{\mu_0}{2} \sum_{i,j} \left[\frac{\boldsymbol{\mu}_i \boldsymbol{\mu}_j}{r^3} - 3 \frac{(\boldsymbol{\mu}_i \mathbf{r})(\boldsymbol{\mu}_j \mathbf{r})}{r^5} \right] ,$$

which can be put into the energy per unit area

$$\mathcal{H}_{ss} = \alpha d \langle u \rangle (B_1 \cos\theta + B_2 \xi \cos\theta_M) \ .$$

The geometrical factors B_1 and B_2 are lattice sums and depend on the lattice symmetry. For thick films B_2 approaches unity, the value obtained by a continuum model [cf. Eqs. (2.3) and (2.3′)].

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