Roughness-induced variation of magnetic anisotropy in ultrathin epitaxial films: The undulating limit

C. A. F. Vaz,* S. J. Steinmuller, and J. A. C. Bland
Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge, CB3 0HE, United Kingdom
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We identify an extra magnetic anisotropy contribution due to roughness in ultrathin single crystalline magnetic films grown on randomly corrugated surfaces. In this picture, the magnetization follows the roughness profile leading to a tilting of the magnetization away from the easy magnetization axis and hence to a modification of the magnetic anisotropy. This "undulating" magnetic state competes with the alternative possibility of uniform in-plane magnetization, which carries a cost in magnetostatic energy due to surface charges. The limits of validity of the model presented are discussed in terms of the roughness amplitude, correlation length, film thickness, and the intrinsic magnetic parameters of the film.

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It has been widely recognized that surface roughness in thin magnetic films leads to an extra magnetostatic energy contribution to the magnetic energy via the so-called "orange-peel" effect,[1] resulting, for example, in changes in the perpendicular magnetic anisotropy,[2–6] extra anisotropy terms in surfaces with directional morphology,[2,7–10] and additional magnetic coupling in multilayer systems.[11–20] Another roughness contribution to the magnetic anisotropy can be envisaged which may be important in ultrathin epitaxial films that cover a randomly corrugated substrate surface: While the magnetization follows the roughness profile, reducing the magnetostatic energy contribution, the tilting away from the easy axis of magnetization leads to an additional magnetic anisotropy contribution. This means that in measurements of the magnetic anisotropy, unless the applied magnetic field completely saturates the sample magnetization (which may require fields of the order of a few T for low anisotropy materials), a residual ripple in the magnetization will contribute to the magnetic anisotropy. This assumes that the epitaxial relation between film and substrate is the same on the flat portions of the surface as on the island slopes, as is expected for ultrathin epitaxial films deposited onto a corrugated surface (Fig. 1, left).[21] Further, we can expect this mechanism to dominate at small thicknesses, while at larger thicknesses the linear increase in exchange and anisotropy energy which is associated with such a magnetic configuration (including a volume magnetostatic energy contribution that results from a slight divergence of the magnetization) will exceed the magnetostatic energy associated with the "orange-peel" configuration, whereby the system is expected to fall into the usual uniform magnetic state (Fig. 1, right). We propose here that this mechanism may be an important contribution to the effective magnetic anisotropy found in single crystalline ultrathin magnetic films, and we calculate by means of a simple model analytical expressions for the effect of roughness on the effective magnetic anisotropy of uniaxial and cubic anisotropy materials. We also discuss qualitatively the transition point in thickness separating this regime from that where the usual "orange-peel" effect contribution sets in.

To estimate the magnetic energy associated with an undulating magnetization, we consider a thin magnetic layer covering a nonmagnetic island, which we assume to have cylindrical symmetry and, in the simplest case, to have a conical shape. Surface roughness is characterized in a first approximation by two parameters, the roughness amplitude σ and the correlation length ξ, which corresponds to the average island size (see Fig. 1); the parameter $\alpha \equiv 4\sigma/\xi$ is a measure of the island slope and is the relevant parameter in problems dealing with surface roughness. The simple conical geometry allows for the analytical calculation of energy expressions for the exchange and magnetocrystalline anisotropy energies, but other shapes can be calculated numerically; as we show, the energy values do not depend significantly on the island shape, indicating that the magnetic energy variation with the roughness parameter is not very sensitive to the exact surface morphology and that our expressions may be used to estimate the variation in anisotropy in real systems.[21] We consider only the case of in-plane magnetized thin film systems with weak anisotropies and we assume that the substrate morphology has no directionality (which otherwise could give rise to extra anisotropy terms[2–4,7,9,10]). Since we assume that the magnetization follows the island surface profile, the magnetization distribution is determined by the substrate surface $S=\{\Phi \equiv \phi(r)-z=0, r \leq R_0\}$ (in cylindrical coordinates, where $R_0 \sim \xi/2$ is the island radius). One requirement is that the magnetization $\mathbf{M}(\mathbf{r})$ is tangent to S, $\nabla\Phi \cdot \mathbf{M}(\mathbf{r})=0$. In order to determine the magnetic anisotropy, we need to calculate the energy difference when the magnetization points (on average) along the hard and easy magnetization axes. We consider first the simpler case of uniaxial anisotropy and next the case of cubic anisotropy.

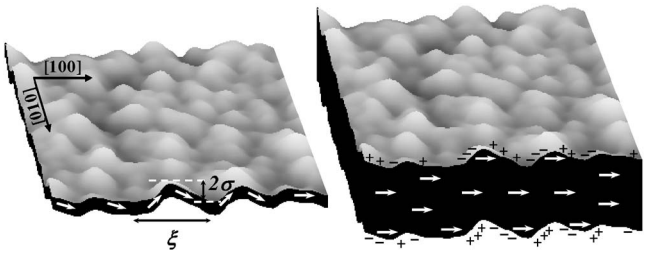

FIG. 1. Schematics of the magnetization profile in a thin and a thick film with coherent interfaces.

For the case of systems with uniaxial anisotropy, we expect no change in energy with roughness when the magnetization lies in the (magnetic hard) plane perpendicular to the easy axis direction, while when the magnetization is aligned (on average) along the easy axis, there is a spread in the magnetization that increases the magnetic energy. Assuming the [100] direction to be the easy axis, we have for the anisotropy energy

$$E_{\text{anis}}^{[100]} = K_u t \int_0^{R_0} r dr \sqrt{1+\phi'^2} \int_0^{2\pi} d\theta \frac{\phi'^2 \cos^2\theta}{1+\phi'^2\cos^2\theta}, \quad (1)$$

where t is the magnetic film thickness, K_u is the uniaxial magnetic anisotropy constant, and $\phi'(r)$ denotes the derivative of $\phi(r)$; this gives for the anisotropy energy density $e_{\text{anis}}^{[100]} = K_u(1-1/\sqrt{1+\alpha^2})$ while along the [010] hard magnetization axis $e_{\text{anis}}^{[010]} = K_u$ and therefore we have for the effective uniaxial anisotropy constant $K_u^{\text{eff}} = e_{\text{anis}}^{[010]} - e_{\text{anis}}^{[100]} = K_u/\sqrt{1+\alpha^2}$. We see that the correction to the effective anisotropy is of second order for small roughness values; we see also that the effect of roughness in this model is always to decrease the effective magnetic anisotropy.

We consider now the more complicated but more interesting case of in-plane cubic anisotropy. We assume for concreteness that the [100] and [110] directions correspond, respectively, to the hard and easy directions (i.e., cubic anisotropy constant $K_1 < 0$) of magnetization in the (001) plane of cubic materials. The relevant quantity is now the energy difference $e_{\text{anis}}^{[110]} - e_{\text{anis}}^{[100]}$, which for a perfect planar film gives $K_1/4$. We next calculate this energy difference as a function of the roughness parameter.

(a) Magnetic anisotropy along the [100] *direction*. We calculate the resultant anisotropy energy $E_{\text{anis}} = K_1 \int \alpha_2^2 \alpha_3^2 dv$, where we assume that the magnetization lies in the (010) plane such as one would expect upon saturation along the [100] direction. We have

$$E_{\text{anis}}^{[100]} = K_1 t \int_0^{R_0} r dr \sqrt{1+\phi'^2} \int_0^{2\pi} d\theta \frac{\phi'^2 \cos^2\theta}{(1+\phi'^2\cos^2\theta)^2} \quad (2)$$

which depends only on the parameter $\alpha = 4\sigma/\xi$. For a general surface $z = \phi(r)$ Eq. (2) is very difficult to calculate analytically or, if possible, it is likely to be too complicated to be useful; however, the case of a conical surface is easy to calculate and yields

$$e_{\text{anis}}^{[100]} = \frac{K_1}{\pi} \frac{\alpha^2}{1+\alpha^2} \left[\frac{1}{2+\alpha^2} + \frac{\arctan(1+\alpha^2)^{-1/2}}{\sqrt{1+\alpha^2}} \right]. \quad (3)$$

In the inset to Fig. 2 we plot the roughness anisotropy energy density contribution (normalized to K_1) as a function of the roughness parameter $\alpha = 4\sigma/\xi$ and we compare it with numerical results for more realistic island shapes, Gaussian and sinusoidal, showing that, in fact, the change in energy is not very sensitive to the exact island shape. The energy reaches a minimum (for $K_1 < 0$) at $\alpha = 1$, which we expect since it corresponds to an island slope of 45°, along which direction the magnetocrystalline anisotropy is lowest in the (100) plane.

(b) Magnetic anisotropy along the [110] *direction*. This problem is identical to that solved previously, but now we require the magnetization to lie in the $(\bar{1}10)$ plane or, equivalently, that $\mathbf{M} \in S$ and $\mathbf{M}\cdot\mathbf{n} = 0$ where $\mathbf{n}$ is a vector normal to the $(\bar{1}10)$ plane. These conditions allow us to write the magnetization as a function of the local coordinates; the magnetic energy is now $E_{\text{anis}} = K_1 \int \Sigma \alpha_i^2 \alpha_j^2 dv$ $(i > j)$, since now all the magnetization components are nonzero in the general case. We can then show that

$$E_{\text{anis}}^{[110]} = \frac{\pi K_1 t}{4} \left[R_0^2 + 3 \int_0^{R_0} dr \frac{r\phi'^2}{1+\phi'^2} \right]. \quad (4)$$

Again, the case of a conical surface can be solved immediately, giving for the energy density

$$e_{\text{anis}}^{[110]} = \frac{K_1}{4} \frac{1}{\sqrt{1+\alpha^2}} \left[1 + \frac{3}{2} \frac{\alpha^2}{1+\alpha^2} \right]. \quad (5)$$

We see in particular that $e_{\text{anis}}^{[110]} = K_1/4$ for $\alpha = 0$, as expected. In the inset to Fig. 2 we show the normalized roughness anisotropy energy density variation with the roughness parameter α along the [110] direction. It is seen that the effect of roughness is to decrease the magnetic anisotropy energy, since more spins now point closer to the [111] direction; for very large roughnesses, however, a larger portion of the spins will point along the [001] direction and therefore the magnetic energy gradually increases to zero $(K_1 < 0)$.

We can now calculate the effective magnetic anisotropy associated with film roughness from the above expressions; since the resulting expressions cannot be simplified beyond their initial form, we shall consider here only their graphical representation, shown in Fig. 2; it shows that the effect of roughness is, within the model proposed here, to decrease the effective magnetocrystalline anisotropy. This decrease is quite significant for large values of α. We should point out that, although we plotted $4\sigma/\xi$ up to 2, such large values are

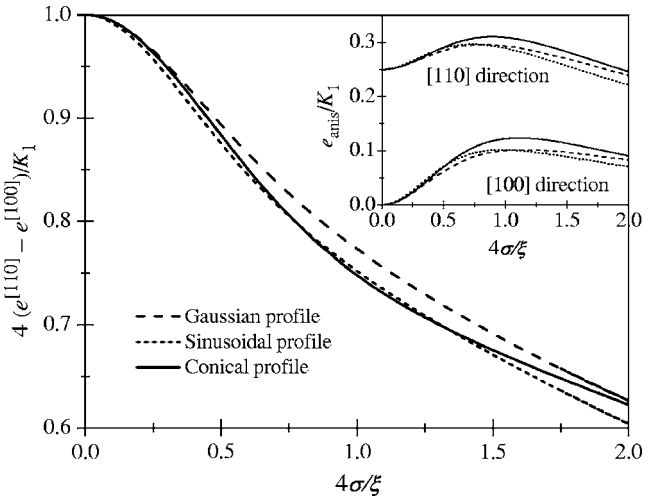

FIG. 2. Variation of the normalized energy density difference with the roughness parameter $4\sigma/\xi$ for several surface profiles. The inset shows the variation of the normalized energy with the roughness parameter $4\sigma/\xi$ for several surface profiles, with the magnetization lying in the (010) and $(\bar{1}10)$ planes.

rather unrealistic, and we expect in all practical cases the latter parameter to be limited up to 0.3 for very rough films; for this value of $4\sigma/\xi$, the effective anisotropy is reduced by $\sim 5\%$, which corresponds to a large change in the measured anisotropy with respect to that of a perfectly flat film.

This reduction in the effective anisotropy is a result of the fact that the roughness-induced anisotropy energy behaves differently along the easy and hard axes; in fact, if the changes in energy with roughness were identical, no variation in the effective magnetic anisotropy would ensue (see inset of Fig. 2).

The remaining question is to determine for which set of sample parameters (t, σ, and ξ) one expects this model for the magnetization configuration to be applicable as opposed to the case of uniform magnetization. Intuitively, one expects the present model to apply for relatively large values of α and small t; the uniform case should be more favorable for small α and large t. Unfortunately the problem is too complicated to allow us a complete study, but we can nevertheless discuss the relative size of the different energy terms involved and the limits for which we expect our model to apply.

For the case of the undulating magnetization profile (which for simplicity we call the "undulating" state, Fig. 1, left), we have both a contribution from the anisotropy and exchange energy; the magnetostatic energy is nonzero, *stricto sensu*: Although there are no surface "magnetic pole charges," there is a volume charge distribution arising from a nonvanishing divergence of the magnetization. The calculation of this energy term is exceedingly complicated, and this term needs to be taken into account in estimating the transition thickness that separates these two magnetic states. The case of uniform magnetization has been widely studied in the literature for $\sigma \ll t$, for instance, by Zhao *et al.*,[5] who provide a convenient expression for the magnetostatic energy of a magnetic film with coherent interfaces. If the demagnetizing energy of the undulating state is not taken into account, the model predicts that the uniform state is higher in energy for an unphysically wide range of thicknesses. This is an indication of the importance of the magnetostatic energy term in determining the details of the spin state of lowest energy in ultrathin films and nanostructures.[22] For ξ comparable to or smaller than the exchange length, the undulating state leads to a very large exchange energy, suggesting that at such short length scales a local uniform magnetization configuration ensues, i.e., the undulating state is expected to be a "macroscopic" state, insensitive to the nanoscale variations in surface roughness (such small scale roughness could give rise to step anisotropies,[23,24] but this should not affect the effective anisotropy for a random roughness profile). For very small roughness values these two different states (uniform and undulating) are largely undistinguishable, but for larger roughness amplitudes and small thicknesses, we expect the undulating state to be lowest in energy.

Variations in the uniaxial, growth-induced, anisotropy of Co films as a function of film roughness have been reported by Li *et al.*,[25] who find that for very large substrate roughnesses ($\alpha \sim 0.4$) the anisotropy vanishes. However, the Co films in this earlier study are very thick (97 nm), are not single crystalline, and the rougher films do not seem to be conformal with the substrate. In the work by Li *et al.* the vanishing of the anisotropy must therefore be due to other factors, such as morphology-induced changes in growth-induced anisotropy. More recently, experimental results on the magnetic anisotropy of fcc Co films grown on rough Cu(001) substrate layers suggest a reduction in the effective anisotropy constant with increasing film roughness that can be accounted for by the present model.[21] In fact, it is found that 7 and 17 nm thick fcc Co(001) films exhibit, respectively, a reduction in magnetic anisotropy with roughness ($\alpha \sim 0.03$ and 0.3 for the smooth and rough films, respectively) of approximately 25 and 8 % while our model predicts a change in anisotropy of about 4.3%. Since real surfaces tend to be more complex than the uniform distribution of islands assumed in our model,[26–28] an exact numeric comparison of the change in magnetic anisotropy with roughness is difficult. However, our model explains well the sign and magnitude of such changes;[21] accuracy may be improved by considering a weighted expansion in the surface roughness components, if known. Our work implies that the substrate roughness may introduce changes in the effective magnetic anisotropy of ultrathin films and that, conversely, a control of the magnetic anisotropy of ultrathin films must take into account roughness effects.

In conclusion, we suggest the existence of a magnetic state for ultrathin films grown on substrates with a significant surface roughness, whereby the magnetization follows the substrate profile in order to avoid the formation of surface charge poles. This leads to a reduction in the magnetic anisotropy and we develop a model that allows one to estimate (by excess) the corresponding change in the effective magnetic anisotropy constant.

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*Author to whom correspondence should be addressed. Present address: Applied Physics, Yale University, New Haven, Connecticut 06520.

[1] L. Néel, Compt. Rend. **255**, 1545 (1962).
[2] E. Schlömann, J. Appl. Phys. **41**, 1617 (1970).
[3] P. Bruno, J. Phys. F: Met. Phys. **18**, 1291 (1988).
[4] P. Bruno, J. Appl. Phys. **64**, 3153 (1988).
[5] Y.-P. Zhao, G. Palasantzas, G.-C. Wang, and J. T. M. De Hosson, Phys. Rev. B **60**, 1216 (1999).
[6] S.-B. Choe and S.-C. Shin, J. Magn. Magn. Mater. **221**, 255 (2000).
[7] E. Schlömann and R. I. Joseph, J. Appl. Phys. **41**, 1336 (1970).
[8] Y. Park, E. E. Fullerton, and S. D. Bader, Appl. Phys. Lett. **66**, 2140 (1995).
[9] R. Arias and D. L. Mills, Phys. Rev. B **59**, 11871 (1999).
[10] J. H. Wolfe, R. K. Kawakami, W. L. Ling, Z. Q. Qiu, R. Arias,

and D. L. Mills, J. Magn. Magn. Mater. **232**, 36 (2001).
[11] L. Néel, Compt. Rend. **255**, 1676 (1962).
[12] E. W. Hill, S. L. Thomlinson, and J. P. Li, J. Appl. Phys. **73**, 5978 (1993).
[13] D. Altbir, M. Kiwi, R. Ramírez, and I. K. Schuller, J. Magn. Magn. Mater. **149**, L246 (1995).
[14] D. Wei and H. N. Bertram, IEEE Trans. Magn. **32**, 3434 (1996).
[15] J. L. Leal and M. H. Kryder, IEEE Trans. Magn. **32**, 4642 (1996).
[16] D.-H. Han, J.-G. Zhu, and J. H. Judy, J. Appl. Phys. **81**, 4996 (1997).
[17] B. D. Schrag *et al.*, Appl. Phys. Lett. **77**, 2373 (2000).
[18] H. D. Chopra, D. X. Yang, P. J. Chen, D. C. Parks, and W. F. Egelhoff, Jr., Phys. Rev. B **61**, 9642 (2000).
[19] C. Tiusan, M. Hehn, and K. Ounadjela, Eur. Phys. J. B **26**, 431 (2002).
[20] W. F. Egelhoff, Jr., R. D. McMichael, C. L. Dennis, M. D. Stiles, A. J. Shapiro, B. B. Maranville, and C. J. Powell, Appl. Phys. Lett. **88**, 162508 (2006).
[21] S. J. Steinmuller, C. A. F. Vaz, V. Ström, C. Moutafis, C. M. Gürtler, M. Kläui, J. A. C. Bland, and Z. Cui, J. Appl. Phys. (to be published).
[22] C. A. F. Vaz, C. Athanasiou, J. A. C. Bland, and G. Rowlands, Phys. Rev. B **73**, 054411 (2006).
[23] D. S. Chuang, C. A. Ballentine, and R. C. O'Handley, Phys. Rev. B **49**, 15084 (1994).
[24] A. Moschel, R. A. Hyman, A. Zangwill, and M. D. Stiles, Phys. Rev. Lett. **77**, 3653 (1996).
[25] M. Li, G.-C. Wang, and H.-G. Min, J. Appl. Phys. **83**, 5313 (1998).
[26] J. M. Bennett and L. Mattson, *Introduction to Surface Roughness and Scattering* (Optical Society of America, Washington, D.C., 1999).
[27] C. A. F. Vaz, G. Lauhoff, J. A. C. Bland, S. Langridge, D. G. Bucknall, J. Penfold, J. Clarke, S. K. Halder, and B. K. Tanner, J. Magn. Magn. Mater. (to be published).
[28] C. A. F. Vaz, S. J. Steinmuller, C. Moutafis, J. A. C. Bland, and A. Yu. Babkevich, Surf. Sci. **601**, 1377 (2007).