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Magnetization of ultrathin ferromagnetic films at finite temperatures

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Abstract. The in-plane magnetization of a two-dimensional film may be stabilized by either dipolar interactions or by an in-plane anisotropy in the absence of an external field. We have calculated the conditions for either of these effects to dominate and show that there is only a restricted range of q values in which dipolar effects are important before the exchange dominates. We report measurements of all relevant anisotropy fields for an epitaxial Ag/2 ML Co/Ag(001) film structurally characterized by angle-resolved Auger spectroscopy and low-energy electron diffraction, together with a measurement of the ground state moment per Co atom (enhanced from the bulk value) using polarized neutron reflection. The results allow us to extend our previous study of the temperature dependence of the magnetization by including the perpendicular fields and the field dependence of the magnetization at 300 K. We show that the field dependence of the magnetization is not consistent with a spin wave gap due to dipolar interactions but is consistent with an anisotropy-induced spin wave gap, confirming the results of a preliminary study.

1. Introduction

The stabilization of long-range ferromagnetic order in 2D has remained an important issue since Mermin and Wagner [1] showed that long-range order which breaks a continuous symmetry cannot be sustained in a system with short-ranged spin interactions. The observation of a robust magnetism [2, 3] in monolayer films may be due either to the occurrence of anisotropy which breaks the continuous symmetry or to long-ranged dipolar effects.

For a film with a uniaxial anisotropy favouring a perpendicular easy magnetization axis, long-range ferromagnetic order can exist at finite temperatures since the anisotropy gives rise in this case to a significant spin wave gap [3, 4], but no such gap arises for the case in which an in-plane easy magnetization is favoured. The problem of the thermal stability of the magnetization has attracted renewed interest since recent investigations of ultrathin epitaxial films establish the existence of long-range magnetic order in plane which is surprisingly robust [5, 6]. For the case of a uniaxial anisotropy favouring in-plane magnetization it has been recently proposed that an effective gap arises due to the long-range character of the dipole interaction [6–8]. Bruno [6] finds, for the case of dipolar interactions only, the magnetization of a Co/Cu(001) monolayer is reduced by ~30% at 100 K. This raises an important question concerning the strength of the in-plane anisotropy required to dominate

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over dipolar effects in controlling the spin wave gap. Recently, Krams *et al* [9] have shown that the magnetocrystalline anisotropy and ferromagnetism both vanish at the same critical thickness in ultrathin fcc Co/Cu films, suggesting that the magnetocrystalline anisotropy is responsible for the stabilization of long-range ferromagnetic order.

In a previous study [10] we measured the in-plane anisotropies and the temperature dependence of the magnetization of an Ag/2 ML Co/Ag(001) structure in an applied magnetic field. We concluded that the temperature dependence of the magnetization was consistent with a spin wave model of the magnetization fluctuations and that the spin wave gap was consistent with that estimated from the magnetocrystalline anisotropies. However, it is important to rule out the possible effect of an applied field in stabilizing the magnetization; moreover, recent studies have shown [11] that large perpendicular anisotropy fields which arise for such ultrathin films can deviate significantly from the bulk values used in our previous study in estimating the spin wave gap. Here we present the results of the two different measurements which show unambiguously that the experimentally observed stabilization of the magnetization of ultrathin Co films on Ag(001) with in-plane remanent magnetization is due to the in-plane magnetocrystalline anisotropy rather than the dipole interactions alone. In one case we have determined all relevant anisotropy fields by MOKE magnetometry and combined these with our extended theoretical analysis to consider the conditions under which dipolar effects are important compared with anisotropy in stabilizing the magnetization. Secondly, we have made additional measurements of the field dependence of the magnetization at 300 K to help rule out the possibility of the applied field stabilizing the magnetization.

2. Theory

Here we present our extended theoretical analysis to examine how dipolar effects and an in-plane anisotropy can stabilize the in-plane magnetization. We first determine a condition that defines the case where dipolar interactions are negligible in comparison with an effective in-plane anisotropy (defined below) in stabilizing the magnetization. Then we determine the temperature dependence of the magnetization from our model and compare this with the expected result when considering dipolar interactions alone.

When the magnetization is close to saturation, its field and temperature dependence is controlled by spin wave excitations as described below. We calculate the effective in-plane anisotropy energy per atom using the expression for the energy of a magnetic film as a function of the magnetization direction. We assume the presence of both twofold and fourfold anisotropies per atom as is appropriate for a cubic film with strain, as used in our experiments.

The out-of-plane (perpendicular) anisotropy is expressed in terms of the modified fourth-order cubic anisotropy K_4', and a second-order uniaxial anisotropy K_2' which includes the effective surface anisotropy term. The anisotropy energy per atom within the plane is given in terms of a modified cubic anisotropy constant K_4 and a uniaxial term K_2 which breaks the fourfold symmetry:

$$E(\theta,\phi) = K_2'\cos^2\theta + \frac{K_4'}{4}\sin^2 2\theta + K_2\sin^2\theta\sin^2\phi + \frac{K_4}{4}\sin^4\theta\sin^2 2\phi. \tag{1}$$

The magnetization lies in plane so the equilibrium angle ϕ_0 and the effective in-plane anisotropy energy K_p may be defined by [12]

$$\left.\frac{\partial E}{\partial \phi}\right|_{\phi_0,\theta=\pi/2} = 0 \tag{2}$$

$$2K_p = \left.\frac{\partial^2 E}{\partial \phi^2}\right|_{\phi_0,\theta=\pi/2}. \tag{3}$$

The following terms contribute to the spin wave energies: isotropic exchange, shape-dependent dipolar interactions, modification of the dipole energies at finite wave vector q (measured in units of $1/a$, where a is the lattice constant of the film), and magnetic anisotropy both perpendicular and in plane. The exchange interaction is by far the largest energy but gives rise to a contribution to the spin wave energies which vary as q^2 for low q. The combination of the shape anisotropy and the perpendicular uniaxial anisotropy which forces the magnetism into the plane is also large but does not create a spin wave gap. The q-dependent dipolar interactions and the in-plane anisotropy are small but are crucially important because the system is not ordered if both of them are absent.

We now investigate the relative importance of the q-dependent dipolar effects and in-plane anisotropy in stabilizing the magnetism for a two-monolayer film. The expression for the spin wave energies may be calculated using the expression from [8] for a film of n monolayers and including the in-plane anisotropy energy [12],

$$\begin{aligned} E_q^2 &= \left\{\varepsilon_q + K_p + K_2' + P_0\left[f - \frac{nq}{4}(1+\cos 2\gamma)\right]\right\}^2 \\ &\quad - \left\{K_p - K_2' - P_0\left[f - \frac{nq}{4}(3-\cos 2\gamma)\right]\right\}^2 \\ &= \left(\varepsilon_q + 2K_p + P_0 nq \sin^2\gamma\right)\left(\varepsilon_q + 2K_2' + 2P_0\left(f - \frac{nq}{2}\right)\right) \end{aligned} \tag{4}$$

where γ is the angle between the magnetization direction and the q vector. ε_q is the exchange contribution to the spin wave energy, and P_0 is the dipolar energy according to the following expressions:

$$\varepsilon_q = Dq^2 \tag{5}$$

$$P_0 = 4\pi\mu_B M_s f. \tag{6}$$

M_s is the saturation magnetization of the film and f is a factor which is necessary to account for the discreteness of the layer. For a two-monolayer fcc film $f \approx 0.883$ [11].

At $q = 0$, equation (4) gives the gap energy E_0:

$$E_0 = \sqrt{4K_{2eff}' K_p} \tag{7}$$

where

$$K_{2eff}' \equiv K_2' + P_0. \tag{8}$$

For finite q both the terms proportional to $P_0 q$ and ε_q contribute; for large values of q, ε_q certainly dominates.

It is important to ask if, as q is increased, the energy passes directly from the anisotropy-dominated regime to the exchange-dominated regime or if a regime exists in which the dipolar terms proportional to $P_0 q$ are dominant. In the low-q regime the largest term in equation (4) is K_{2eff}'.

Hence,

$$E_q^2 \approx 2K_{2eff}'\left[\varepsilon_q + 2K_p + P_0 nq \sin^2\gamma\right]. \tag{9}$$

The values of q such that the in-plane anisotropy contribution is overwhelmed by either the dipolar term or the exchange are written q_d and q_e respectively, since we are looking for

the condition that the dipolar effects are irrelevant we set $\sin^2\gamma = 1$ which is the maximum allowed value. They are defined by

$$2K_p \cong P_0 n q_d \tag{10}$$

$$2K_p \cong \varepsilon_{q_e} = Dq_e^2. \tag{11}$$

At large values of q the exchange term certainly dominates and hence the dipolar term always gives a negligible contribution to equation (4) if $q_d > q_e$. This condition means that at very low q the spin wave energy is dominated by the gap. As q is raised, the exchange regime takes over while the dipolar contribution is small compared with the larger of the exchange or anisotropy energy in all cases. When this condition holds we can safely ignore the q-dependent dipolar energies.

From equations (10) and (11) we can derive a condition for q-dependent dipolar energies to be negligible ($q_d > q_e$) in comparison with the in-plane anisotropy K_p

$$K_p \geqslant \frac{P_0^2 n^2}{2D}. \tag{12}$$

We can evaluate this condition on K_p from the experimentally determined anisotropy constants of our cobalt films (see section 4) and can show that it is easily satisfied. This is a useful result because when the q-dependent dipolar terms are dropped the spin wave energy given in equation (4) depends on q only through ε_q ($= Dq^2$).

$$E_q^2 = \left[\varepsilon_q + K_p + K'_{2eff}\right]^2 - \left[K_p - K'_{2eff}\right]^2. \tag{13}$$

The excitations with q perpendicular to the film can be neglected for ultrathin films because they involve small wavelengths and high energies. In this case the magnetization deviation for a film of n layers and spin s per unit cell calculated within the spin wave theory can be given exactly for any combination of anisotropy constants [13]:

$$\Delta M = \frac{M_0}{2\pi sn} \int_0^\pi \frac{W(q) q \, \mathrm{d}q}{\mathrm{e}^{E_q/kt} - 1} \tag{14}$$

$$W(q) = \frac{\varepsilon_q + K'_{2eff} + K_p}{E_q} \tag{15}$$

where $W(q)$ is the spin wave weight. Using equations (5) and (13) we find

$$\mathrm{d}E_q = \frac{\mathrm{d}E_q}{\mathrm{d}q} \, \mathrm{d}q = 2DqW(q) \, \mathrm{d}q \tag{16}$$

and

$$\frac{\Delta M}{M_0} = \frac{1}{4\pi sDn} \int_{E_0}^{E_\pi} \frac{\mathrm{d}E_q}{\mathrm{e}^{E_q/kt} - 1} = \frac{-kT}{4\pi sDn} \ln\left[\frac{\left(1 - \mathrm{e}^{-E_\pi/kT}\right)}{\left(1 - \mathrm{e}^{-E_0/kT}\right)}\right] \cong \frac{kT}{4\pi sDn} \ln\frac{E_0}{kT} \tag{17}$$

assuming $E_0 \ll kT$ and $E_\pi \gg kT$. It is a special feature of two dimensions that ΔM is exactly integrable and depends only on the maximum and minimum of the energy. The gap energy was given by equation (7). We therefore use our measurements of K'_{2eff} and K_p for the cobalt film described below to fit the observed $M(T)$ for the two models to show that in-plane anisotropies stabilize the in-plane magnetization.

The result for the effect of dipolar interactions alone may be calculated from the spin wave energies in a similar manner [7, 8]. Bruno [6] showed that it may be approximated by an expression like equation (17) with E_0 replaced by

$$E_0 \rightarrow E_{pseudo} = \frac{\sqrt{2}P_0 n}{4}\sqrt{\frac{K'_{2eff}}{D}}. \tag{18}$$

An alternative condition that dipolar interactions may be neglected may be obtained from $E_0 = \sqrt{4K_p K'_{2eff}} > E_{pseudo}$.

3. Sample growth and characterization

The sample was prepared by evaporation in UHV (pressure less than 9×10^{-11} Torr) at 300 K on a single-crystal Ag(001) surface as reported previously [14]. Figure 1 shows the Auger spectrum versus time curve for a typical growth of a Co/Ag(001) sample. After the shutter is opened at t_0 the curve follows an exponential decay until $I/I_0 = 0.77$ after which the curve forms a series of straight sections interrupted by breaks at t_1, t_2 and t_3. The times between the breaks at t_1 and t_2 and between t_2 and t_3 are approximately equal indicating monolayer growth between each break. This means that up until t_1 more than a single monolayer is deposited, assuming one monolayer to mean that all fourfold sites on the Ag surface are occupied by one Co atom. LEED showed a sharp p(1 × 1) pattern for the clean Ag surface which deteriorated slightly as the Co was deposited until the change in the Auger spectrum against time curve at $I/I_0 = 0.77$. It remained as a sharp p(1 × 1) pattern until t_3 (3 ML), after which it began to fade, in agreement with previous results [15] which indicate the formation of a disordered structure above ~3 ML but with a cubic structure forming below that thickness. These results imply that there is some interdiffusion of the Co into the Ag substrate to form an interfacial alloy structure. This was evidenced by the fact that small traces of Co could be observed after most of the Co was removed by a single cycle of Ar^+ bombardment. In fact it took up to four or five Ar^+ bombardment cycles to completely remove all traces of Co. Polarized neutron reflection measurements indicate that the average magnetization is reduced for samples of thickness >3 ML, confirming that a structural change takes place above 3 ML [16], whereas for samples less than 3 ML a significantly enhanced average moment is observed.

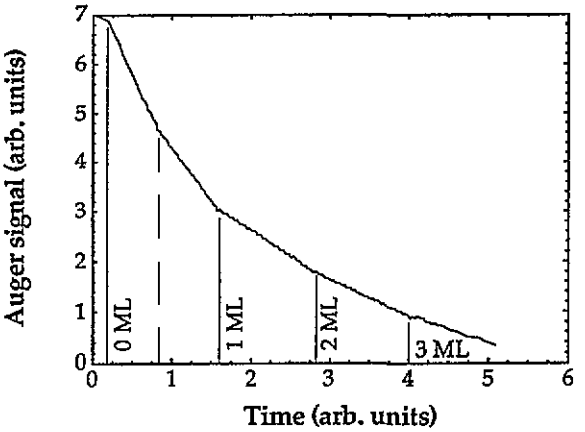

Figure 1. Auger signal–time curve for the growth of Co on Ag(001) showing well defined breaks indicating the completion of successive monolayers.

Angle-resolved Auger measurements of the growth of Co/Ag(001) have been carried out, as shown in figure 2. The measurements for the Ag [100] azimuth show dominant peaks at $\theta = 0°$ and close to 45° with respect to the surface normal, as expected from the forward scattering of electrons by layers of the fcc lattice along the [001] and [101] axes. The [110] azimuth scan is featureless at low coverage indicating no third-layer forward scattering. Our data would therefore suggest that the fcc phase is stabilized for a 2 ML Co thickness. However, Li and Tonner [15] find that for a 3 ML Co/Ag(001) film, the angle-resolved Auger spectra is consistent with a bct structure. This is consistent with the effect of a strong strain influencing the crystal structure for films of this thickness. Whether the film is bct or fcc is not important for our study, but we can conclude that a cubic in-plane anisotropy should arise and that the films are strongly strained.

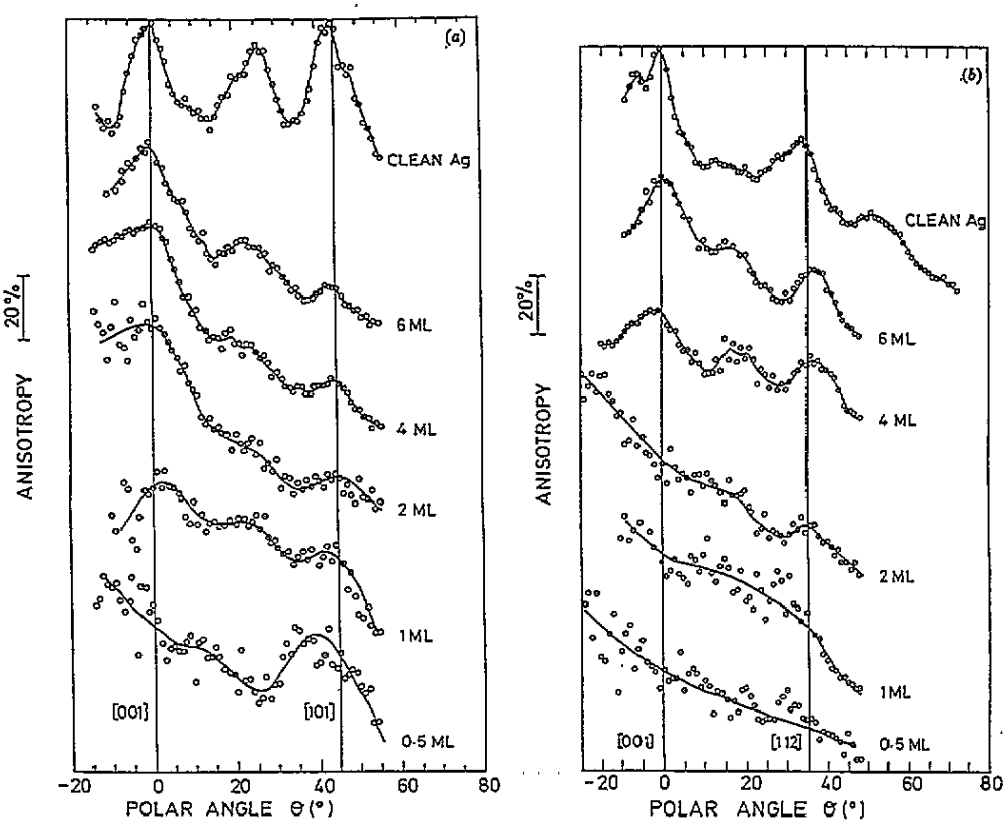

Figure 2. Polar angle Auger electron intensity variation for clean Ag(001) and with various coverages of Co in the [100] Ag azimuth (a), and the [110] Ag azimuth (b).

A conventional longitudinal MOKE arrangement was used to determine the in-plane magnetization curves which were measured at room temperature. An intensity-stabilized HeNe laser of wavelength 632.8 nm and nominal power 10 mW illuminated the sample which was mounted on a rotary stage between the poles of an electromagnet. The sample could be rotated in its own plane in the magnetic field to produce a set of MOKE loops that show the nature of the in-plane anisotropy. A Hall sensor was attached to one of the pole pieces of the electromagnet and calibrated so that the measured Hall voltage could be converted into the value of the field at the centre of the pole pieces where the sample was located. The experiment was controlled by a computer which was used to vary the field

produced by the electromagnet, while recording the Hall voltage and photodiode signal. An intensity stability of better than 0.1% was achieved and was necessary for the sensitive measurements of the 2 ML thick sample anisotropy constants as described later.

Polar MOKE measurements were made using a 7 T superconducting magnet. The sample was placed at the end of an insert tube, close to the centre of the magnet, at room temperature. This geometry was designed so that the laser light did not pass through any windows or lenses in the vicinity of the field, thus eliminating problems due to Faraday rotation or birefringence. Both sets of MOKE data were taken at room temperature.

4. Discussion

We have measured MOKE M–H loops in order to determine all the anisotropy constants of the film. In-plane MOKE measurements give information about K_4 and K_2, while perpendicular MOKE measurements provide information about K'_{2eff}, and the surface anisotropy term K_s. Larger fields were used here compared to our previous study [10], thus permitting a more accurate determination of the anisotropy strengths.

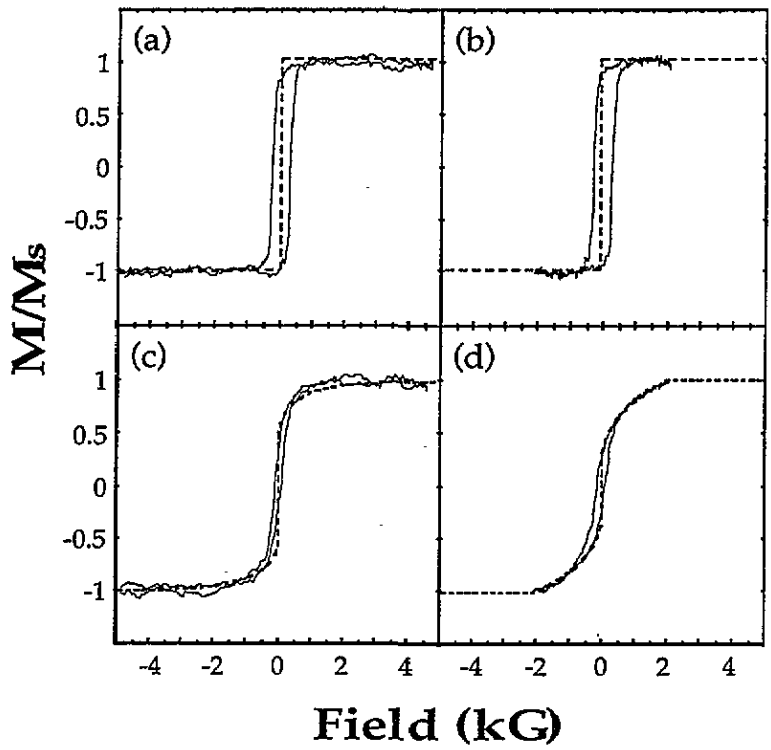

Figure 3. Experimental MOKE loops (solid lines) and calculated M/M_s against field (dashed lines) for the 2 ML Co/Ag film at various in-plane angles relative to the easy axis: (a) −15°; (b) 0° (easy axis); (c) 75°; (d) 90° (hard axis).

Figure 3 shows experimental (solid lines) and calculated (dashed lines) normalized magnetization (M/M_s) against field (H) loops for the 2 ML Co/Ag film for four different in-plane orientations of the applied field relative to the easy axis ($\gamma = -15°$, 0° (easy axis), 75° and 90° (hard axis)). This set of loops clearly confirms the presence of a significant

uniaxial anisotropy with its easy axis loop at $\gamma = 0°$. Detailed fits as described below are required to fit values of the anisotropy constants from this data.

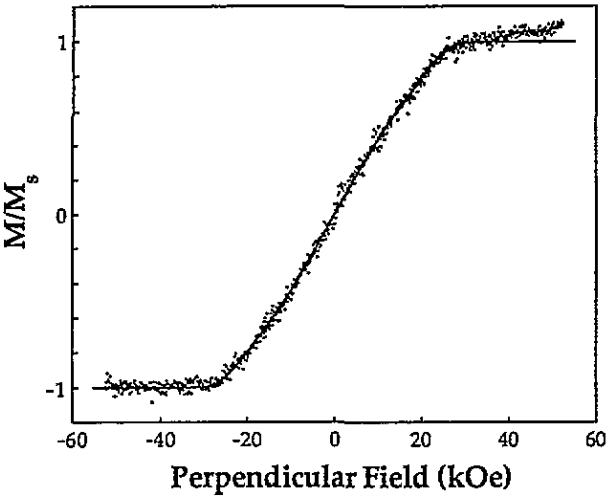

Figure 4. The experimental polar MOKE loop obtained with a superconducting magnet (dots) and calculated M/M_s against field (solid line) for the 2 ML Co/Ag film.

Figure 4 shows the experimental (dots) and calculated (solid line) perpendicular M/M_s against H loop for the same film, corrected for a linear background contribution. The data are well fitted by the calculated curve except at very high positive field. In this region an experimental artefact possibly associated with the magnetooptical signal (rather than the magnetization itself) causes a deviation from the saturation level. The saturation level is clearly seen in the negative field data and therefore the saturation field can be accurately fitted. The large saturation field observed here implies that either the magnetization is enhanced (in line with the PNR measurements), or that a strong surface anisotropy favouring an in-plane orientation is present, or both. An important feature of the data is the curvature of the low-field M–H curve which can be accounted for by the presence of a significant out-of-plane fourth-order anisotropy term K_4'.

The calculations for the fits to both the in-plane and polar MOKE curves were made using a minimum-energy technique [17] that minimizes the total magnetostatic energy as defined by equation (1) as a function of the orientation and strength of the applied field. The values of the anisotropies were determined by fitting the calculated data to the experimental loops, and the fitted parameters are shown in table 1. In addition, an offset of 1° of the applied field direction to the normal of the sample plane was used to correct for misalignment of the sample during the polar MOKE experiment.

A previous polarized neutron reflection measurement [16] gave a value of 2 μ_B for the effective ground state moment per atom (compared with 1.7 μ_B for bulk cobalt) which corresponds to a value for H_d ($=4\pi M_s f$) of 18.5 kOe. Thus the observed perpendicular anisotropy field of 23 kOe which included the contribution of H_d means that $K_2'/\mu_B = 4.5$ kOe. Such strong anisotropies have been observed previously for fcc Co [11]. The existence of a large in-plane uniaxial anisotropy we attribute to the effect of the large in-plane strain in such films, though the exact mechanism is unclear. One possibility is that atomic steps give rise to the observed uniaxial anisotropy or the strain itself is anisotropic. We can rule out the possibility of islands given that a well defined and large perpendicular saturation field is observed, which implies that the demagnetizing factor

Table 1. Magnetic properties of the Ag/2 ML Co/Ag(001) structure. The in-plane anisotropies K_2 and K_4 correspond to values used in fitting the in-plane MOKE data of figure 1, and K'_{2eff} was given by the fit to the polar MOKE data shown in figure 2. The methods of estimating H_{ex} are explained in the text. The anisotropies are given per atom assuming a moment per atom of $2\mu_B$.

K'_2/μ_B	K'_{2eff}/μ_B	K_4/μ_B	K_2/μ_B	$K'_4/2\mu_B$	H_p	H_d	$H_{ex}^{T_c}$	H_{ex}^{D}	f
4.49 kOe	22.97 kOe	−1170 Oe	828 Oe	−3.93 kOe	584 Oe	18.48 kOe	1870 kOe	3729 kOe	0.883

of the full completed film is present.

It is convenient to write the energies appearing in equation (12) as effective fields:

$$H_p = \frac{K_p}{2\mu_B} \qquad H_{ex} = \frac{D}{2\mu_B}. \tag{19}$$

In this notation equation (12) becomes

$$H_p > \frac{H_d^2 n^2}{8H_{ex}}. \tag{20}$$

The value of H_{ex} was estimated to be $H_{ex}^{T_c} \cong 1900$ kOe by Bruno [6] using the measured transition temperature. It may also be estimated from the measured value of D for cobalt [18] of 560 meV Å^2, which leads to the value of $H_{ex}^{D} \cong 3700$ kOe. These estimates are very uncertain because it is not clear how the finite geometry will affect the exchange stiffness. In a Heisenberg model it would depend critically on the range of the exchange interactions (Bruno assumed nearest-neighbour interactions). However, the films are metallic with enhanced moments which may lead to a relatively enhanced value of D. Using these estimates in equation (12) with $n = 2$ we see that the in-plane anisotropy dominates provided that $H_p > 84$ Oe and $H_p > 44$ Oe using the large and small estimates for H_{ex} respectively. These conditions are seen to be both satisfied very easily for our sample.

Figure 5 shows the experimental loop of figure 3(a) (solid line) together with the calculated magnetization deviation (dashed line) as a function of field at 300 K using the anisotropy constants determined from the loop fits, as shown in table 1. The two dots correspond to the magnetization deviation in zero field at 300 K assuming only dipolar interactions. Clearly the observed loop indicates that dipolar interactions alone cannot account for the magnetization deviation, and that anisotropies must also be taken into account.

5. Conclusions

We have shown that the effect of an in-plane anisotropy is dominant in stabilizing the in-plane magnetization of a two-dimensional ferromagnetic film when compared to the effect of dipolar interactions alone. First we derived a condition for when in-plane anisotropy effects should dominate dipolar effects in the spin wave energies. Our measurements of the anisotropies and magnetization for a strained fcc Co/Ag film show that this condition is satisfied, and therefore that anisotropy effects dominate over dipolar effects in stabilizing the magnetization. Measurements of the anisotropy and magnetization are important because these quantities are seen to differ significantly from the bulk values; exact knowledge of them is important in testing the theoretical result. Second we derived an expression for the

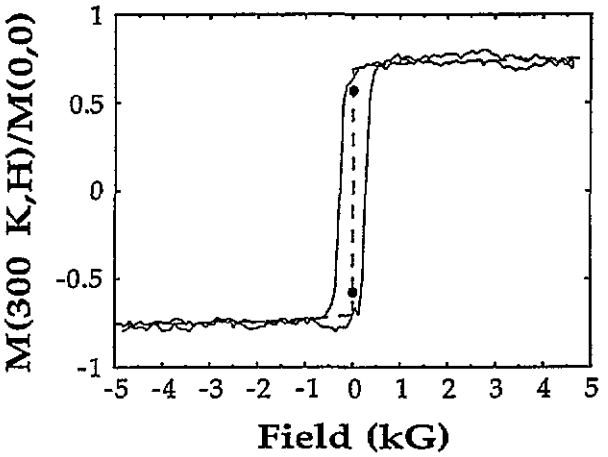

Figure 5. The *M–H* loop obtained from the 2 ML Co/Ag sample at −15° to the easy axis and at 300 K. The dashed line shows the calculated magnetization deviation as a function of field at 300 K using a value for the gap temperature calculated from the measured anisotropies. The two dots correspond to the magnetization deviation expected at zero field and 300 K assuming only dipolar interactions.

magnetization deviation as a function of temperature at zero applied field assuming that the in-plane anisotropy again dominates over dipolar effects. The measured deviation at 300 K and at zero field i s again in agreement with this result.

Acknowledgments

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