Influence of film morphology on perpendicular magnetic anisotropy

J. Camarero,* J. J. de Miguel, and R. Miranda
Departamento de Fsica de la Materia Condensada and Instituto de Ciencia de Materiales "Nicolas Cabrera," Universidad Autsnoma de Madrid, Cantoblanco, E-28049-Madrid, Spain

V. Raposo† and A. Hernando‡
Instituto de Magnetismo Aplicado "Salvador Velayos," UCM-RENFE, P.O. Box 155, Las Rozas, E-28230-Madrid, Spain
(Received 25 February 2001; published 6 September 2001)

Perpendicular magnetic anisotropy (PMA) appears in epitaxial films mainly due to the broken symmetry at the interface. As a result, ultrathin magnetic layers tend to be perpendicularly magnetized. With increasing film thickness, the shape anisotropy overcomes this interface contribution and forces the sample magnetization into the surface plane. We show that previous experimental studies of the magnetic anisotropy energies in the Co/Cu(111) system are affected by the large roughness of the Co films, resulting in underestimated values for the Co-Cu interface anisotropy. By using a surfactant (Pb) to assist the growth of Co layers we are able to prepare Co films and Cu/Co bilayers of homogeneous thickness and negligible roughness, and to determine a more accurate value for the Co-Cu interface anisotropy. With the aid of a simple model calculation, we demonstrate that roughness will substantially affect experimentally determined values of PMA.

DOI: 10.1103/PhysRevB.64.125406 PACS number(s): 68.55.−a, 75.30.Gw, 75.70.Ak, 81.15.−z

I. INTRODUCTION

The existence of magnetic anisotropy, i.e., of special directions (the ''easy'' and ''hard'' axes) for the magnetization is one of the most important characteristics of magnetic materials. This phenomenon is caused by the spin-orbit interaction, which couples the individual magnetic moments in the solid to the crystal lattice.[1] From the point of view of applications, anisotropy is at the base of all relevant processes in ferromagnetic materials, such as the appearance of spontaneous magnetization or its mechanisms of reversal. The total anisotropy energy results from a combination of many different aspects associated with the material structure, such as its shape, its crystalline phase, or the existence of elastic strains.[2]

In thin films it is frequently assumed that the total magnetic anisotropy energy can be phenomenologically divided between a volume and a surface term. The latter was first introduced by Néel,[3] and confirmed experimentally by Gradmann and Müller.[4] It is due to the broken symmetry at the interface. In this way, the anisotropy energy is written as follows:

$$K_{eff}=K_V+nK_S\left(\frac{1}{t}\right), \tag{1}$$

where t is the film thickness and n the number of interfaces (assumed to be identical). K_V is the volume term, including contributions from shape ($K_{sh}=-2\pi M_S^2$, with M_S the saturation magnetization), magnetocrystalline (K_{mc}), and magnetoelastic (K_{me}) anisotropies:

$$K_V=K_{sh}+K_{mc}+K_{me}. \tag{2}$$

In this context, K_S/t is the difference in anisotropy energy between an atom at the surface (or interface) and in the film volume.

Ultrathin magnetic layers with perpendicular magnetization are especially attractive candidates for high-density information storage materials. With conventional longitudinal recording approaching its fundamental superparamagnetic limit, perpendicularly magnetized media are receiving increased attention because of their potential to extend this limit to significantly higher densities due to the use of a thicker recording layer. Besides, data retrieval by means of the magneto-optic Kerr effect is favored because the polar Kerr rotation can be one order of magnitude larger than for longitudinal geometry.[5] The increased signal-to-noise ratio should allow the use of smaller bit sizes. Nevertheless, PMA is only found for a limited number of combinations of magnetic and nonmagnetic materials (for instance, Co with either Pd, Pt, or Cu),[2] and this for the fcc (111) orientation, which presents severe growth difficulties.[6] It is therefore necessary to carefully study the anisotropy characteristics of these materials, especially at room temperature, as a first step towards developing strategies to enhance the effects leading to the appearance of PMA. The interfacial contribution to the magnetic anisotropy can be maximized by preparing multilayers; however, it is crucial to ensure the sharpness of the interfaces and the accurate control of the thicknesses in those samples.

II. EXPERIMENT

In this work, we have used the surface magneto-optic Kerr effect (SMOKE) to study the magnetic properties of epitaxial Co films and {Co/Cu} bilayers grown on Cu(111), paying special attention to the morphological and structural aspects that influence the magnetic anisotropy in these samples. Our experimental system has been described previously.[7] The epitaxial layers are grown by molecular beam epitaxy (MBE) on a single-crystal Cu(111) substrate, either clean or precovered with 1 monolayer (ML) of Pb acting as surfactant.[8] The film thickness and surface ordering are controlled in real time during deposition by monitoring

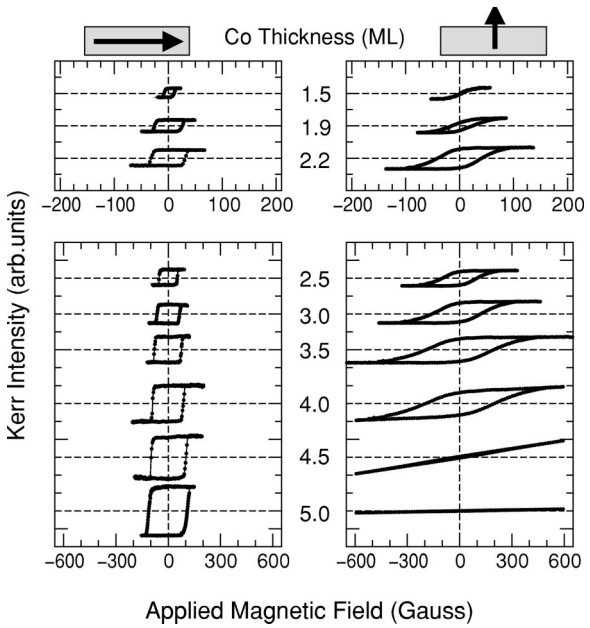

FIG. 1. Kerr effect measurements showing the evolution of magnetic anisotropy as a function of Co thickness t_{Co} (shown in the central column). Thinner films can be magnetized in both directions; with increasing thickness the easy axis lies clearly within the surface plane. All films have been grown using 1 ML of Pb as surfactant.

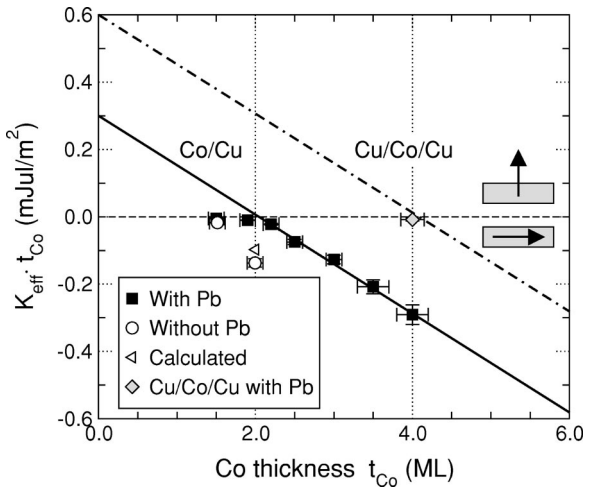

FIG. 2. Uniaxial magnetic anisotropy for Co films of different thicknesses, as determined from Kerr effect measurements. All samples were grown at RT, with (solid squares) or without (open circles) 1 ML Pb as surfactant. The solid line is a least-squares fit to the former data using Eq. (1); the dashed line is the predicted behavior for a Cu/Co/Cu bilayer structure; the diamond is the corresponding experimental data point, confirming the expectation. The triangle is calculated for a Co film grown without surfactant, based on our structural studies of this system.

the specularly reflected intensity of a neutral He beam.[9] After growth, the sample is transferred under ultrahigh vacuum to an auxiliary chamber for the Kerr effect measurements, which are always performed at room temperature (RT).

III. RESULTS

A. Co films grown with the aid of surfactant

We have determined the perpendicular magnetic anisotropy (PMA) in our Co films from sets of hysteresis curves such as those depicted in Fig. 1, in the range of thicknesses where it was possible to saturate the sample magnetization in both polar and longitudinal geometries with the magnetic field available in our setup (± 600 Oe). To do this we proceeded in the usual way, calculating and subtracting the energies needed to magnetize the film along both axes, starting from a demagnetized state:

$$K_{eff} = M_S \left[\int_0^{I_S} H_\perp dI - \int_0^{I_S} H_\parallel dI \right]. \qquad (3)$$

Here, M_S is the saturation magnetization (1442 emu/cm^3 for bulk Co at RT), $H_\perp$ and $H_\parallel$ are the fields applied along the directions normal and parallel to the surface, respectively, and I_S is the normalized saturation Kerr intensity. To obtain correct values for K_{eff} it is necessary to know the relationship between the Kerr intensity measured in our SMOKE experiment and the actual sample magnetization. This is often difficult, but we have shown previously[7] that the saturation Kerr signal of our Co films grown with surfactant increases linearly over an extended range of thicknesses. One can therefore safely assume that the Kerr intensity measured in those films corresponds to the atomic magnetic moment of bulk Co.

From Eq. (3) it follows that a positive anisotropy energy implies out-of-plane magnetization, whereas for $K_{eff} < 0$ the easy axis lies within the film plane. Our results are plotted in Fig. 2: the solid squares correspond to samples grown assisted by 1 ML of Pb as surfactant, while the open circles are for Co films prepared on the clean Cu(111) substrate. All depositions were performed at RT. We will first focus on analyzing the surfactant-assisted films. Their anisotropy energy clearly follows a linear behavior with increasing Co thickness. The straight line is a least-squares fit to the data using Eq. (1). The first two data points (for coverages of 1.5 and 1.9 ML Co, respectively) have been omitted from the fit; they deviate from the expected behavior because for such low thicknesses some patches of the Co film are only 1 ML thick and have Curie temperatures (T_C) below RT.[7,10] Therefore they do not contribute to the magnetic signal detected in our experiment. From the fit we obtain $K_V = -(0.74 \pm 0.04)$ MJ/m^3 and $K_S = (0.32 \pm 0.03)$ mJ/m^2. During surfactant-assisted growth, the Pb layer continuously segregates to the surface of the epitaxial layer. Thus, all these Co films are covered by Pb. We assume that the Co/Pb interface does not contribute to the magnetic anisotropy. We have tested the validity of this hypothesis by depositing 1 ML of Pb on top of a 2.5-ML Co film grown on clean Cu(111). At this coverage, the sample magnetization has just switched from out-of-plane to in-plane. The addition of the Pb-Co interface produces no significant change in the anisotropy energy, thereby justifying our assumption. Hence, the value of K_S must be attributed entirely to the Co-Cu interface.

Table I shows a comparison between our results for the

TABLE I. Comparison of reported values for the different contributions to the magnetic anisotropy in Co/Cu(111) and Cu/Co/Cu(111) systems.

t_{Co} [ML]	t_{Cu} [ML]	K_S [mJ/m^2]	K_V [MJ/m^3]	$K_{mc}+K_{me}$ [MJ/m^3]	Ref.
{2-7}	4	(0.32±0.03)	-(0.74±0.04)	(0.53±0.04)	This work
{2-15}	11	0.10	-0.80	0.47	11
	23	0.12	-0.80	0.47	11
{2-20}	5	0.12	-0.86	0.41	12
{2-20}	-	0.18	-0.64	0.63	13
-	-	0.13	-0.90	0.37	14
{0-17}	14	0.26	-0.60	-	15
		0.4/0.5	0.0	-	15
5.5	-	-0.51	1.34	-	16
5.5	-	0.37	-0.06	-	16
1	-	0.57	-	-	17

different contributions to the magnetic anisotropy and those obtained by other authors.[11–17] Several points deserve being mentioned. First, the sum of the shape ($K_{sh}=-1.27$ MJ/m^3) and bulk magnetocrystalline ($K_{mc}=0.53$ MJ/m^3) (Ref. 18) anisotropies equals exactly our experimental value of the volume term K_V. This indicates that the magnetoelastic term does not make a significant contribution in this case, in good agreement with previous suggestions.[13] This is important for the analysis of our data, because in general K_{me} can also depend on the film thickness through lattice misfit relaxation.[19]

Still more interesting is the fact that the Co-Cu interface anisotropy coming out of our experiments is significantly larger than almost all others reported before[11–14] obtained by the same ''area method.'' The only possible exception[16] will be discussed later. Our experimental result would thus be the closest approximation to the theoretical prediction (0.57 mJ/m^2).[17] This is in all likelihood due to the great improvement in the structural quality of our samples, achieved thanks to the use of the surfactant layer. Systematic studies using scanning tunneling microscopy (STM) and low-energy electron diffraction (LEED) have shown that Co grows on clean Cu(111) in a multilayer fashion, forming pyramids with a large number of exposed atomic levels.[20,21] The large roughness on such samples reduces the influence of the interface anisotropy because the islands actually have heights much larger than the nominal film thickness. It has been argued that roughness could also reduce the shape anisotropy by introducing in-plane demagnetizing fields.[22] However, it seems that at least in this case this effect is negligible. Our own Co films grown without surfactant (open circles, Fig. 2) also show a reduced PMA, similar to the findings by the other authors cited before and consistent with the above discussion.

Accepting that our value for K_S represents the best experimental approximation to the ideal one, it is easy to calculate the effect that adding a second Co-Cu interface (as in a Cu/Co/Cu bilayer) will have on the anisotropy energy. This is represented by the dash-dotted line in Fig. 2, and it predicts that the transition from perpendicular to parallel magnetization should take place at $t_{Co}=4$ ML. This forecast can be readily checked against our own Kerr effect measurements. The results are shown in Fig. 3. They demonstrate that the switching of the magnetization direction does indeed occur at the expected Co coverage. The transition is an abrupt one, and the anisotropy in this case is quite strong: for Co thicknesses up to 3.5 ML, the strongest field available in our experiment is not enough to magnetize the sample parallel to the surface, while at 4 ML the easy axis is already in-plane, with a low saturation field.

The excellent agreement between this experiment and the corresponding prediction lends additional support to several of our previous assertions. First, it confirms that the Pb overlayer directly in contact with the Co film had negligible influence on the PMA. And second, it implies that the second Co-Cu interface is totally equivalent to the first one, both magnetically as well as morphologically. Since the first one was obtained growing Co on the substrate surface, carefully cleaned and ordered, this result provides a striking albeit indirect demonstration of the capabilities of surfactant-assisted growth.

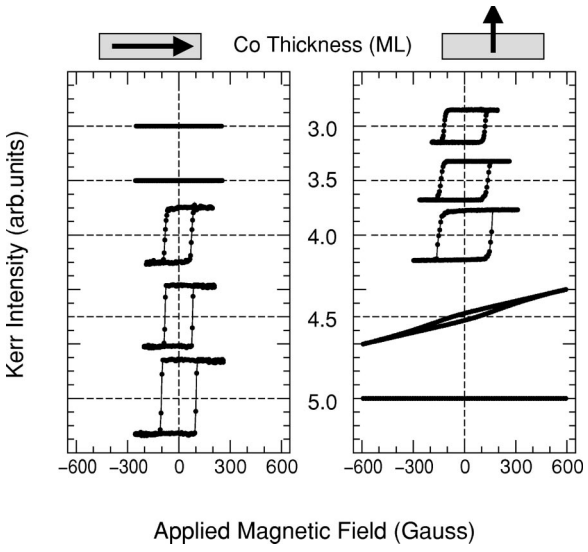

FIG. 3. Transition from out-of-plane to in-plane magnetization as a function of increasing Co thickness, for Cu/Co/Cu(111) bilayer structures.

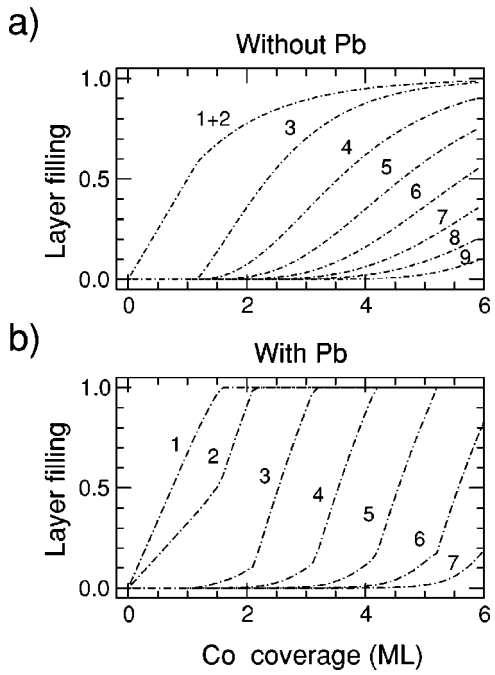

FIG. 4. Evolution of atomic layer fillings during growth of Co on Cu(111) at room temperature: (a) without surfactant, (b) with 1 ML Pb.

B. Co films grown without surfactant

We now turn to analyze the data shown in Fig. 2 for Co films grown without surfactant. Unfortunately, we were not able to obtain more data points; the maximum magnetic field applicable with our setup was not strong enough to measure polar hysteresis cycles for Co films of equivalent thickness higher than 2 ML. To understand these results it is necessary to have detailed knowledge of the sample morphology. We have performed a thorough structural characterization of this system, employing a variety of experimental techniques which include surface x-ray diffraction (SXRD) and thermal energy atom scattering (TEAS).[23] In these experiments, we monitor continuously the evolution of the intensity diffracted by the sample surface, in real time during deposition. Analyzing those data with the aid of a kinetic growth model developed by us[6] and kinematic diffraction theory allows us to determine the gradual filling of atomic levels as growth proceeds. Our results for Co films prepared without and with surfactant are presented in Figs. 4(a) and 4(b), respectively. These graphs completely specify the film morphology at any stage during growth. Clear differences can be seen between them: without surfactant, bilayer islands are formed at the Co-Cu interface, followed by statistical filling of upper atomic levels, corresponding to growth with negligible interlayer diffusion. With surfactant, on the contrary, we find quasi-layer-by-layer growth from the second monolayer on, with a self-replicating front. This ensures that any influence of the sample topology on its magnetic properties will be constant for a wide range of thicknesses.

IV. DISCUSSION

From Fig. 4(a) one can directly obtain the fractional occupation of each atomic level for Co films of any equivalent thickness, grown without surfactant. Then, by applying Eq. (1), one can estimate the anisotropy energy K_{eff} that should be expected for such rough films. We have done so, and the result of this calculation for the 2.0-ML film is shown with a triangle in Fig. 2. Although the calculated value shows the right trend, it still falls short of the experimental value, which reveals a stronger in-plane anisotropy. We believe that an additional contribution might result from dipolar interactions between the islands in the films grown without surfactant, analogous to the so-called ''orange peel'' effect.[24]

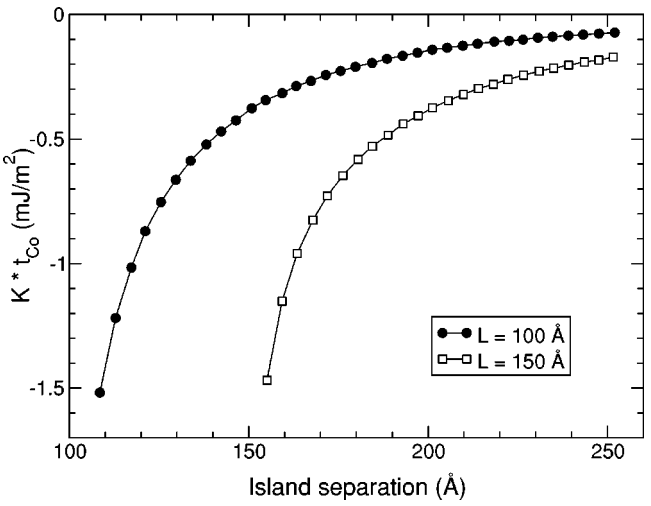

FIG. 5. Calculation of the dipolar magnetostatic interaction between in-plane magnetized Co islands of pyramidal shape, representing a 2-ML-thick Co film grown on Cu(111) without surfactant. L is the base size.

We have performed a simple calculation in order to estimate the magnitude that this interaction can have in samples with typical morphology, as determined from experiments.[20,23] For simplicity, we have considered pyramidal Co islands with square bases of size $(L\times L)$. The calculation is done for an equivalent Co thickness of 2 ML (as in the experiment of Fig. 2), taking the level occupations from Fig. 4(a). The islands are discretized in small cubes of side d situated at positions given by the coordinates (i,j,k). All of them have saturation magnetization of module M_S pointing along directions determined by the unit vectors $\vec{m}_{i,j,k}$. The magnetostatic energy between two islands (u,v) is evaluated as a function of their separation by taking the sum of all the interactions between the cubes forming them:

$$U_{u,v}=M_S^2\sum_{i,j,k}\left[\vec{m}_{ijk}\sum_{i'j'k'}\left(-\frac{\vec{m}_{i'j'k'}}{r^3}+\frac{3(\vec{r}\cdot\vec{m}_{i'j'k'})}{r^5}\vec{r}\right)\right], \tag{4}$$

where $\vec{r}=\vec{r}_{i'j'k'}-\vec{r}_{ijk}$, $r=|\vec{r}|$ and the subindices (i,j,k), (i',j',k') describe positions within the islands u and v, respectively. The results of this calculation for in-plane magnetized Co islands of two typical sizes (100 and 150 Å) are plotted in Fig. 5.

This graph demonstrates that the magnetostatic interaction has the same order of magnitude as the measured anisotropies. This means that the effect of roughness must be taken

into account when evaluating the experimental data. In our case, the difference between the experimental energy for our 2-ML Co film grown without surfactant (open circle in Fig. 2) and the calculated one (triangle) amounts to 40 $\mu J/m^2$. In our model, this is the expected interaction energy between in-plane magnetized islands of 100 Å separated by ~ 300 Å, a typical terrace size for our crystal.[25] Previous reports have already pointed out that the magnetocrystalline anisotropy can be affected by the film's morphology due to the existence of atomic steps[26] or interdiffusion.[27] Our work demonstrates that dipolar magnetostatic interactions can also have a decisive influence on the magnetic properties of rough films.

The reduced K_S values reported in the literature can thus be explained by the existence of this unnoticed interaction; in rough films, grown without surfactant, it will be operative as soon as they become in-plane magnetized. Our model also provides an alternative explanation for the results of Farle *et al.*[16] These authors found a temperature dependence of the anisotropy of their Co films grown on Cu(111) without surfactant. They suggest that this behavior is of thermodynamic origin, and point out that measurements of anisotropy should therefore be performed at constant reduced temperature $\tau = T/T_C$.

In our opinion, though, such a temperature dependence might rather be related to the film roughness. The Curie temperature of ultrathin ferromagnetic films is known to depend on their thickness.[28,29] For Co/Cu(111) most of the evolution towards bulk T_C takes place between 1 and 6 ML.[7,10] Having been grown without surfactant, the Co films studied by Farle *et al*. must contain islands with a wide range of thicknesses, as demonstrated by Fig. 4(a). Co islands with different heights will also have diverse T_C's, which by the way would preclude the definition of a single reduced temperature for this system. Upon lowering the sample temperature, the thinner areas will gradually descend below their Curie temperatures downwards and become ferromagnetic, thereby providing additional and stronger dipolar interactions parallel to the surface plane. This effect can account for the increased in-plane anisotropy at low temperature observed in those experiments.[16]

V. SUMMARY

We have measured the PMA in ultrathin Co films and Cu/Co bilayers epitaxially grown on Cu(111), with and without the aid of a monolayer of Pb acting as surfactant. The presence of the latter induces high-quality layer-by-layer growth of Co films with very homogeneous thickness. From the analysis of our Kerr effect measurements we can determine the different contributions to the films' magnetic anisotropy. Our values for the volume terms are similar to those reported by other authors and coincident with the expectations; for the Co-Cu interface, on the contrary, we obtain a larger anisotropy than other experimental values published to date. We have also demonstrated that those results were strongly affected by the films' roughness. Our work shows that the dipolar magnetostatic interactions between regions of inhomogeneous thickness can have a profound influence on the sample's magnetic anisotropy. This effect has been minimized in our experiments, and therefore our value should be a reliable approximation to the ideal case.

ACKNOWLEDGMENT

This work has been financed by the CICyT through Project No. MAT98-0965-C04-02.

*Present address: Laboratoire Louis Néel, CNRS; BP 166, 38042 Grenoble Cedex, France.

†Also: Instituto de Ciencia de Materiales de Madrid, ICMM-CSIC, Cantoblanco, E-28049-Madrid, Spain.

‡Also: Departamento de Física de Materiales, Universidad Complutense de Madrid, E-28040-Madrid, Spain.

[1] C. Kittel, *Introduction to Solid State Physics*, 6th ed. (Wiley, New York, 1986).

[2] M.T. Johnson, P.J.H. Bloemen, F.J.A. den Broeder, and J.J. de Vries, Rep. Prog. Phys. **59**, 1409 (1996).

[3] L. Néel, J. Phys. Radium **15**, 225 (1954).

[4] U. Gradmann and J. Müller, Phys. Status Solidi **27**, 313 (1968).

[5] S.D. Bader, J. Magn. Magn. Mater. **100**, 441 (1991).

[6] J. Camarero, J. de la Figuera, J.J. de Miguel, R. Miranda, J. Álvarez, and S. Ferrer, Surf. Sci. **459**, 191 (2000).

[7] J. Camarero, J.J. de Miguel, R. Miranda, and A. Hernando, J. Phys.: Condens. Matter **12**, 7713 (2000).

[8] J. Camarero, T. Graf, J.J. de Miguel, R. Miranda, W. Kuch, M. Zharnikov, A. Dittschar, C.M. Schneider, and J. Kirschner, Phys. Rev. Lett. **76**, 4428 (1996).

[9] J.J. de Miguel, A. Cebollada, J.M. Gallego, S. Ferrer, R. Miranda, C.M. Schneider, P. Bressler, J. Garbe, K. Bethke, and J. Kirschner, Surf. Sci. **211/212**, 732 (1989).

[10] A. Díaz-Ortiz, F. Aguilera-Granja, and J.L. Morán-López, Phys. Rev. B **53**, 6514 (1996).

[11] F.J.A. den Broeder, W. Hoving, and P.J.H. Bloemen, J. Magn. Magn. Mater. **93**, 5652 (1991).

[12] Z.S. Shan, S. Nafis, J. Woollan, S.H. Liou, and D.J. Sellmyer, J. Appl. Phys. **73**, 6347 (1993).

[13] J. Kohlhepp, H.J. Elmers, and U. Gradmann, J. Magn. Magn. Mater. **121**, 487 (1993).

[14] B. Hillebrands, P. Krams, J. Fassbender, C. Mathieu, G. Güntherodt, R. Jungblut, and M.T. Johnson, Acta Phys. Pol. A **85**, 179 (1994).

[15] H. Laidler and B.J. Hickey, J. Phys.: Condens. Matter **12**, 8401 (2000).

[16] M. Farle, W. Platow, E. Kosubek, and K. Baberschke, Surf. Sci. **439**, 146 (1999).

[17] G.H.O. Daalderop, P.J. Kelly, and M.F.H. Schuurmans, Phys. Rev. B **42**, 7270 (1990).

[18] W.J.M. de Jonge, P.J.H. Bloemen, and F.J.A. den Broeder, in *Ultrathin Magnetic Structures I*, edited by J.A.C. Bland and B. Heinrich (Berlin, Springer, 1994).

[19] C. Chappert and P. Bruno, J. Appl. Phys. **64**, 5736 (1988).

[20] J.E. Prieto, J. de la Figuera, and R. Miranda, Phys. Rev. B **62**, 2126 (2000).

[21] Ch. Rath, J.E. Prieto, S. Müller, R. Miranda, and K. Heinz, Phys. Rev. B **55**, 10 791 (1997).

[22] P. Bruno, J. Appl. Phys. **64**, 3153 (1988).

[23] J. Camarero, J. de la Figuera, J. Álvarez, S. Ferrer, J.J. de Miguel, and R. Miranda (unpublished).

[24] D. Altbir, M. Kiwi, R. Ramírez, and I.K. Schuller, J. Magn. Magn. Mater **149**, L246 (1995).

[25] The nominal miscut of our crystal is 1°; the equilibrium terrace width is therefore ~120 Å. However, actual terraces are usually larger, due to the existence of step bunching on this surface, as described in Ref. 6. Since our Co films are grown under conditions of high atomic mobility, only a few islands nucleate on each terrace.

[26] P. Bruno, J. Phys. F: Met. Phys. **18**, 1291 (1991).

[27] J.M. MacLaren and R.H. Victoria, J. Appl. Phys. **76**, 6069 (1994).

[28] C.M. Schneider, P. Bressler, P. Schuster, J. Kirschner, J.J. de Miguel, and R. Miranda, Phys. Rev. Lett. **64**, 1059 (1990).

[29] F. Huang, M.T. Kief, G.J. Mankey, and R.F. Willis, Phys. Rev. B **49**, 3962 (1994).