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On tilted magnetization in thin films

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Abstract

We suggest a new mechanism for explaining the tilt of the magnetization away from the surface normal in certain magnetic ultra-thin films. Our arguments are based on a simple classical spin Hamiltonian in which the magnetocrystalline surface anisotropy is described by $H_a = -\sum_i \lambda_i (s_i^z)^2 + \sum_i \gamma_i (s_i^z)^4$, where λ_i and γ_i are non-negative phenomenological constants, s_i^z denotes the z-component (normal to the surface) of the spin at the site labelled by i. In this paper we study only the ground state. In contrast to the usual explanation which attributes the experimentally observed tilted magnetization to the fourth-order term involving γ_i, we show that the second-order term alone can lead to this interesting phenomenon. Our explanation implies that the magnetization of the successive layers are not collinear. As an illustration of our arguments we discuss the experimentally observed orientational transition of the Co/Au(1 1 1) system in quantitative details. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

As is well known by now, ultra-thin films of Fe, Co and Ni on non-magnetic substrates or sandwiched between non-magnetic metals, such as Cu, Ag or Au are frequently magnetized along out-of-plane directions, whereas on the basis of magnetostatic arguments one would expect their magnetization to lie in-plane [1]. Currently, this surprising phenomenon is at the centre of much experimental [1–9] and theoretical [10–15] attention. In this paper we shall study two mechanisms which can give rise to a ground-state magnetization tilted at an angle θ with respect to the surface normal of the film.

This problem is of interest because in many cases, such as Au/Co/Au or Cu/Fe/Cu sandwiches [4–6], where the ground state of a monolayer is

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perpendicularly magnetized and the rotation in-plane, as further layers are added, takes place gradually with the tilt angle θ changing continuously from $\theta = 0$ at $N = 1$ to $\theta = \pi/2$ at the critical thickness of N_c layers. In the face of it, this may be a textbook example of the well-known orientational transition from one magneto-crystalline easy axis (perpendicular) to another one (in-plane) [16]. Indeed, this is the view taken by all the authors who have discussed the matter so far [4–7,12]. In what follows, we shall suggest an alternative explanation which implies a physical picture very different from that of the conventional argument.

As the others [12,15] we shall study a model of classical vector spins $\boldsymbol{\mu}_i = \mu_i \boldsymbol{s}_i$ ($|\boldsymbol{s}_i| = 1$) localized at the lattice sites labelled by i. It is described by the Hamiltonian

$$H = -\frac{1}{2}\sum_{i \neq j} J_{ij} \boldsymbol{s}_i \cdot \boldsymbol{s}_j + \frac{1}{2}\sum_{i \neq j} \omega_{ij}\left(\frac{\boldsymbol{s}_i \cdot \boldsymbol{s}_j}{r_{ij}^3} - 3\frac{(\boldsymbol{s}_i \cdot \boldsymbol{r}_{ij})(\boldsymbol{s}_j \cdot \boldsymbol{r}_{ij})}{r_{ij}^5}\right) - \sum_i \lambda_i (s_i^z)^2 + \sum_i \gamma_i (s_i^z)^4, \quad (1)$$

where the first term is an exchange interaction energy, the second one is due to the dipolar interaction between the spins while the third and the fourth terms describe the magneto-crystalline anisotropy. To simplify matters we shall assume that $J_{ij} = J$ for all nearest neighbor sites and zero otherwise. Similarly, we shall neglect the variation of the magnetic moment μ_i from site to site, even near the surface, and adsorb it into the dipolar coupling constant $\omega_{ij} = \omega = (\mu_0/4\pi)\mu^2/a^3$, where a is the (in-plane) lattice constant. Consequently, the magnitudes of the position vectors $\boldsymbol{r}_{ij}$ are measured in units of a. The single-site magneto-crystalline anisotropy terms have been added to describe the effects of spin–orbit coupling [10]. Due to the reduced symmetry at the surface (interface), the coefficients λ_i are much larger on the surface (interface) than in the bulk [17]. Therefore, we shall assign finite values to them only on the surface (interface) layers. On the contrary, when not zero γ_i will be taken to be the same on all layers. An example of the geometries of interest is illustrated in Fig. 1.

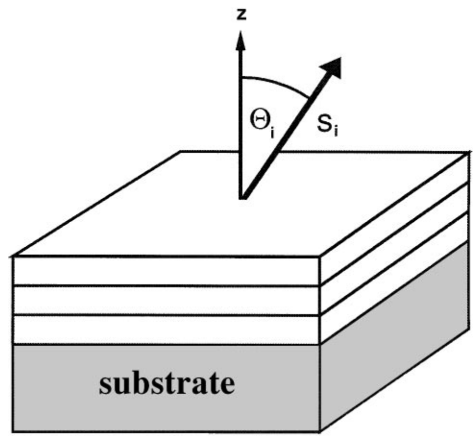

Fig. 1. Scheme of a thin magnetic film on a substrate displaying the convention used for the orientations of magnetization.

Of course, the above model is only a caricature of the real physical situation which concerns itinerant electrons. Nevertheless, although the first principles account of magneto-surface anisotropy by Szunyogh et al. [17] could address most of the problems we shall investigate in this paper, for the point we wish to make, the above model will suffice.

To illustrate the dilemma, let us assume that all the spins are parallel and make an angle θ with the surface normal. Furthermore, let us denote the energy of such a configuration for N (100) layers of an FCC lattice by $E_N(\theta)$ and seek the ground state of the classical Hamiltonian (1) – neglecting for the moment the last term – by minimizing $E_N(\theta)$ with respect to θ. For $N = 1$

$$E_1(\theta) = -2J - \tfrac{1}{4}A\omega + (\tfrac{3}{4}A\omega - \lambda)\cos^2(\theta), \quad (2)$$

where A is the so-called dipolar Madelung constant [17]. Evidently, for $\lambda > \frac{3}{4}A\omega$ the ground state corresponds to $\theta = 0$, while for $\lambda < \frac{3}{4}A\omega$ to $\theta = \pi/2$. For $N > 1$, $E_N(\theta)$ can also be readily determined, and – as it will be shown in Section 3 – yields a ground-state orientation $\theta = 0$ for $N < N_c$ and $\theta = \pi/2$ for $N < N_c$, with no intermediate tilting angle θ in between. Thus the question arises: "what does give rise to a tilt?"

As discussed by Landau and Lifshitz [16], for bulk magnets with two symmetry-determined easy axis one can predict a magnetization which points at an intermediate direction between them if one includes a contribution to the spin Hamiltonian analogous to the fourth-order term in Eq. (1). Indeed, it can be readily seen that for $N = 1$ and $\gamma \neq 0$ the minimum in the energy occurs at a tilted angle $\theta = \arccos(\sqrt{(\lambda - \frac{3}{4}A\omega)/2\gamma})$. Thus, it is natural that Chappert and Bruno [4] and Jensen and Bennemann [12] explained the observed tilt for the magnetic thin films by invoking such higher-order contribution to the anisotropy energy. The principle aim of the present paper is to point out that this is not the only mechanism for the tilted magnetization in the ground state. In fact, as we shall show presently, the above solution of the energy minimization problem to find the ground state is incorrect and misleading. If we allow the spin orientation to vary from layer to layer there is another stationary state whose energy is lower than that of the uniform configuration discussed above and corresponds to an average orientation tilted with respect to the surface normal even when $\gamma = 0$. The possibility of such non-collinear ground-state spin configuration in magnetic thin films was first noted by Mills [18,19]. Here we investigate in more detail the circumstances when it can arise. To illustrate the mechanism at work, in Section 2 we study the case of a bilayer. In Section 3 we investigate the orientational transition from perpendicular to in-plane magnetization with increasing N, while in Section 4 the effect of the fourth-order anisotropy term is studied. Quantitative fits to experimental data for the Co/Au(1 1 1) system are performed in the last section stressing the importance of non-collinearity even in the presence of fourth-order anisotropy ($\gamma \neq 0$).

2. The case of a bilayer

Confining ourselves to spin states in which the spins are parallel in a given layer but their orientations may differ from layer to layer, the energy of N layers per 2D unit cell implied by Eq. (1) can be written as

$$E_N(\theta_1, \theta_2, \dots, \theta_N) = -\tfrac{1}{2}\sum_{p,q=1}^{N}(Jn_{pq} + \tfrac{1}{2}A_{pq}\omega)\sin(\theta_p)\sin(\theta_q) - \tfrac{1}{2}\sum_{p,q=1}^{N}(Jn_{pq} - A_{pq}\omega)\cos(\theta_p)\cos(\theta_q) - \sum_{p=1}^{N}\lambda_p\cos^2(\theta_p) + \sum_{p=1}^{N}\gamma_p\cos^4(\theta_p), \tag{3}$$

where n_{pq} is the number of nearest neighbors of a site in layer p within layer q and A_{pq} is the dipole coupling constant determined by the lattice geometry. In order to find the extremal values of the energy, Eq. (3), as a function of the set of angles $(\theta_1, \theta_2, \dots, \theta_N)$ we seek the solutions of the Euler–Lagrange equations:

$$\frac{\partial E_N(\theta_1, \theta_2, \dots, \theta_N)}{\partial \theta_p} = -\sum_{q=1}^{N}(Jn_{pq} + \tfrac{1}{2}A_{pq}\omega)\cos(\theta_p)\sin(\theta_q) + \sum_{q=1}^{N}(Jn_{pq} - A_{pq}\omega)\sin(\theta_p)\cos(\theta_q) + 2\lambda_p\sin(\theta_p)\cos(\theta_p) - 4\gamma_p\sin(\theta_p)\cos^3(\theta_p) = 0. \tag{4}$$

The aim of this and the forthcoming section is to elaborate on the observation that allowing different spin orientations in different layers naturally yield a solution to the above equation which corresponds to an average magnetization tilted with respect to the surface normal even without requiring the presence of the fourth-order anisotropy term in Eq. (3) [15,18,19]. Consequently, in the present (and also in the next) section γ_p is chosen to be zero. Noting that many of our general conclusions are also valid in the multilayer cases, we now confine ourselves to the case of a bilayer. In particular, we will be looking for the set of anisotropy constants (λ_1, λ_2) at given J and ω for which tilted average magnetization can occur.

To begin with, we observe that the uniform perpendicular and in-plane magnetizations, $\theta_1 = \theta_2 = 0$ and $\theta_1 = \theta_2 = \pi/2$, respectively, satisfy Eq. (4). Interestingly, along the line in the parameter space (λ_1, λ_2),

$$\lambda_1 + \lambda_2 = \tfrac{3}{2}(A_{11} + A_{12})\omega, \tag{5}$$

the energies of the two solutions are degenerate. Below this line the easy axis lies in the plane, whilst above it is perpendicular to it. Clearly, if the magnetization is to change continuously across this line, solutions corresponding to tilted magnetizations should exist in its vicinity. An efficient way to investigate these is to choose a pair of angles θ_1^*, θ_2^* and determine the parameters λ_1, λ_2 by demanding that θ_1^* and θ_2^* satisfy Eq. (4). Substituting these parameters into Eq. (3), we can easily express the energy of the tilted solutions as a function of the orientations in the two layers:

$$E_2(\theta_1^*, \theta_2^*) = -(Jn_{11} + \tfrac{1}{2}A_{11}\omega) - \tfrac{1}{2}(Jn_{12} + \tfrac{1}{2}A_{12}\omega)\left(\frac{\sin(\theta_1^*)}{\sin(\theta_2^*)} + \frac{\sin(\theta_2^*)}{\sin(\theta_1^*)}\right). \qquad (6)$$

Note that the position of the minimum, θ_1^* and θ_2^*, as well as the minimum energy $E_2(\theta_1^*, \theta_2^*)$ are functions of the parameters J, ω, λ_1 and λ_2. Evidently, for $\theta_1^* \neq \theta_2^*$ the average magnetization is tilted with respect to the surface normal. It is reassuring to note that for no interaction between the two layers, i.e., for $n_{12} = 0$ and $A_{12} = 0$, and on substituting $n_{11} = 4$ for FCC (1 0 0) layers as well as abbreviating A_{11} by A, Eq. (6) gives the double of the energy in Eq. (2) for $\theta = \pi/2$.

By using Eq. (6) we now can compare the energies of the different types of solutions, i.e., those of the collinear ($\theta_1 = \theta_2 = 0$ or $\pi/2$) and the non-collinear ones. After straightforward algebra we find

$$E_2(\theta_1 = \theta_2 = 0) - E_2(\theta_1^*, \theta_2^*) = \tfrac{1}{2}(n_{12}J - A_{12}\omega)\left(\frac{\cos(\theta_1^*)}{\cos(\theta_2^*)} + \frac{\cos(\theta_2^*)}{\cos(\theta_1^*)} - 2\right) \geqslant 0, \qquad (7)$$

$$E_2\left(\theta_1 = \theta_2 = \tfrac{\pi}{2}\right) - E_2(\theta_1^*, \theta_2^*) = \tfrac{1}{2}(n_{12}J + \tfrac{1}{2}A_{12}\omega)\left(\frac{\sin(\theta_1^*)}{\sin(\theta_2^*)} + \frac{\sin(\theta_2^*)}{\sin(\theta_1^*)} - 2\right) \geqslant 0. \qquad (8)$$

Remarkably, whenever they exist, the energy of non-collinear states is always below or equal to the energies corresponding to the perpendicular or in-plane magnetizations.

Of course, there are regions in the parameter space where $\theta_1 = \theta_2 = 0$ or $\pi/2$ are the only solutions. It is particularly interesting to investigate the boundaries of such regions in the λ_1 versus λ_2 plane for fixed values of the parameters J and ω. It is straightforward to show that the lower and upper bound of the tilt area are given by

$$(2\lambda_1 - Jn_{12} - (\tfrac{3}{2}A_{11} + \tfrac{1}{2}A_{12})\omega)(2\lambda_2 - Jn_{12} - (\tfrac{3}{2}A_{11} + \tfrac{1}{2}A_{12})\omega) = (Jn_{12} - A_{12}\omega)^2, \qquad (9)$$

and

$$(2\lambda_1 + Jn_{12} - (\tfrac{3}{2}A_{11} + A_{12})\omega)(2\lambda_2 + Jn_{12} - (\tfrac{3}{2}A_{11} + A_{12})\omega) = (Jn_{12} + \tfrac{1}{2}A_{12}\omega)^2, \qquad (10)$$

respectively. The derivation of the above equations with a complete study of the magnetic ground state of the model will be published elsewhere. Here we restrict our considerations to the evolution of the tilt in the λ_1 versus λ_2 parameter space. The ground-state phase diagram is shown in Fig. 2 for $\omega = 0.1$ as measured in units of J. We note that the area of the tilt magnetization is getting larger as ω is increasing. Moreover, as the dipolar coupling goes to zero ($\omega \to 0$) the upper and lower bound of the tilt zone tends to the line defined in Eq. (5) and we get back the Nèel model with ferromagnetic ground state. Since the exchange coupling J is generally much larger than the strength of the dipole–dipole interaction ω as well as the magnitudes of the anisotropy term λ_p, the orientations of the spins in the different layers tend to be almost parallel, i.e., the difference between the angles of the tilted solutions will be small, typically less than 0.1 rad as was discovered by Mills et al. [18,19]. It is also worthy to mention that collinear solution other than those perpendicular or parallel to the layer can exist only for $\lambda_1 = \lambda_2 = \tfrac{3}{4}(A_{11} + A_{12})\omega$ where, however, the orientations of the magnetization are undefined (critical point).

3. Multilayers

Although the mathematics is somewhat more complicated, the above discussion can be repeated

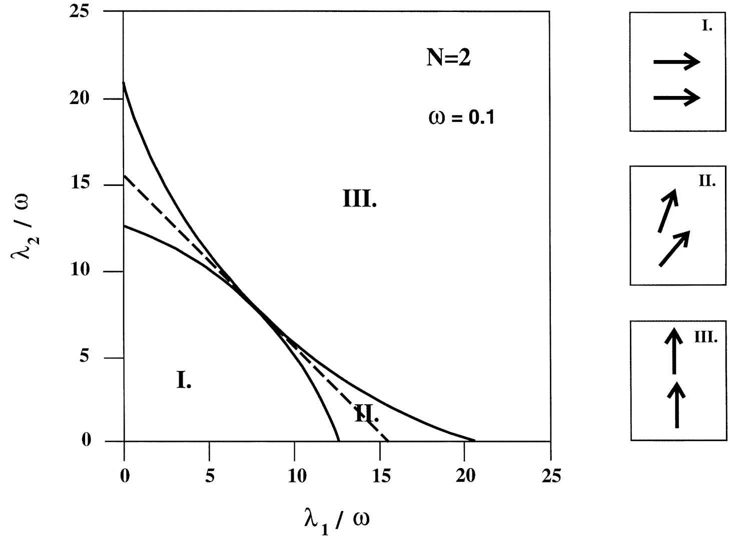

Fig. 2. The ground-state phase diagram of a bilayer on the λ_1/ω versus λ_2/ω plane for $\omega = 0.1$ (in units of J) and $\gamma_1 = \gamma_2 = 0$. Schematic pictures of the spin directions in the three characteristic regions of the phase diagram are shown on the right-hand side. The dashed line is defined by Eq. (5).

for N (> 2) layers starting from Eq. (4). In particular, it can be shown that if a non-collinear solution $(\theta_1^*, \theta_2^*, \dots, \theta_N^*)$ exists for the multilayer system, its energy is always smaller than those corresponding to the ferromagnetic states being magnetized perpendicular or parallel to the surface. In order to dramatize the effect we are considering, the anisotropy constants of the inner layers $(\lambda_2, \lambda_3, \dots, \lambda_{N-1})$ have been chosen to be zero and as in Section 2 the ground-state phase diagrams are presented in the λ_1 versus λ_N parameter space. This choice is consistent with the results of first-principles calculations [17,20] which generally predicted large magnetic anisotropy at the surface of ferromagnetic films, or rather at the interface of the film and the substrate, while yielding small values in between. Note that in order to keep correspondence with the previous section we continue to use the notations λ_1 and λ_2 for the two non-vanishing anisotropy constants.

Since the region where tilted solutions exist is expected to separate those of the uniform in-plane and perpendicular magnetizations it has to be close to the line along which the energy of these two solutions are equal,

$$\lambda_1 + \lambda_2 = \tfrac{3}{4} \sum_{p,q=1}^{N} A_{pq}\omega, \tag{11}$$

where the right-hand side is an obvious generalization of that of Eq. (5). Since the dipolar Madelung constants A_{pq}, which depend only on the actual layer geometry, rapidly decrease with an increasing value of $|p - q|$ [17], the right-hand side of Eq. (11) increases monotonously with N. Therefore, for a given set of parameters λ_1, λ_2 and ω Eq. (11) clearly determines the value of N_c where – as mentioned in Section 1 – within the subset of collinear magnetic states a first-order transition from perpendicular to in-plane magnetization occurs. As in the case of Eqs. (9) and (10) the phase boundaries in the λ_1 versus λ_2 plane can be determined yielding a phase diagram for different multilayers as shown in Fig. 3 for $\omega = 0.01$. Evidently, the larger the number of layers (N) the larger the area of the tilt zone. As far as the N dependence of the orientation of the magnetization for a given set of (λ_1, λ_2) is concerned, there are two possibilities: the point (λ_1, λ_2) is outside or inside one of the tilt zone. In

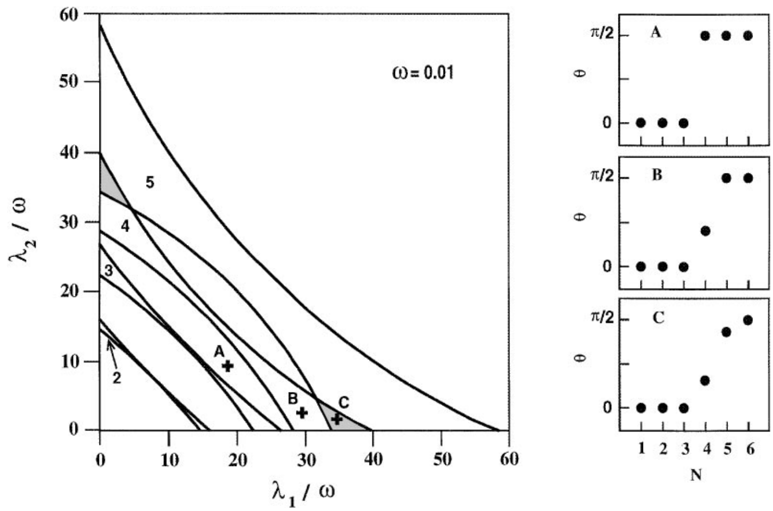

Fig. 3. The ground-state phase diagram of multilayers with $N = 2$–5 on the λ_1/ω versus λ_2/ω plane for $\omega = 0.01$ (in units of J) and $\gamma_p = 0$ ($p = 1, \dots, N$). The average angle versus N phase diagrams are depicted for the points A, B and C on the right-hand side (see in the text).

the first case (point A in Fig. 3) there is a discontinuous reorientation transition as the number of layers exceeds a critical value N_c. In the second case (point B in Fig. 3) at the critical thickness, the point (λ_1, λ_2) is lying in the tilt zone of which, the layer-averaged magnetization will have an intermediate angle and mimic a continuous transition. Of course, since the physical values of N form a discrete set the word 'continuous' in the present context needs to be interpreted with appropriate care. Experimentally, during the growth process partially filled layers can be achieved representing a non-integer value for N, however, a theoretical description of such cases is beyond the scope of the present work. Interestingly, the tilt regions corresponding to different values of N can overlap (see point C in Fig. 3). This implies that for a given set of parameters from such a region there exist (at least) two different thicknesses of the multilayer with tilted ground-state magnetization.

4. The effect of the fourth-order term

Above, we have argued that the fourth-order term in Eq. (1) is, in principle, not necessary to explain the presence of tilted magnetizations detected in several experiments [4–6]. In what follows we reintroduce the fourth-order term into the theory and study its effect on the ground-state phase diagram. This will allow us to make contact with the conventional arguments of Refs. [4,12].

An estimate, based on very general symmetry arguments, of the fourth-order anisotropy leads to the conclusion that it is comparable on all layers and also not much different from its bulk value [4,12]. Therefore, to simplify the foregoing discussion we let $\gamma_p = \gamma$ for all layers p. At the same time, as before, we take λ_p's to be non-zero only on the top and the bottom layers. As it turns out, the extra term $\sum_{p=1}^{N} \gamma \cos^4(\theta_p)$ in $E_N(\theta_1, \theta_2, \dots, \theta_N)$ does not complicate matters too much. For instance, the line along which the energies of the uniform perpendicular and parallel magnetizations are equal, i.e. the generalization of Eq. (11), is given by

$$\lambda_1 + \lambda_2 = \frac{3}{4} \sum_{p,q=1}^{N} A_{pq}\omega + N\gamma. \tag{12}$$

Whilst it is not surprising that the present generalized theory leads to a ground-state magnetization which is tilted towards the surface normal, it is unexpected that, again, the absolute minima refer to non-collinear solutions. To demonstrate this statement we generalize Eqs. (7) and (8) for a bilayer in the presence of fourth-order anisotropy terms to yield

$$E_2(\theta_1 = \theta_2 = 0) - E_2(\theta_1^*, \theta_2^*) = \tfrac{1}{2}(n_{12}J - A_{12}\omega)\left(\frac{\cos(\theta_1^*)}{\cos(\theta_2^*)} + \frac{\cos(\theta_2^*)}{\cos(\theta_1^*)} - 2\right) + \gamma\sin^2(\theta_1^*) + \gamma\sin^2(\theta_2^*) \geqslant 0, \quad (13)$$

$$E_2(\theta_1 = \theta_2 = \tfrac{\pi}{2}) - E_2(\theta_1^*, \theta_2^*) = \tfrac{1}{2}(n_{12}J + \tfrac{1}{2}A_{12}\omega)\left(\frac{\sin(\theta_1^*)}{\sin(\theta_2^*)} + \frac{\sin(\theta_2^*)}{\sin(\theta_1^*)} - 2\right) + \gamma\cos^2(\theta_1^*) + \gamma\cos^2(\theta_2^*) \geqslant 0. \quad (14)$$

Introducing the notation $\lambda_p' = \lambda_p + 2\gamma\cos^2(\theta_p)$ we find the same boundaries for λ_1' and λ_2' as for λ_1 and λ_2 in Eqs. (9) and (10). Since at the boundary between the areas of the tilted and in-plane magnetizations on the ground-state phase diagram θ_p tends to $\pi/2$, consequently, λ_p' tends to λ_p, the fourth-order anisotropy does not affect the shape of the lower boundary of the tilt region. However, at the upper boundary $\theta_p \to 0$ and, therefore, $\lambda_p' \to \lambda_p + 2\gamma$. As a result, this boundary is simply shifted by 2γ on the phase diagram as compared to the case of $\gamma = 0$. As mentioned in Section 1, in a monolayer the fourth-order anisotropy causes a tilting of the magnetization. In a multilayer this is not necessarily the case. However, it makes the tilt area broader relative to the case of $\gamma = 0$. This broadening of the tilt region is shown in Fig. 4 for a bilayer.

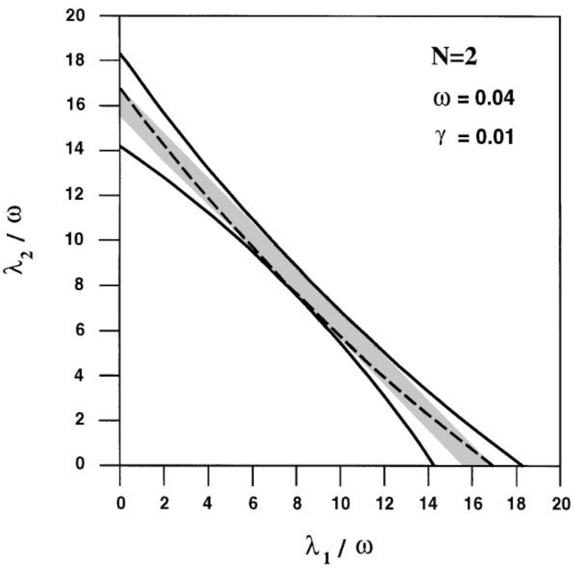

Fig. 4. The ground-state phase diagram of a bilayer on the λ_1/ω versus λ_2/ω plane for $\omega = 0.04$ and $\gamma = 0.01$ (both in units of J). As a comparison, the upper boundary corresponding to the case of $\gamma = 0$ is shown by a dashed line. The region where tilted collinear spin-states, although not necessarily ground states, exist is indicated by shading.

5. Application to the Co/Au(1 1 1) system and conclusions

Having discussed the possibility of spin tilting in general, the question arises whether measurements on real systems can be reproduced by the spin Hamiltonian (1) with a reasonable set of parameters. The most appealing system for such a study is the Co/Au(1 1 1) system where both the geometrical and the magnetic structure is simple, and the intermediate transition region, where spin tilting is found experimentally, is relatively broad.

In Fig. 5 the experimentally measured mean orientations of magnetization in the Co_N/Au(1 1 1) thin films [5,6] are recorded. Characteristically, up to a film thickness of 3 layers the magnetization points normal to the surface, while above 6 layers it lies parallel with it. In between there is a continuous

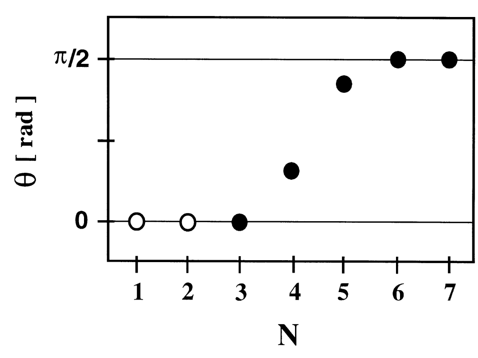

Fig. 5. Mean angles of the magnetization in the Co/Au(1 1 1) system as deduced from Refs. [5,6]. The open circles indicate that, most likely because of the lower Curie temperature, at 300 K no magnetization for $N < 2$ was found.

transition in the angle of magnetization. Since the present formalism accounts merely for perfect, two-dimensional translation invariant layers, from the set of points we used in our numerical fit we excluded the value for the non-integer coverage ($N = 4.5$) also given in Refs. [5,6].

Because of the small number of data points, we had to decrease considerably the number of parameters in the spin Hamiltonian. Some of such restrictions were introduced before, namely, $J_{pq} = J$, $\omega_{pq} = \omega$, $\lambda_p = 0$ for $p = 2, \dots, N$ and, in the presence of fourth-order anisotropy, $\gamma_p = \gamma$ for all p. Additionally, we choose λ_p at only one terminal layer to be non-zero. This approach is partially supplied by our experience of previous first-principles studies predicting generally much larger anisotropy at the interface between the magnetic film and the non-magnetic substrate than at the surface [17,20]. Therefore, in total there remained four independent parameters which define our model Hamiltonian (1): J, ω, λ and γ. If, however, one is interested only in the angles which characterize the ground states, but not in the ground-state energies, it is sufficient to deal with three ratios $J' = J/\omega$, $\lambda' = \lambda/\omega$ and $\gamma' = \gamma/\omega$. It should also be noted that we calculated the dipole–dipole Madelung constants A_{pq} for an FCC(1 1 1) geometry.

To highlight the role of various factors which determine the tilt angle, we used three different schemes to fit the experimental data: (A) without fourth-order anisotropy ($\gamma = 0$), (B) with fourth-order term and also, (C) based on the collinear tilted state for $\gamma \neq 0$. It is noteworthy that whilst within scheme C the parameters λ' and γ' unambiguously determine the magnetic orientation, a numerical fit of the proper, non-collinear ground states of the Hamiltonian (1) to a measurement of the magnetic directions also determines the Heisenberg exchange parameter J.

The parameters as obtained from the different kinds of numerical fits are listed in the upper part of Table 1. In all the three cases the mean angles presented in Fig. 5 could be recovered with an accuracy less than 0.02 rad. Apparently, the value of J' for case A is seven times smaller than that for case B. In view of the previous sections, this in fact, is not surprising, since for $\gamma = 0$ the only way to broaden the tilting regions corresponding to a given thickness of the film – thus to achieve overlap between different layer thicknesses (see Fig. 3) – is to increase the ratio ω/J or equivalently decrease J'. Since for B and C the competition of the second and fourth-order anisotropies broadens the tilting regions, the value of J' can be considerably increased in these cases. A less pronounced difference between schemes B and C is a decrease of γ' by about 20%. This trend is again clear, since by taking into account the proper non-collinear ground states the tilting region broadens relative to that of the collinear case (see Fig. 4).

Table 1
Parameters fitted to the measured mean directions of magnetization in the Co/Au(1 1 1) system. Three different models were used: non-collinear ground states without (A) and with (B) fourth-order anisotropy term, and collinear tilted state in the presence of fourth-order anisotropy term (C). Note that in case C there is no information on the exchange parameter J. The strength of the dipolar interaction was fixed to $\omega = 6.63$ μeV (see in the text)

	A	B	C
$J' = J/\omega$	179	1250	–
$\lambda' = \lambda/\omega$	37.3	43.0	44.3
$\gamma' = \gamma/\omega$	–	1.25	1.56
J (meV)	1.19	8.29	–
λ (meV)	0.247	0.285	0.294
γ (μeV)	–	8.29	10.34
K_S (mJ/m^2)	0.554	0.638	0.657
K_2 (kJ/m^3)	–	79.1	98.8

In order to provide parameters which can be compared to first-principles calculations, or even to other experiments, we made an estimate of the magnetic moment of Co to be 1.7 μ_B [21] and then, by using the 2D lattice constant of the (1 1 1) facet of FCC gold ($a = 5.43$ a.u.), we calculated $\omega \simeq 6.63$ μeV. The corresponding values for J, λ and γ are seen in the middle part of Table 1. The surface and second hexagonal anisotropy constants, K_S and K_2, respectively, as computed from the parameters λ and γ are presented in the lower part of Table 1. While the surface anisotropy constants compare reasonably well with that given in Refs. [5,6] (0.62 mJ/m^2), our values for K_2 are somewhat lower than the bulk value of

K_2 (143 kJ/m^3). A possible reason for that might be that in our model we considered an FCC(1 1 1) layer geometry with the lattice constant of bulk gold rather than the hexagonal Co bulk structure.

As pointed out above, an interesting feature of the non-collinearity of ground states is that they give information on the exchange parameter J. The fact, however, that the experiments were carried out at room temperature, implies that – at least for $N > 2$ [5,6] – the Curie temperature, T_C, for the thin film must be higher. From this point of view, J for case A seems to be rather low, while for case B it displays a fairly reasonable value. This may be taken as an indication that fourth-order anisotropy does play a role in the Co/Au(1 1 1) system. However, the connection between J and T_C for thin films is not firmly established and we would prefer to defer judgement on this matter pending further experiments. A particularly useful one of these would be a study of the reorientation as a function of λ_1–λ_2. Evidently, for the fourth-order (γ only) mechanism, a change in λ_1–λ_2 would have a little effect, while the non-collinear state would be dramatically altered. Perhaps the easiest experiments would be to compare the behavior of a non-magnetic substrate/magnetic film/open surface system with that of a magnetic film sandwiched between two equivalent non-magnetic covers. As Fig. 4 indicates, in the latter case, $\lambda_1 = \lambda_2$, the transition region would be dominated by γ. On the other hand, in the asymmetric system the non-collinear magnetism could produce a significantly broader transition.

In summary, we have presented a careful investigation of the ground states of a simple spin-Hamiltonian containing magnetostatic dipole–dipole interaction, as well as second- and fourth-order surface magneto-crystalline anisotropies. We have shown that continuous reorientation transition with respect to the thickness of an ultra-thin magnetic film is a direct consequence of the model even in the absence of fourth-order anisotropy. Interestingly, we find that the spins corresponding to the tilted average magnetization are necessarily non-collinear. Fits to the measured directions in the Co/Au(1 1 1) thin films show that at present there are not enough experimental data to distinguish between alternative explanations for the phenomenon. Finally, we proposed some new experiments which could do so.

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