Invited paper

Magnetization, coercive forces and magnetoresistance in simple and double Co films with perpendicular magnetization


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Our Au(111)/Co/Au samples show perpendicular spontaneous magnetization for Co thicknesses \( t \) below \( t^* = 1.2 \text{ nm} \). Recent RHEED results are in favour of the existence of an important "strain-induced" anisotropy, helping to overcome the shape anisotropy. From the measured parameters of anisotropy and roughness, we propose a quantitative model that explains the fast variation of the perpendicular coercive force with \( t \) below \( t_c \). Finally, disymmetric Au/Co\((t_1)/\text{Au}/\text{Co}(t_2)/\text{Au} \) bilayers \( (t_1 \text{ and } t_2 < t_c) \) show enhanced magnetoresistance (5%) in the field range where the layers are magnetized antiparallel.

1. Introduction

In recent years, the research on ferromagnetic ultrathin films and multilayers has undergone an astonishingly fast evolution from pure fundamental work to applications, starting for instance from the first announcement in 1986 of perpendicular magnetic anisotropy on films of a few atomic layers of Co on a gold substrate [1], to the first realization of a read/write process for perpendicular magneto-optical recording on Pt/Co multilayers [2] and the present realization of prototype giant storage density disks based on Pt/Co multilayers [3].

Indeed, at least three important effects have been experimentally evidenced on these compounds, that, together with the fast progress in deposition techniques, widely opened the field of applications:

(1) the possibility to realize films with perpendicular anisotropy [1] and to stack these films in multilayers of macroscopic magnetization while preserving this anisotropy [4].

(2) the possibility to easily follow the magnetization of these ultrathin films and multilayers using magneto-optical effects [5], with even enhanced Faraday and Kerr rotations compared with bulk Co for instance [6].

(3) the discovery of enhanced [7] and sometimes giant [8] magnetoresistances in these structures.

The two first effects lead directly to perpendicular high density magneto-optical storage, while the third should soon find application in magnetoresistive heads.

Much theoretical [9, 12, 22–24] and experimental [10, 11, 15, 21, 26] work has been devoted lately to the interpretation of these effects. On the other side, "technical" effects of utmost importance for applications, such as hysteresis and aftereffects, are beginning to be studied on such structures [15–17].

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In this paper, we will review our latest results in this field.

2. Samples

Our experiments have been performed on Au/Co/Au simple sandwiches and Au/Co/Au/Co/Au bilayers. Detailed reports of the samples preparation and structural characterization have been published elsewhere [18, 19], and we will present here only new RHEED results.

From previous experiments, we knew that the surface of the Au substrate layer consisted mainly of (111) compact terraces, separated by monoatomic steps about 20–30 nm apart. Recent RHEED experiments [20] confirmed the above description. As expected for a rather smooth surface, the RHEED pattern of the Au substrate is made up of lines. With the deposition of Co, the diagram evolves slowly; however, a line diagram remains up to about 3 nm: this is also in agreement with our previous observations that the surface roughness increases slowly with the cobalt thickness. Moreover, measurement of the distances between the lines shows that the cobalt lattice is expanded in the plane of the film: indeed in bulk gold (111) planes, the interatomic distance is about 13% greater than in bulk cobalt (0001) planes. As expected, this lattice expansion decreases with the cobalt thickness $t$, as displayed in fig. 1.

More puzzling is the fact that, from the deposition of the very first atomic layer of the gold overlayer, the lines recover the spacing of gold substrate. Furthermore, transmission electron microscopy on simple sandwiches with cobalt thickness around 2 nm never detected any lattice expansion of the cobalt film, to a precision of 1%. We thus must conclude that, at this point, we have no definite evidence that, in the sandwiches, the cobalt film is expanded as measured by RHEED on free cobalt surfaces, although some strain must finally remain. We have so far no indication on the lattice parameter perpendicular to the film.

3. Perpendicular magnetic anisotropy

Perpendicular spontaneous magnetization in thin films requires the existence of a large uniaxial anisotropy to overcompensate the dipolar shape anisotropy. From ferromagnetic resonance experiments on simple Au/Co/Au sandwiches of cobalt thickness $t$ between 1.1 and 8 nm [1], we could measure the anisotropy constants $K_1$ and $K_2$ of the cobalt films. For the first constant $K_1$ we found the dependence: $K_1 = K_{\text{in}} + 2K_s/t$, with $K_s = 0.5$ erg cm$^{-2}$ at room temperature and $K_s = 0.7$ erg cm$^{-2}$ at $T = 10$ K. The observed behaviour, crossing from an in-plane anisotropy above $t^*_s = 1.2$ nm to perpendicular one below, comes from this rapid increase of $K_1$ at low thicknesses.

This $1/t$ dependence is a signature of a “surface” effect and, since its first observation [14], $K_s$ was identified as the “surface anisotropy”, due to the breaking of the crystalline symmetry at the interface, as predicted by Néel.
Several attempts have been made to calculate $K_s$, either from the phenomenological Néel's model [12], or from more sophisticated band calculations on unsupported monolayers [22–24]. We proposed another mechanism [15, 26] that could coexist with Néel's "surface" anisotropy. As in Néel's model, it refers to magnetostriction constants, but this time through the strains that must exist in any real film due to the mismatch $\eta = (a_f - a_s)/a_s$ between film ($a_f$) and substrate ($a_s$) bulk lattice parameters ($\eta = -13.2\%$ for a Co/Au interface). When the film thickness increases, this mismatch is accommodated through strains, whose energy is proportional to the volume, and interfacial dislocations, whose energy is proportional to the surface. The competition of these two energies leads to the existence of a critical thickness $t_c$: below $t_c$ exists exact pseudomorphism, dislocations beginning to appear only above $t_c$ [27]. A straightforward calculation [15] to first order in $\eta$, neglecting the interaction between dislocations, predicts for the in-plane strain $\varepsilon$ a decrease above $t_c$ as

$$\varepsilon = -\frac{\eta t_c}{t}. \quad (1)$$

With the above dependence of $\varepsilon$, the magneto-elastic anisotropy energy becomes above $t_c$ equivalent to a "surface" energy [26], thus with an effective "strain induced" surface anisotropy constant $K_s$. Moreover, as $t_c$ depends on $\eta$ as $1/|\eta|$, the $1/t$ variation of $\varepsilon$ above $t_c$ will depend only on the parameters of cobalt: whatever the substrate, measurements on samples with thicknesses above $t_c$ should give the same value of $K_s$.

Introducing in this model parameters of bulk cobalt gives a value of $t_c$ about 3 Å [26]. In fig. 1 we have displayed the behaviour calculated from eq. (1), together with our RHEED measurements of in-plane strain on cobalt films with free surface: the agreement is excellent.

As we have no indication on the out-of-plane strain in our films, we have several ways to evaluate $K_s$ from magnetostriction constants. When allowing the out-of-plane strain to relax to minimize the elastic energy of the film, one obtains $K_s = 1.8\text{ erg cm}^{-2}$, a value of the same sign, although higher than experimental value. $K_s$ falls to $0.7\text{ erg cm}^{-2}$ when neglecting out-of-plane strain.

Moreover, our measurements of $K_s$ on Au/Cu/Co/Cu/Au samples (with $t > 1.5$ nm, thus above $t_c$ for this interface [28]) gave the same value of $K_s$ as for Au/Co/Au samples.

It thus seems clear from the above discussion that, together with the more classical magnetocrystalline surface anisotropy, strain induced anisotropy might play an important role in these thin films, though our results so far do not allow determination of their respective weights.

4. Hysteresis properties at low temperature

Hysteresis cycles recorded at low temperature ($T = 10\text{ K}$) on simple sandwiches Au/Co/Au [15, 26] confirm exactly the results of the ferromagnetic resonance experiments. Particularly, samples with $t$ below the critical thickness $t^* = 1.2$ nm display perpendicular magnetic anisotropy, with rather square hysteresis loops in fields applied perpendicularly to the film. A more puzzling effect is the thickness dependence of the "perpendicular" coercive force $H_c$. As is displayed in fig. 2, $H_c$ increases rapidly, roughly as $1/t^2$, when $t$ decreases below $t^*$. In a bulk sample, with uniaxial anisotropy constant $K$ and exchange stiffness $A$, the energy of a Bloch wall of surface $S$ would be given by $E_w = 4S\sqrt{AK}$. In a thin film such as ours, the energy of a domain wall would thus depend on thickness through both thickness fluctuations and anisotropy. This led us to make the hypothesis that coercive forces in our samples are due to the motion of domain walls, in a spatially fluctuating tension $\gamma$ (wall energy per unit area). This hypothesis was recently confirmed by domain imaging experiments on the same samples [34]. However these experiments also showed that small variations in the quality of the samples could lead
to a totally different hysteretic behaviour, driven mainly by domain nucleation.

To simplify the problem, we will consider a film of constant thickness \( t \) (\( t \) is the average thickness of real films) and lateral dimensions \( L \times L \) (cf. fig. 2). As \( t \) is below \( t^* \), the film has a perpendicular easy magnetization axis. We suppose this film is divided in two domains of magnetization \(+ M\), and \(- M\), separated by a Bloch wall. To minimize its energy, the wall will deform itself, and move under the effect of a perpendicular applied field \( H \). We choose the coordinate axis so that the \( z \) axis is perpendicular to the film; the \( y \) axis corresponds to the average direction of the wall, which then moves along the \( x \) axis (cf. fig. 2). Deformation of the wall along the \( z \) axis is unlikely, as it would create “magnetic charges” and thus cause a considerable energy increase: we then have to treat a two dimensional problem.

To take into account the fluctuating local thickness \( h(x, y) \) of real films, we introduce an effective wall tension:

\[
\gamma(x, y) = \left( \frac{h(x, y)}{t} \right) 4 \sqrt{2AK_s/h(x, y)} = \sqrt{h(x, y)/t} \gamma_0, \tag{2}
\]

where \( \gamma_0 \) is the wall tension without thickness fluctuations and \( 2K_s/h(x, y) \) is the local anisotropy constant.

The mean quadratic value of the fluctuations of \( \gamma \) can then be approximated as

\[
\delta \gamma = \sigma \gamma_0 / 2t, \tag{3}
\]

where \( \sigma \) represents the local thickness fluctuations amplitude.

Let \( \xi \) be the correlation length of \( \gamma \); it can be visualized as a kind of average extension of zones of maximum or minimum thickness in the film.

The problem of the motion of a deformable wall is in general impossible to solve exactly. We will use the approximate calculation method presented by Néel [29] for the 3D case. If \( \delta \gamma \ll \gamma_0 \), one can linearize the problem of the calculation of the wall energy. Minimization of \( E^c_w \) is then easy in the Fourier space, and the problem reduces to that of a plane rigid wall moving with a fluctuating energy:

\[
E^c_w = tL \gamma_0 \left( 1 + \left( \frac{\delta \gamma}{\gamma_0} \right)^2 f \right). \tag{4}
\]

The first term is the energy of the undeformed wall, the second (negative) is the gain in energy with deformation. \( f \) is a dimensionless function, fluctuating with the average position \( x_0 \) of the wall, and whose calculation requires the knowledge of all Fourier coefficients of \( \gamma \). However, a few simple remarks can help to obtain a sufficient knowledge.

1. The real wall deforms itself to jump over the obstacles (the maxima of \( h \)) one after the other, to minimize barrier heights. Each obstacle to pass corresponds to a maximum of \( f \). Thus the amplitude of the fluctuations of \( f \) must not depend on \( L \). As \( f \) must also be a function of dimensionless parameters, we propose that the average fluctuation amplitude of \( f \) be proportional to \( \xi/L \). Note that this will finally correspond to the barrier height for a rigid wall passing one obstacle: \( t \xi \delta \gamma \), with a reduction factor \( \delta \gamma/\gamma_0 \) linked to the deformation of the wall.

2. When moving on a distance \( \xi \), the effective rigid wall will encounter about \( L/\xi \) obstacles;
measurements at room temperature on the same samples seem in good agreement with the above model [16, 33].

5. Magnetoresistance

The mechanism most generally proposed to explain the large magnetoresistance observed in bilayers and multilayers is a spin dependent transmission of the electrons at the interfaces between magnetic and non-magnetic metals [9]. This phenomenon leads to a strong reduction of the carrier transmission and thus to a large increase of the resistivity when the neighbouring ferromagnetic layers have antiparallel magnetizations. This configuration happens naturally in multilayers with antiferromagnetic interactions.

Fig. 3. Coercive forces $H_c$ measured on a series of Au/Co/Au simple sandwiches, versus the inverse of the square of the Co thickness $t$. The dashed line is just a guide for the eye. The solid line corresponds to the behaviour predicted by eq. (5), with $\sigma = 0.3$ and 20 nm.

Thus the average distance between maxima of $E_w$ is about $\xi^2/L$.

To evaluate an average propagation field $H_p$, we follow Néel [31] and replace the real variation of $E_w$ by a succession of parabolic arcs, and obtain

$$H_p = \frac{2 \delta \gamma^2}{\gamma_0 \xi M_s} = \frac{2 \sigma^2 \gamma 2\pi At^*}{t^{5/2} \xi}, \quad (5)$$

where we have introduced $K = 2\pi M_s^2 t^*/t$ to get rid of $M_s$.

To compare expression (5) to experiment, we made the hypothesis that in our very thin films the main roughness comes from the substrate, and thus chose $\xi = 20 \text{ nm}$ and $\sigma = 0.3 \text{ nm}$ (cf. Section 2). $A$ has been taken equal to the bulk value: $2.1 \times 10^{-6} \text{ erg cm}^{-1}$. In fig. 3 is displayed the theoretical behaviour, together with the experimental values of $H_c$: the overall agreement is excellent, considering both the roughness of the model and the fact that we used no adjustable parameter. Moreover, recent aftereffects mea-

Fig. 4. Compared variation at $T = 4.2 \text{ K}$, versus perpendicular applied field $H$, of the relative magnetoresistance $(R - R_0)/R_0$, and relative magnetization $M/M_s$ of a bilayer Au(25 nm)/Co(0.65 nm)/Au(5 nm)/Co(0.35 nm)/Au(25 nm).
It may happen artificially in bilayers with different coercive forces [10, 11]. Fig. 4 displays the magnetoresistance (a) and the hysteresis loop (b) measured in perpendicular applied field at $T = 4.2$ K on a bilayer: Au(25 nm)/Co(0.65 nm)/Au(5 nm)/Co(0.35 nm)/Au(25 nm). The hysteresis loop is clearly that of two independent ferromagnetic layers with perpendicular anisotropy, having as expected very different coercive forces (cf. Section 4) and saturation magnetizations in the ratio of thicknesses. Thus there exists a range of fields where the layers are both monodomains, with opposite magnetizations. Indeed, the magnetoresistance exhibits a large (about 5%) plateau in this exact range, in agreement with the predictions of the above discussed mechanism.

In fig. 5 we show the magnetoresistance in perpendicular field of the same bilayer, but recorded at room temperature. At this temperature, coercive forces are much lower, and become determined mostly by dynamical phenomena [16]. Particularly at $t = 0.35$ nm, the relaxation time of the magnetization is negligible (about 20 ms, cf. ref. [6]) compared with the variation rate of the field in the recording of fig. 5. That explains why the magnetoresistance exhibits a plateau between nearly zero and the coercive force of the thick layer. The reduction in magnitude of the effect (1.5% instead of 5% at 4.2 K) is linked to the reduction of electron mean free path, and is quantitatively explained in our most recent calculations [32].

6. Conclusion

Clearly more experiments are needed to better understand the “surface induced” anisotropy effect. One way would be to evaluate the exact strain in the film and compare with magnetic properties, for instance through “in situ” magnetic and structural measurements on samples with free surface. Another possibility would be to measure $K_c$ on samples of very low thicknesses, on a couple $M/M'$ with a high value of $t_c$ [28].

At the same time, new possibilities in domain imaging should soon allow us to study more directly hysteretic behaviours [34].

Finally, we think that enhanced magnetoresistance caused in multilayers by different coercive forces might prove very useful in applications: as shown in fig. 5, after previous saturation only a weak field is needed to produce large resistivity changes, moreover nearly reversible.

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References

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