

Enhanced magnetoresistance of ultrathin (Au/Co)_n multilayers with perpendicular anisotropy

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We present magnetoresistance (MR) measurements performed on two Au/Co/Au sandwiches with ultrathin cobalt layers (0.32 and 0.76 nm) and on a bilayer Au/Co/Au/Co/Au (0.75 nm Co). The easy magnetization axis is shown to be perpendicular to the films, in agreement with previous magnetization and ferromagnetic resonance measurements. The hysteretic behavior is easily observed by measuring the MR, leading to a determination of the coercive fields of a few 10^2 G. The Co bilayer exhibits a drastic enhancement of the MR effect with respect to the corresponding monolayer ($\delta R/R=1.3\%$ at 300 K and 3% at 4.2 K) and a square hysteresis loop. Different mechanisms for the enhancement of the MR are proposed.

The magnetic properties and especially the surface and interface anisotropy of ultrathin magnetic films are of considerable interest, both from a fundamental point of view and for their potential applications. Perpendicular anisotropy in such systems is particularly attractive for future applications in high-density magnetic recording. Since the pioneering work of Gradmann,¹ much effort has been devoted to understanding magnetism in thin metallic films. Many types of magnetic systems consisting of sandwiches, multilayers, and superlattices have been experimentally and theoretically investigated, as described in several review papers.²⁻⁵ However to date little experimental data are available concerning the interface anisotropy of ferromagnetic films.

We will concentrate here on ultrathin films of cobalt sandwiched between gold layers. Recent experimental results [superconducting quantum interference device (SQUID) measurements of the parallel and perpendicular remanent magnetizations, and ferromagnetic resonance experiments] have shown that the magnetic interface anisotropy leads to a perpendicular magnetization of the ferromagnetic film for very thin Co deposits (one or two monolayers). When increasing the Co thickness, the magnetization becomes in plane, due to the competition between interface and shape anisotropy.⁵⁻⁷ In this Rapid Communication we report the first observation of magnetoresistance (MR) effects in the same Au/Co systems, both at low temperature and at room temperature, which confirms the preceding studies.

Despite the fact that the measurement of anisotropic MR in ferromagnetic metals is a very old technique, first discovered by Lord Kelvin in 1857,⁸ and widely used for films of a few tens of nanometers,⁹ there are, to our knowledge, no examples of such measurements on ultrathin ferromagnetic films. We intend to show that the study of the MR yields the most important characteristics

of magnetic films down to the monolayer, i.e., coercive and saturation fields, and magnetic anisotropy.

The samples are Au/Co/Au (111) and Au/Co/Au/Co/Au (111) grown by slow evaporation in ultrahigh vacuum (10^{-10} Torr) onto a glass substrate with very small surface roughness (0.5 nm). The film thickness is monitored by a quartz oscillator during deposition. The first Au layer (thickness 25 nm) consists of large polycrystals (typical lateral dimensions 200–300 nm) with random orientations in the plane of the film but with a (111) oriented surface. The surface exhibits terraces with monoatomic steps. The characterization of these sandwiches is performed by *in situ* resistivity, followed by post-deposition grazing x-ray diffraction and by transmission electronic microscopy (TEM). The TEM results obtained on a 8-nm-thick Co film show that the Co film on Au (111) is hcp with the *c* axis perpendicular to the film.¹⁰ The sandwiches have sharp interfaces and good crystallinity. The upper Au layer (25 nm thick) protects the sample against oxidation. The three samples discussed in this article have the following characteristics: sample 1, Au (25 nm)/Co (0.32 nm)/Au (25 nm); sample 2, Au (25 nm)/Co (0.76 nm)/Au (25 nm); sample 3, Au (25 nm)/Co (0.75 nm)/Au (3 nm)/Co (0.75 nm)/Au (25 nm).

In order to measure their MR, the samples were cut to the following dimensions: 1×10 mm². Electrical contacts were made by sticking gold leads with silver paint on the top gold covering. Four-point measurements were performed with an alternating bridge operating at low frequency (32 Hz), with very weak power dissipation (10^{-8} – 10^{-9} W) and high precision ($\Delta R/R=10^{-5}$ – 10^{-6}). The MR was measured both at room temperature in the field range 0–0.8 T and at low temperature (1.3–4.2 K) for magnetic fields in the range 0–5 T. Several field orientations with respect to the film plane

and the electric current could be obtained. In this Rapid Communication we will use the following notations: H_{\perp} for a field perpendicular to the film plane, and H_t for a transversal field, i.e., in the film plane but perpendicular to the electric current.

For all three samples, the room-temperature value of the resistance is consistent (to within 10%) with that of a pure gold sample having the same geometry before any thermal cycling. The resistance increases after each thermal cycle down to helium temperature due to the appearance of strain-induced defects. Nevertheless, the phonon-linked part of the resistivity is the same as for pure gold (to within 1%) in all the sandwiches and does not depend on the history of the samples.

Despite the fact that the resistance is essentially due to the two thick gold layers in the sandwiches, the magnetic behavior of the cobalt layer hysteresis is clearly seen in the MR, the maximum value of $\Delta R(H)/R(0)$ being of the order of 10^{-2} . Therefore, the Co magnetism causes strong scattering of the electrons. A tentative explanation of this phenomenon will be given later. The value of the resistance is linked to the Co film magnetization. $R(0)$ reflects the remanent magnetization of the film; R is maximum (R_c) near the coercive field H_c when the domain structure of the magnetic film is the most complex. For fields $H > 3T$, a parabolic contribution to $R(H)$ is due to the MR of gold. We note $R_{Au}(H)$ this parabolic magnetoresistance which can be extrapolated to $H=0$, $\delta R(H) = R(H) - R_{Au}(H)$, and H_c the field giving the maximum value of δR noted δR_c . The parameter $r = [\delta R_c - \delta R(0)]/\delta R_c$ can be used to characterize the form of the Co layer hysteresis loop. The value of r increases from 0 to 1 when the hysteresis loop changes from flat (no hysteresis) to perfectly squared.

In the three samples the comparison of $R(H)$ for H_{\perp} and H_t confirms the existence of an easy magnetization axis perpendicular to the film plane. In the transverse configuration $\delta R(0) \sim \delta R_c$, whereas in the perpendicular direction, the hysteresis loop is broad, and the maximum relative MR $\delta R_c/R$ is about 1%.

For sample 1 (0.32 nm Co), $T_c = 18$ K (Ref. 6) and is not far out from the temperature range of the experiments. A clear temperature effect is observed, both on the perpendicular H_c value (0.3 T at 4.2 K and 0.54 T at 1.3 K) and on r , which is 0.25 at 4.2 K and 0.47 at 1.3 K. The perpendicular MR $\delta R_c/R$ is 2% at 1.3 K (Fig. 1). The perpendicular saturation field is 2.2 T at 4.2 K and 3 T at 1.3 K.

For sample 2 (0.76 nm Co), the hysteresis loop for H_{\perp} is closer to a square than for sample 1: $r = 0.78$ (Fig. 2). The coercive field is equal to 0.22 T at low temperature. The Curie temperature is greater than 300 K in this case, and so no temperature effect can be observed in the helium range.

The most interesting behavior is that of the Co bilayer (sample 3) in which drastic effects have been recorded both at 4.2 and 300 K [Figs. 3 and 4(b)]. In that sample the perpendicular MR exhibits very abrupt variations in the vicinity of the coercive field. The zero field and saturation field resistance are exactly the same, which is characteristic of a perfectly square hysteresis loop for H_{\perp}

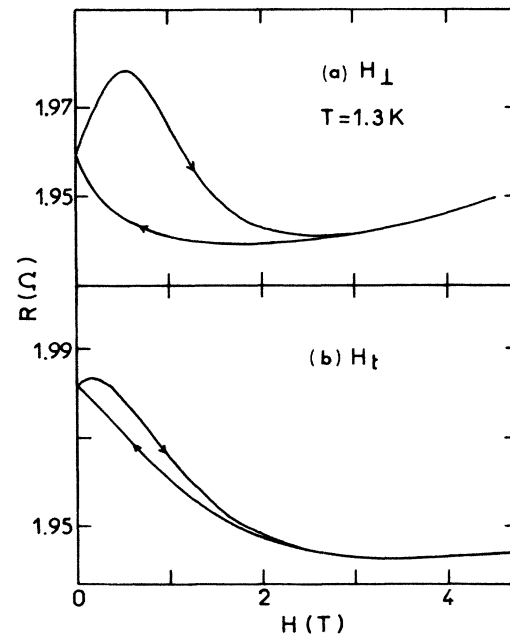


FIG. 1. Magnetoresistance of sample 1: Au/Co (0.32 nm)/Au at 1.3 K for magnetic fields (a) perpendicular (H_{\perp}) and (b) transversal (H_t) (see text). At high fields, the classical parabolic magnetoresistance of metals is observed.

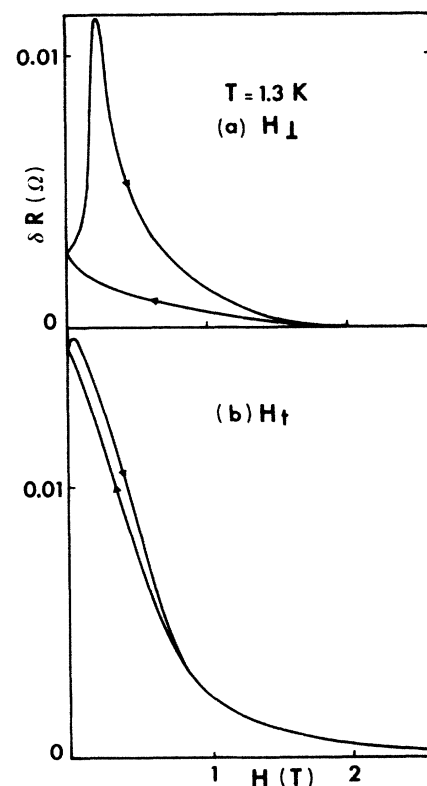


FIG. 2. Magnetoresistance of sample 2: Au/Co (0.76 nm)/Au at 1.3 K for (a) H_{\perp} and (b) H_t . The parabolic magnetoresistance of gold has been subtracted. The minimum value of R is 2.12 Ω .

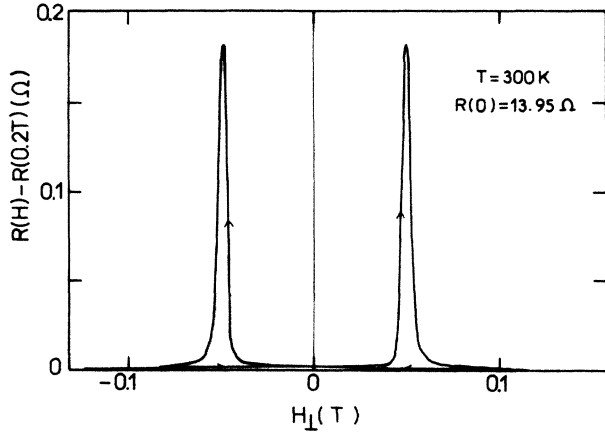


FIG. 3. Room-temperature perpendicular magnetoresistance of sample 3: Au/Co (0.76 nm)/Au (3 nm)/Co (0.76 nm)/Au. The coercive field is $H_c = 493$ G and $\delta R_c/R = 1.3\%$.

to the film ($r=1$). The coercive field H_c is equal to 2 kG at 4.2 K and 490 G at 300 K; $\delta R_c = 0.16$ Ω at 4.2 K and 0.18 Ω at 300 K, giving $\delta R_c/R = 3\%$ at 4.2 K and 1.3% at 300 K. For transverse fields, a very smooth variation of the MR is contrarily observed [Fig. 4(a)]. We have just observed an exactly similar behavior of the MR at room temperature in another cobalt bilayer: Au (25 nm)/Co (0.75 nm)/Au (13 nm)/Co (0.75 nm)/Au (25 nm).

At room temperature, the rotation of the field in the

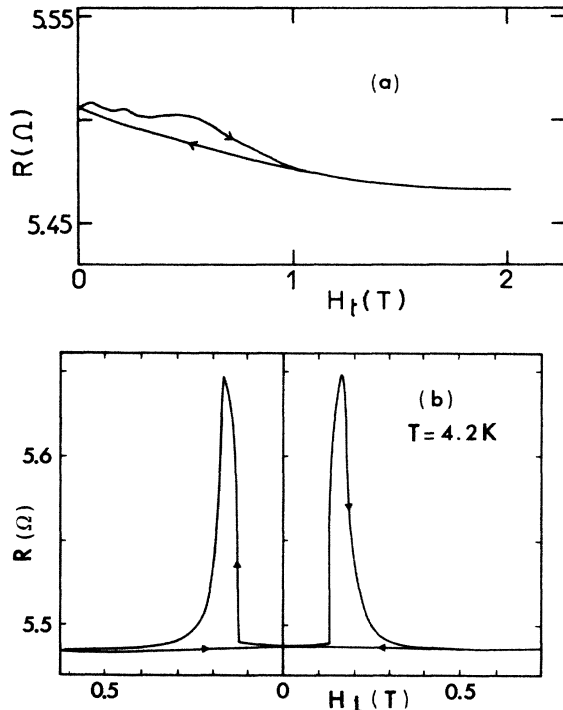


FIG. 4. Magnetoresistance of sample 3 at 4.2 K for (a) H_t and (b) H_{\perp} . Perpendicularly to the film, a coercive field of 2×10^3 G and a maximum magnetoresistance $\delta R_c/R = 3\%$ are observed.

plane perpendicular to the electric current leads to a variation of the coercive field approximately given by $H_c(\theta) = H_c(0)/\cos\theta$ for $\theta < 60^\circ$, where θ is the angle between the applied field and the sample normal. It thus seems that the component of the field perpendicular to the film, i.e., parallel to the easy magnetization axis of this system is the only effective one. The magnetization remains fixed along the perpendicular direction and does not rotate with the field. It only flips from up to down at H_c .

From the experimental results it appears that the high MR after subtracting the parabolic contribution in high field is due to the perpendicular anisotropy of the cobalt. In a perpendicular magnetized Co domain with a mean diameter Λ_D much larger than the Co thickness, the magnetic induction B is given by

$$B = H + 4\pi M_s - H_D, \quad (1)$$

where H is the external field, and where the demagnetizing field H_D is about $-4\pi M_s$; M_s is the saturation magnetization. For $H=0$, B would be equal to zero, but the defects, particularly the interface roughness, produce a weak induction $B = 4\pi M_s \eta$ with $\eta \ll 1$ (for cobalt, $4\pi M_s = 18$ kG). On the other hand, in Bloch walls where the magnetization rotates over 180° over the short distance Λ_w , the induction is of about $4\pi M_s$. We can then conceive three principal mechanisms for the electronic diffusion.

(a) In zero applied field there is a perpendicular magnetic induction $B(r)$ in gold in the vicinity of the cobalt layer, due to the magnetic induction B in the cobalt (1), which deviates the current lines in gold. The Lorentz deviations of the electrons are not compensated by the Hall effect because of the alternating values of $B(r)$.

In order to get a rough estimate of the resulting MR, we will assume that the magnetic induction in gold is approximately constant over distances of the order of Λ_D , and anyway less than the induction inside the cobalt (this assumption is not unrealistic because the thickness of each gold layer is much smaller than the domain size Λ_D). In this case, the resulting MR would be

$$\frac{\delta R_c}{R} \sim \left(\frac{eB}{m} \right)^2 \frac{\lambda^2}{v_F^2}. \quad (2)$$

In our samples, the elastic mean free path λ is 100 nm, the Fermi velocity $v_F = 1.4 \times 10^6$ ms^{-1} . For $\eta = 0.1$, we obtain $\delta R_c/R < 5 \times 10^{-6}$, which is too weak in comparison with the experiments [even with $\eta = 1$, i.e., $B = 4\pi M_s$, formula (2) would give $\delta R_c/R \sim 5 \times 10^{-4}$].

(b) The high induction in the Co walls produces some magnetostriction which deforms the atomic array. An electron which crosses the Co films in a wall is then strongly diffused. The corresponding gold-wall interface resistance is proportional to the number of walls.

(c) The Co atoms laying in gold generate an important density of three-dimensional virtual bound states which can be very near the Fermi level in the walls. An electron crossing a wall undergoes an important resonant diffusion.

If we consider the effects (b) and (c) as a gold-wall in-

terface resistance, we obtain

$$\frac{\delta R_c}{R} \sim \frac{\Delta_w}{\Lambda_D + \Lambda_w} \frac{\lambda}{t} |\langle \cos \phi_D - \cos \phi_w \rangle|, \quad (3)$$

where t is the whole thickness of the sample, ϕ_D and ϕ_w are the angular deviations of an electronic path crossing a domain or a wall, and the average is taken over all the electronic paths. Assuming physically reasonable values $\Lambda_D = 200$ nm, $\Lambda_w = 2$ nm, $|\langle \cos \phi_D - \cos \phi_w \rangle| \sim 0.5$ we obtain $\delta R_c/R \sim 10^{-2}$ which is comparable to the experimental values.

The three mechanisms (a), (b), and (c) are ineffective in the case of in-plane magnetized films because, in that case, the induction in the domains is about the same as in the walls. The detailed calculations of formulas (2) and (3) will be exposed in a forthcoming paper.

In conclusion, we have shown that MR measurements give a simple and easy way of determining unambiguously the easy axis and the coercive field of ferromagnetic metals of a few atomic layers. We have applied this method to carefully characterized sandwiches and bilayers of co-

balt in gold. The perpendicular anisotropy of these ultrathin hcp cobalt films was confirmed and a temperature dependence observed in the thinnest sample, in contrast with the results obtained by Pescia *et al.* in the case of cobalt on copper.¹¹ A possible interpretation of these results in terms of a domain structure induced in the gold by the cobalt magnetization or a gold-wall interface resistance is proposed; for in-plane magnetizations, the MR mechanism is different, as pointed out above. We have observed a strong enhancement of the MR anisotropy in a cobalt bilayer. This phenomenon is attractive for use in miniaturized magnetic field detectors for magnetic recording.

Further work is in progress on MR and magnetization of Au/Co/Au sandwiches with various Co thicknesses and on the effect of gold interlayer thickness in Co bilayers.

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