Ferromagnetic resonance studies of very thin cobalt films on a gold substrate

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Ferromagnetic resonance measurements in Co films of 11.3, 20, and 80 Å thickness sandwiched by Au have been made as a function of the dc magnetic field orientation in a plane perpendicular to the film. These polycrystalline films were measured to have the hcp structure with the c axis perpendicular to the film plane within a few degrees. The experimental results are well explained by a theoretical model where an axial magnetic anisotropy up to the second-order term is included.

I. INTRODUCTION

The specific magnetic phenomena which occur in very thin ferromagnetic crystalline films and at their surfaces (or interfaces) when these are coated by another material, magnetic or nonmagnetic, are of interest from a fundamental point of view. The recent progress in evaporation techniques for preparing ultrathin films and multilayered metallic films[1] has now opened a new field. In such ultrathin films of magnetic material, size effects and surface effects are expected to be revealed in the magnetization and in the magnetic anisotropy.

We have investigated the properties of very thin cobalt films sandwiched by gold. We will present and discuss here a study of the ferromagnetic resonance (FMR) of such thin films, a technique which is particularly suitable for providing information on their magnetic anisotropy (and especially the surface-induced one) as already demonstrated in the past in similar studies.[2] Our cobalt films are polycrystalline, but with a preferred orientation of the crystallites (and a crystallographic structure which has been proven to be hexagonal close packed) so that this FMR study, as a function of film thickness, may be compared to recent ones performed on thin metallic monocrystalline films, such as Fe epitaxially grown on GaAs.[3] Finally we stress the fact that for this hexagonal structure, we have fully taken into account in our theoretical analysis the second-order terms in the magnetic anisotropy, a point which appears essential for the interpretation of the behavior of the thinnest film.

II. THEORETICAL MODEL

The coordinate system used in our calculations of the magnetostatics and the fields for resonance is shown in Fig. 1. The energy density function appropriate to a system with hexagonal structure, up to the second-order term in the magnetic anisotropy, is then given by the expression

$$E=-HM\sin\theta\cos(\varphi_H-\varphi)+\tfrac{1}{2}(4\pi M^2)\sin^2\theta\sin^2\varphi -(K_1+2K_2)\sin^2\theta\sin^2\varphi+K_2\sin^4\theta\sin^4\varphi\ , \quad (1)$$

where the first term represents the Zeeman energy, the second the magnetostatic energy, and the last two terms the axial anisotropy energy with the c axis parallel to the y axis. K_1 and K_2 are the first- and second-order anisotropy constants. The effective values of K_1 and K_2 in Co films depend on the volume magnetocrystalline anisotropy, the surface anisotropy, and the stress anisotropy. Since the dc field H is applied in the xy plane, the static equilibrium position of the magnetization M is given by $\theta_{eq}=\pi/2$ and for φ_{eq} by the relation

$$MH\sin(\varphi_H-\varphi_{eq})=(4\pi M^2-2K_1-4K_2)\sin\varphi_{eq}\cos\varphi_{eq} +4K_2\sin^3\varphi_{eq}\cos\varphi_{eq}\ . \quad (2)$$

In regard to the magnetostatics, the magnetic structure of the Co film *in zero external field* depends critically on K_1 and K_2. Besides the trivial limiting cases, $\varphi_{eq}=0$ (small magnetic anisotropy) and $\varphi_{eq}=\pi/2$ (very high positive

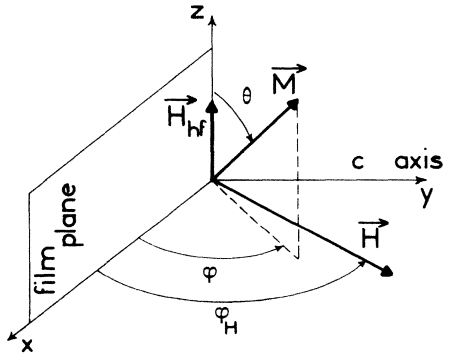

FIG. 1. Orientation of the dc magnetic field **H** and of the magnetization **M** with respect to the coordinate system used in the calculations. The film is parallel to the xz plane.

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value of K_1), there is a solution of a conical[4] structure around the c axis with a half angle α given by

$$\sin^2\alpha=\frac{4\pi M^2-2K_1}{4K_2}\ , \tag{3}$$

where $\alpha=(\pi/2)-\varphi_{eq}$ represents the angle **M** makes with the c axis.

The resonance field H_{res} can be calculated by using, for example, the general equation derived by Smit and Beljers[5]

$$\left[\frac{\omega}{\gamma}\right]^2=\frac{1}{M^2\sin^2\theta}\left[\frac{\partial^2E}{\partial\theta^2}\frac{\partial^2E}{\partial\varphi^2}-\left[\frac{\partial^2E}{\partial\theta\partial\varphi}\right]^2\right]. \tag{4}$$

The second derivatives in Eq. (4) must be evaluated for θ_{eq} and φ_{eq}. We obtain the following expression:

$$\left[\frac{\omega}{\gamma}\right]^2=[H\cos(\varphi_H-\varphi_{eq})+(4\pi M-H_A)\cos2\varphi_{eq}+H_{A2}(3\sin^2\varphi_{eq}\cos^2\varphi_{eq}-\sin^4\varphi_{eq})]$$
$$\times[H\cos(\varphi_H-\varphi_{eq})-(4\pi M-H_A)\sin^2\varphi_{eq}-H_{A2}\sin^4\varphi_{eq}]\ , \tag{5}$$

where $H_{A1}=2K_1/M$, $H_{A2}=4K_2/M$, and $H_A=H_{A1}+H_{A2}$.

Our cobalt films are polycrystalline with an hcp structure of the Co but with a small spread of the c-axis orientation from one crystallite to the other, as evidenced by detailed measurements[6] mentioned below. From the spatial variation of the c axis and the distribution of internal field $H_i=4\pi M-H_A$, one can expect a spread of the resonance fields and consequently two sources of linewidth ΔH besides the one which would arise from an intrinsic damping mechanism. The first contribution corresponds to $\Delta H=\Delta\varphi_H(\partial H_{res}/\partial\varphi_H)$, where $\Delta\varphi_H$ is the assumed angular spread of the c axis, and the second one to $\Delta H=\Delta H_i(\partial H_{res}/\partial H_i)$ for an internal field inhomogeneity ΔH_i. In each case the variation of H_{res} is to be deduced from Eq. (5). Calculations of the angular dependence of ΔH, based on these two expected sources, have been compared to experiments and will be presented in Sec. III.

III. EXPERIMENTAL RESULTS AND INTERPRETATION

The samples studied are Au/Co/Au sandwiches with different thicknesses e of the cobalt layer. They were made by slow evaporation of the metals on a polished glass platelet at $T=300$ K in ultrahigh vacuum (10^{-10} Torr). For the measurements the films are then peeled off from the glass substrate making use of a special varnish. The Au substrate is polycrystalline (mean lateral size 2000 Å), monocrystalline through its total thickness (around 260 Å), with an atomically flat surface perpendicular to the [111] direction. Detailed measurements[6] using transmission electron microscopy and electron diffraction at normal incidence have shown that the Co film and the covering Au layer follow the polycrystalline structure of the initial Au (111) plane, and furthermore that the crystallographic structure of the cobalt is hexagonal close packed with a lattice constant equal to the bulk value and an orientation of the c axis along the [111] direction of the gold, i.e., perpendicular to the film plane.

We carried out FMR measurements at the X band (9.23 GHz) on three such polycrystalline Au/Co/Au films corresponding to the following Co thicknesses: $e=80$ Å, $e=20$ Å, and $e=11.3$ Å. We investigated in particular the FMR spectrum, at $T=291$ K and $T=10$ K, as a function of the orientation of the applied dc field **H** in a plane perpendicular to the film (angle φ_H in Fig. 1) and also followed the evolution of the resonance spectrum versus temperature (in the range 5–300 K) in the parallel geometry ($\varphi_H=0$). The signal detected corresponds to the field derivative of the absorbed power. One observes nearly antisymmetrical lines with an A/B ratio (of the amplitudes of the lobes above and below the baseline) close to but different from 1, which varies in a systematic fashion with the orientation φ_H. The data for the three samples studied, which will be presented and discussed, concern the field for resonance H_{res} and the linewidth ΔH (see Figs. 2–7). These quantities have been derived, respectively, from the field H_0 (intersection with the baseline) and peak-to-peak linewidth $\Delta H_{p.p.}$ after a correction relevant to the observed A/B value. These corrections have been made assuming they are the same as the ones expected for a Lorentzian line shape[7] corresponding to the same A/B ratio. They are only appreciable for the thinnest sample ($e=11.3$ Å) where the A/B value can go up to ~3 around the perpendicular geometry.

It is seen from expressions (2) and (5) that the resonance field H_{res} depends on two parameters, viz., the internal field $H_i=4\pi M-H_A$ and H_{A2}. In the numerical calculations we used the fitted values of H_i and H_{A2} for the two particular geometries: $\varphi_H=0°$ and $\varphi_H=90°$ (except for the 80-Å sample where the line cannot be observed in $\varphi_H=90°$ geometry due to the insufficiency of the available field) for which expression (5) reduces to

$$\left[\frac{\omega}{\gamma}\right]^2=H_{\|}(H_{\|}+4\pi M-H_A)\ , \tag{6}$$

and

$$\frac{\omega}{\gamma}=H_{\perp}-(4\pi M-H_{A1})\ , \tag{7}$$

where $H_{\|}$ and $H_{\perp}$ are the resonance fields for $\varphi_H=0$ and $\varphi_H=90°$, respectively. We assume further that the magnetization in our thin films is equal to that of bulk cobalt. The $4\pi M$ value is then taken as 18 kOe and 17.6 kOe at 10 and 300 K, respectively, since the Curie temperature is very high even for the thinnest film.[8] The fitted values of H_A and H_{A2} for different samples are given in Table I.

TABLE I. The values of the parameters $4\pi M - H_A$ and the anisotropy fields H_A and H_{A2}, defined in Eq. (5), as deduced from a fit to the experimentally observed angular variation of H_{res} for the three samples studied, at two temperatures: $T = 291$ K and $T = 10$ K (see Sec. III). The bulk cobalt values of these parameters, taken from Ref. 9, are given for comparison. The bulk cobalt value of $4\pi M$ has been taken to extract H_A from $4\pi M - H_A$ in the three samples studied.

T (K)	Samples	$4\pi M - H_A$ (kOe)	H_A (kOe)	H_{A2} (kOe)
291	Bulk	7.04	10.56	4.26
	80 Å	9.22±0.18	8.38±0.18	3±1
	20 Å	4.54±0.08	13.06±0.08	0.94±0.14
	11.3 Å	−1.96±0.31	19.56±0.31	1.84±0.71
10	Bulk	3.86	14.14	4.73
	80 Å	11.66±0.27	6.34±0.27	3±1
	20 Å	5.8 ±0.1	12.2 ±0.1	1.44±0.16
	11.3 Å	−0.9 ±0.37	18.9 ±0.37	0.68±0.56

The values for bulk cobalt are taken from Ref. 9.

For the 80-Å film at 291 K, one finds that the H_A and H_{A2} values are a little smaller than the bulk ones. Furthermore H_A is reduced at $T = 10$ K, compared to room temperature, contrary to the strong increase observed in the bulk. This latter feature is common to the three samples which show a monotonic decrease of the resonance field $H_{||}$ with decreasing temperature. This may be ascribed to internal stresses produced by different thermal contractions of the Au and Co layers. The effect of these internal stresses is also apparent in the line broadening of these polycrystalline films, which show a monotonic increase of their linewidth ΔH with decreasing temperature. However, the strong angular dependence of the linewidth observed at $T = 291$ K can be well accounted for essentially in terms of the effect of the distribution of the c-

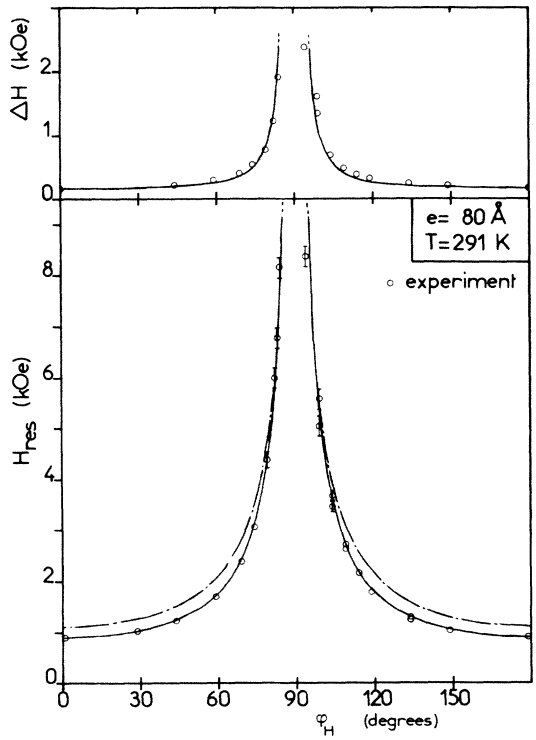

FIG. 2. Resonance field H_{res} and linewidth ΔH, at $T = 291$ K, as a function of the dc magnetic field orientation (angle φ_H) in the 80-Å sample. The solid line for H_{res} is a calculated curve, from Eqs. (5) and (2), using $4\pi M - H_A = 9.22$ kOe (fitted in parallel geometry) and $H_{A2} = 3$ kOe (chosen to give the best overall angular fit). The dashed curve corresponds to the bulk cobalt values (Table I) of these parameters. The solid line for ΔH is a calculated curve, according to the expression $\Delta H = \Delta H_0 + \Delta\varphi_H(\partial H_{res}/\partial\varphi_H)$, using $\Delta H_0 = 0.16$ kOe (the fitted value in $\varphi_H = 0$) and assuming an angular spread of the c axis $\Delta\varphi_H = 2°$ (see Sec. II).

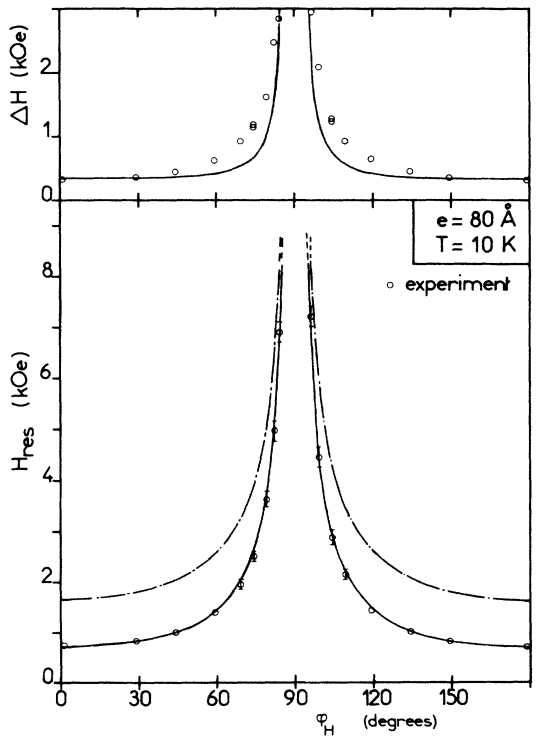

FIG. 3. Resonance field H_{res} and linewidth ΔH, at $T = 10$ K, as a function of the dc field orientation (angle φ_H) in the 80-Å sample. The solid line for H_{res} is a calculated curve, using $4\pi M - H_A = 11.66$ kOe and $H_{A2} = 3$ kOe, according to the procedure detailed in Fig. 2. The dashed curve is the one expected for bulk cobalt. The solid line for ΔH is a calculated curve (as in Fig. 2), using $\Delta H_0 = 0.32$ kOe and $\Delta\varphi_H = 2°$.

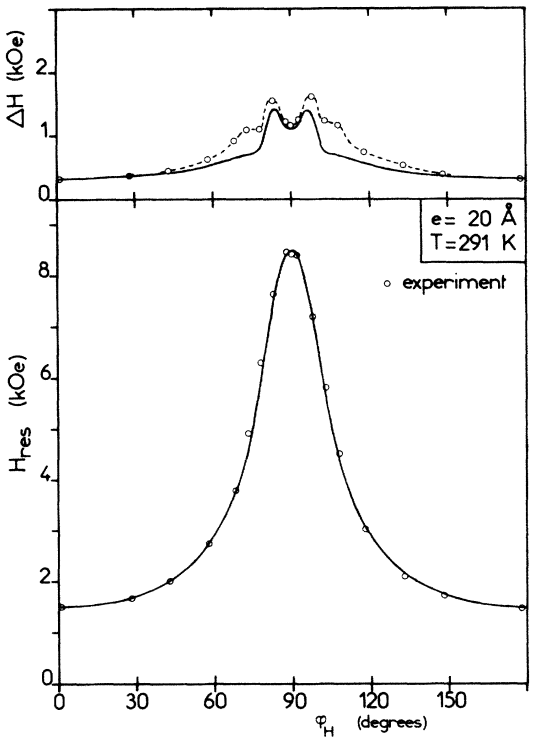

FIG. 4. Resonance field H_{res} and linewidth ΔH, at $T=291$ K, as a function of the dc field orientation (angle φ_H) in the 20-Å sample. The solid line for H_{res} is a calculated curve, from Eqs. (5) and (2), using $4\pi M - H_A = 4.54$ kOe and $H_{A2} = 0.94$ kOe as deduced from a fit to experiment through Eqs. (6) and (7). The solid line for ΔH is a calculated curve, according to the expression $\Delta H = \Delta H_0 + \Delta\varphi_H(\partial H_{res}/\partial\varphi_H) + \Delta H_i(\partial H_{res}/\partial H_i)$, using $\Delta H_0 = 0.11$ kOe (the fitted value in $\varphi_H = 0$), $\Delta\varphi_H = 2°$ and assuming an internal field inhomogeneity $\Delta H_i = 1$ kOe (see Sec. II).

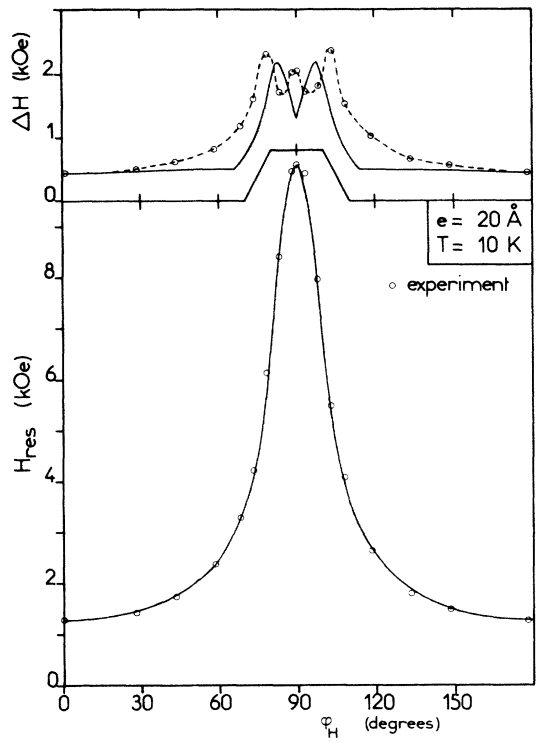

FIG. 5. Resonance field H_{res} and linewidth ΔH, at $T=10$ K, as a function of the dc field orientation (angle φ_H) in the 20-Å sample. The solid line for H_{res} is a calculated curve, using $4\pi M - H_A = 5.8$ kOe and $H_{A2} = 1.44$ kOe, according to the procedure detailed in Fig. 4. The solid line for ΔH is a calculated curve (as in Fig. 4), using $\Delta H_0 = 0.295$ kOe, $\Delta\varphi_H = 2°$ and $\Delta H_i = 1$ kOe.

axis orientations of the different crystallites as shown in Fig. 2. At low temperature the poorer quality of the fit to the observed ΔH indicates a more complicated behavior (Fig. 3), probably due to the stress effects.

For the 20-Å sample at 291 K, the deduced H_A value is now found to be greater than the bulk one (Table I), contrary to the thicker sample. For this thinner sample the increase in H_A is likely to arise from the growing contribution of the surface anisotropy,[10] which in the present case has axial symmetry. It also appears that this surface-induced anisotropy essentially enhances the K_1 value. At low temperature the H_A value is not very different from the bulk one, revealing a strong competition between surface anisotropy and stress effects. As shown by the different curves of Fig. 4, the linewidth would arise from both the spatial fluctuations of the c axis and the spread of internal fields. Indeed, the former gives rise to a maximum of ΔH near the perpendicular direction (c axis) whereas the shoulder observed near the maximum and the important ΔH value for $\varphi_H = 90°$ are manifestations of the latter mechanism. Again, at low temperature, the complex behavior of the angular variation of the observed linewidth (Fig. 5) may be associated, as already mentioned, to the effect of internal stresses.

Finally, for the 11.3-Å sample which contains, on the average, only about six atomic layers, one can expect a spectacular manifestation of the surface anisotropy. The unusual angular dependence of H_{res} (the higher value of H_{res} in $\varphi_H = 0°$ compared to $\varphi_H = 90°$ and the presence of a minimum of H_{res} for an intermediate value of φ_H) results directly from the fact that H_A is now greater than $4\pi M$ and H_{A2} remains positive and appreciable (Fig. 6). Indeed, the observation of such a minimum of H_{res} can only be understood if we include in the theoretical analysis the second-order terms as described by H_{A2}. If we take the values of H_A and H_{A2} given in Table I (which represent average values of these parameters for our polycrystalline sample) we find that the numerator of expression (3) is very small and negative. This means that one is in the limiting case where the magnetization begins to align itself along the c axis. However, taking into account the error bars on the listed values of H_A and H_{A2}, other sets of values of these parameters can easily lead to a different solution of conical structure (see Sec. II) for which the presence of second-order effects is also essential. Moreover, the expected distribution of H_A and H_{A2} values in our polycrystalline sample should give rise to a complex magnetic structure with a ferromagnetic component along the c axis. The effect of this strongly

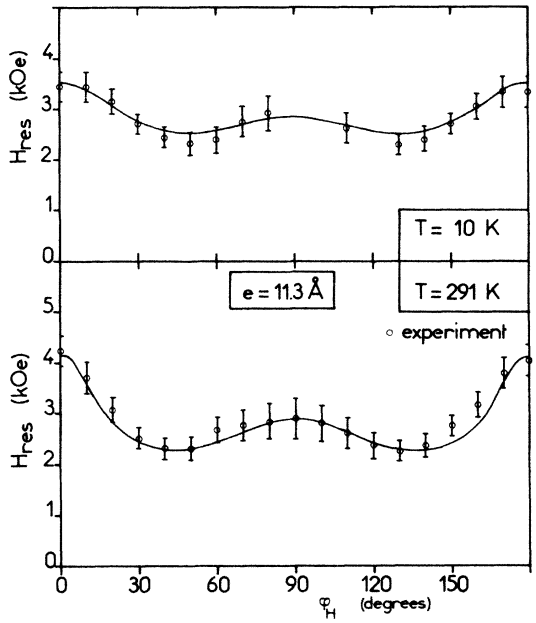

FIG. 6. Resonance field H_{res}, respectively at $T=291$ K and $T=10$ K, as a function of the dc field orientation (angle φ_H) in the 11.3-Å sample. At each temperature, the solid line for H_{res} is a calculated curve, from Eqs. (5) and (2), using $H_i=4\pi M-H_A$ and H_{A2} as deduced from a fit to experiment through Eqs. (6) and (7). At $T=291$ K we used $H_i=-1.96$ kOe and $H_{A2}=1.84$ kOe and at $T=10$ K, $H_i=-0.9$ kOe and $H_{A2}=0.68$ kOe.

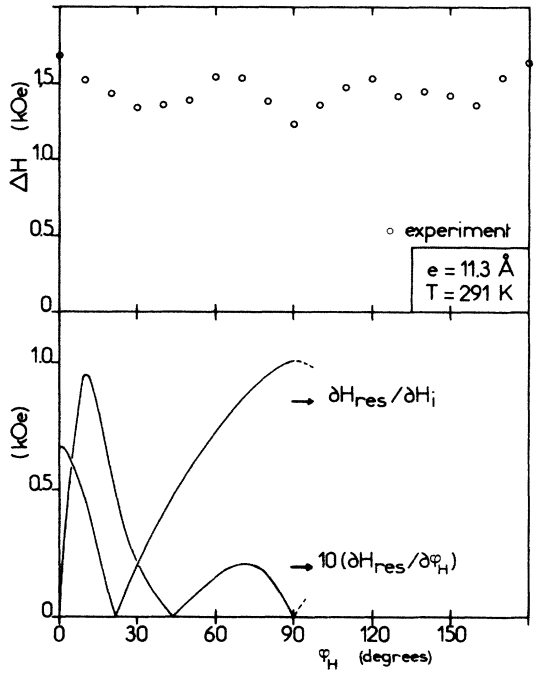

FIG. 7. Linewidth ΔH, at $T=291$ K, as a function of the dc field orientation (angle φ_H) in the 11.3-Å sample (experimental results in the upper part of the figure). The two solid curves in the lower part of the figure correspond to the calculated slopes of the H_{res} curve, $(\partial H_{res}/\partial H_i)$ and $(\partial H_{res}/\partial \varphi_H)$, respectively. These variations of H_{res} are deduced from Eqs. (5) and (2), using for H_i and H_{A2} the values which fit the H_{res} curve at $T=291$ K (see Fig. 6). The quantities plotted have been converted into linewidths (kOe unit) using $\Delta H_i=1$ kOe and $\Delta \varphi_H=10°$.

enhanced crystalline anisotropy, observed for this thinnest sample, is consistent with the observation[8] of a remanent magnetization predominantly along the film normal for the thinnest Co layers, contrary to the observation in the thicker samples, where it lies in the film plane.

The distribution of H_A and H_{A2}, i.e., of the internal field, should lead to important contributions to the linewidth. This can first explain the very large magnitude of ΔH observed in $\varphi_H=0$ for the thinnest sample as compared to the thicker ones. Secondly, the relatively small angular variation of ΔH (Fig. 7) is consistent with the expected combined effects of the distribution of c-axis orientations and of internal fields when one uses for this simulation the listed values of $4\pi M-H_A$ and H_{A2} which fit the angular variation of H_{res}.

IV. DISCUSSION

Following the method of analysis previously used, in particular that in Refs. 11 and 12, for the effects arising from a surface-induced magnetic anisotropy in thin ferromagnetic films, one may express the total magnetic anisotropy energy density of ultrathin films (whose thickness e is smaller than a characteristic length[12] associated with the exchange interaction) as

$$K=K_v+\frac{1}{e}(K_s^{(1)}+K_s^{(2)}) . \tag{8}$$

This corresponds to a sum of two contributions: a homogeneous volume contribution K_v, and a surface-induced one, inversely proportional to the thickness e and characterized by surface energy densities $K_s^{(1)}$ and $K_s^{(2)}$, one for each interface of the magnetic film. In the present case we are dealing with a symmetric interface with gold, and a magnetic interface anisotropy energy $K_s^{(i)}$ of axial symmetry is expected to arise from the hcp structure of the Co film (with the c axis perpendicular to the film plane) so that one can write, for either interface

$$K_s^{(i)}=K_s \sin^2\alpha , \tag{9}$$

where α is the angle the magnetization **M** makes with the film normal.

Looking at the deduced values of the anisotropy fields H_A and H_{A2}, displayed in Table I for the three samples studied, one notices first that there is no clear dependence of H_{A2} on the film thickness. As was already mentioned, it appears that the surface-induced anisotropy essentially enhances the first-order anisotropy constant K_1 with reduced thickness so that, in Fig. 8, we have plotted H_{A1} as a function of $1/e$ at the two temperatures studied. Keeping in mind that this study concerns only three different thicknesses of the Co film, one sees that a linear variation of H_{A1} versus $1/e$ describes our data reasonably well. From the slope of these lines, according to Eq. (8), the value of the interface anisotropy constant K_s can be de-

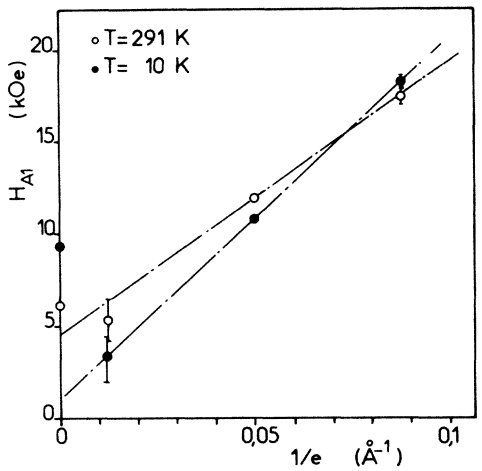

FIG 8. Anisotropy field $H_{A1}=2K_1/M$, deduced from the values of the parameters displayed in Table I, plotted as a function of the reciprocal of the Co thickness e for the two temperatures $T=291$ K and $T=10$ K. The symbols on the vertical coordinate axis correspond to the bulk Co values.

duced and is found to be $K_s=+0.5$ and 0.7 erg cm^{-2} at 291 and 10 K, respectively. These values of K_s found for the interface Co(0001)/Au(111) correspond to an interface anisotropy higher in magnitude and opposite in sign compared to the one ($K_s \sim -0.2$ erg cm^{-2}) recently determined[13] experimentally for Ni(111) films in contact with various metals. As seen from Fig. 8, even at low temperature where stress effects are important, H_{A1} increases linearly with $1/e$. This implies that the stress-induced anisotropy, nearly independent of the film thickness, corresponds to a volume effect. The increase of the magnitude of K_s observed when the temperature is decreased is characteristic of an anisotropy of magnetocrystalline origin. Moreover, the high values of K_s found in these Co films appear consistent with the fact that the Co moment remains close to the bulk value even when the film thickness is reduced down to 4 Å in our samples.[8]

A second observation that can be drawn from Fig. 8 is that an extrapolation of the straight lines to $1/e=0$ leads to H_{A1} values different from the bulk cobalt ones. As was already pointed out in Ref. 12, such a difference is compatible with the presence in these thin films of an additional uniaxial volume anisotropy of the same form as (9), i.e., $K_u \sin^2\alpha$ (with the axis $\hat{u}$ parallel to the film normal). Such a uniaxial anisotropy may result from a volume stress-induced anisotropy, as mentioned above, or more specifically with the growth conditions of the Co films. In particular we suggest that such a contribution may arise, in our cobalt films, from the presence of a weak density of stacking faults undetected in the electron diffraction measurements of Ref. 6. Explicitly the point is as follows. In hcp Co, adjacent layers A,B of atoms in (0001) planes are displaced with respect to each other in such a way that the structure can be viewed as a succession of $ABABAB\ldots$ planes. In fcc Co the (111) planes are similar to those A,B planes but the pattern repeats every third layer and is thus represented by the succession of $ABCABC\ldots$ planes. It is clear that a stacking fault in hcp Co like $ABCABAB$ or $ABCBABA$ would strongly affect two adjacent atomic layers since the involved atoms are now in a cubic environment. As a consequence of the presence of such stacking faults in our Co films the average value of K_1 is expected to decrease.

V. CONCLUSION

In conclusion, the FMR study of these very thin Co(0001) films sandwiched by Au has revealed the increasing role of the crystalline surface anisotropy with the reduction of the Co layer thickness. Indeed, for our thinnest Co film ($e=11.3$ Å) this uniaxial anisotropy is getting so large that it overcomes the effect of the demagnetizing field and leads to an equilibrium orientation of the magnetization out of the film plane. Besides, for this system with hexagonal structure, the comparison with experimental results has stressed the need to take into account in the theoretical analysis the presence of second-order terms in the crystalline anisotropy. Further studies of similar Co films on Cu substrates are now in progress.

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