RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 40, NUMBER 10 1 OCTOBER 1989

Magnetic aftereffect in ultrathin ferromagnetic films

G. Bayreuther
Institut für Angewandte Physik, Universität Regensburg, Universitätsstrasse 31, Postfach 397, D-8400 Regensburg, Federal Republic of Germany

P. Bruno
Institut d'Electronique Fondamentale, Bâtiment 220, Université de Paris-Sud, F-91405 Orsay CEDEX, France

G. Lugert and C. Turtur
Institut für Angewandte Physik, Universität Regensburg, Universitätsstrasse 31, Postfach 397, D-8400 Regensburg, Federal Republic of Germany

(Received 31 July 1989)

By using the recently developed alternating-gradient magnetometer, that combines very high sensitivity and measurement rapidity, we have made observations of magnetic aftereffect in ferromagnetic films of a few atomic layers. These Co films have a perpendicular easy axis due to a large interface anisotropy and square hysteresis loop. The study of the field dependence of the relaxation rate gives information on the activation energy. The results are discussed in connection with the possible existence of domains and wall motion.

It is well known, after Néel,[1] that magnetic aftereffect due to thermal activation is a general property of all ferromagnetic materials. In bulk materials, the time scale for magnetic aftereffect is often so large that aftereffect cannot be observed. On the other hand, when the dimensions are very small, aftereffect becomes very important (small particles, rock magnetism). One can therefore raise the question of whether aftereffect can be observed when *one* dimension only is very small, i.e., in ultrathin films.

In order to be able to observe aftereffect in ultrathin films, one has to fulfill the following conditions: (1) Because of the ultralow thickness of the samples, one must use a very sensitive magnetometer. (2) One must be able to measure rapidly enough to follow the time dependence of the magnetization. The vibrating sample magnetometer is fast but not sensitive enough, whereas the torsion oscillation magnetometer and the SQUID magnetometer are very sensitive but not fast enough.

The recently developed alternating-gradient magnetometer[2] (AGM) is both very sensitive and relatively fast, and is therefore well suited for studying dynamical effects in ultrathin films. We have used a laboratory built AGM working at room temperature to study the time dependence of magnetization in Au/Co/Au sandwiches with ultralow Co thickness (a few atomic layers), which, due to a large interface anisotropy, have a perpendicular easy axis below 12 Å and a square hysteresis loop.

Our measurements unambiguously show the presence of an unusually important aftereffect in Co ultrathin films. To our knowledge this is the first observation of such effects in ultrathin films. The field dependence of the relaxation rate gives information on the activation energy. It is shown that the time stability of the remanent magnetization varies by several orders of magnitude in a narrow thickness range. We discuss the mechanism of magnetization reversal, and suggest that the latter is due to wall motion.

The existence of walls and domains in the monolayer range is currently of great interest. It has been recently studied theoretically by Yafet and Gyorgy.[3] However, on the experimental point of view, this question remains open. Our method for studying the dynamical behavior of the magnetization proves to be very efficient and provides useful information on the dynamics of (possibly existent) Bloch walls in the monolayer range.

I. THE SAMPLES

A detailed description of the preparation and structural characterization of the Au/Co/Au sandwiches can be found in Ref. 4. The samples are grown on glass by thermal evaporation in ultrahigh vacuum. After annealing, the gold substrate, about 250 Å thick, is polycrystalline, with (111) texture and a crystallite size around 2000 Å. The surface is constituted of atomically flat terraces of width 200 to 300 Å, separated by monoatomic steps. The Co film grows expitaxially on Au, with a hcp (0001) structure. The growth is not layer by layer, so that the second interface has a larger roughness than the first one. The film is then covered by a Au layer of about 250 Å in order to prevent corrosion of the Co. This Au layer grows epitaxially on Co, so that the crystalline coherence is kept through the total thickness. For the magnetic measurements, the Au/Co/Au sandwiches are peeled off from the glass with amyl-acetate varnish.

The magnetic properties of the samples have been studied by ferromagnetic resonance and SQUID magnetometry. These experiments, reported in previous publications,[5,6] show the presence of a large interface anisotropy competing with the demagnetizing anisotropy, and driving the easy axis out of plane for Co thicknesses below 12 Å.

II. EXPERIMENTAL RESULTS

The perpendicular hysteresis loop of a 8.1-Å-thick film is shown in Fig. 1 for two different sweeping times (2 and 45 min). One first observes the high remanence and the squareness of the loop, indicating the out-of-plane easy axis. The negative slope in high field is due to the diamagnetism of the sample holder. The important point is the influence of the sweeping time: The coercive field decreases as the sweeping time is increased (respectively 650 and 560 Oe for 2 and 45 min). This is clear evidence for the presence of magnetic aftereffect in these ultrathin films.

In order to study the time dependence of the macroscopic magnetization M, we have made the following measurements: The sample is first saturated in a positive field along its easy axis; the field is then continuously decreased and stopped at a given negative value, and the time variation of the magnetization is recorded in this constant field. Figure 2 shows the time variation of the magnetization for a 5.4-Å-thick film, at different values of the applied field. It appears that the relaxation rate of the magnetization increases rapidly as the absolute value of the field is increased. This strongly suggests that the aftereffect is due to thermal activation of magnetization reversal.[7] As a matter of fact, the energy barriers must decrease when the absolute value of the field is increased.

If we have barriers of energy E_A, the relaxation of the magnetization will be

$$M(t)-M_\infty=[M(t=0)-M_\infty]\exp(-t/\tau)\,, \qquad (1)$$

where the temperature dependence of the relaxation time τ should follow an Arrhenius law:

$$\tau=\tau_0\exp(E_A/k_BT)\,. \qquad (2)$$

The equilibrium value of the magnetization (or anhysteretic magnetization) M_∞ depends on the field, and is not known in our experiments. We will assume that it is $+M_S$ $(-M_S)$ when the field is positive (negative). Because of the squareness of the loop, and because we make

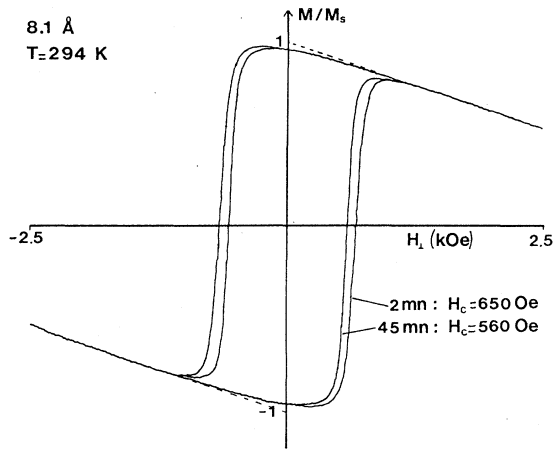

FIG. 1. Perpendicular hysteresis loop of a 8.1-Å-thick film for sweeping times of 2 and 45 min.

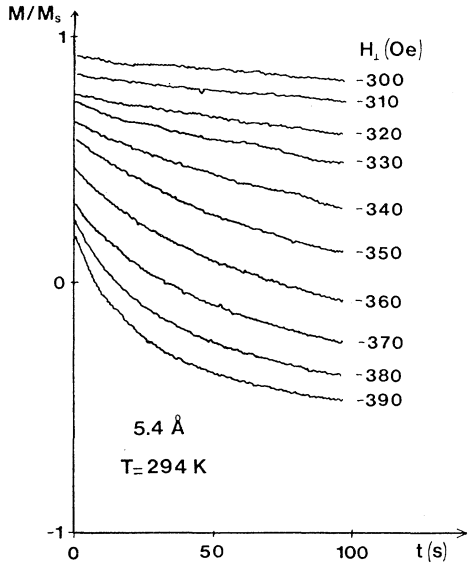

FIG. 2. Relaxation of the magnetization of a 5.4-Å-thick film for different values of the field.

measurements in the neighborhood of the coercive field, this is probably a good approximation.

When there is a distribution of activation energies of width ΔE_A, the magnetization relaxation is quasilogarithmic if $\Delta E_A \gg k_BT$, and quasiexponential if $\Delta E_A \ll k_BT$.[8] Figure 3 shows the relaxation of the magnetization for the 8.1-Å film in a field of -500 Oe. It appears that the relaxation is neither exponential [Fig. 3(a)] nor logarithmic [Fig. 3(b)]. This is due to the fact that ΔE_A is of the same order of magnitude as k_BT. Indeed, it can

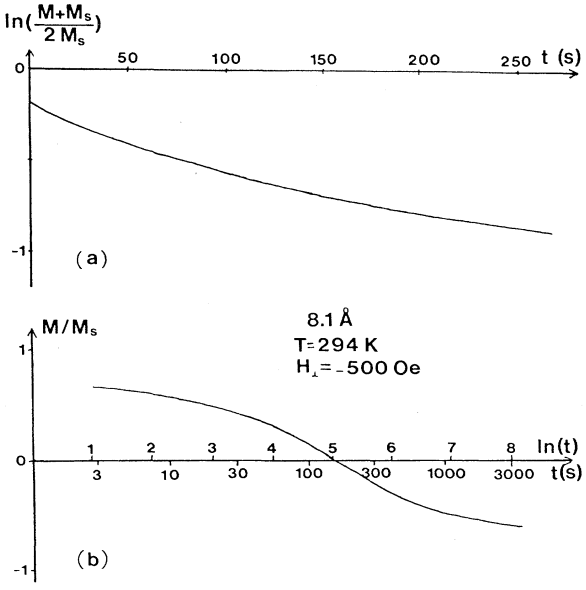

FIG. 3. Time dependence of the magnetization of the 8.1-Å film for $H=-500$ Oe. (a) $\ln[(M+M_S)/2M_S]$ vs t. (b) M/M_S vs $\ln(t)$.

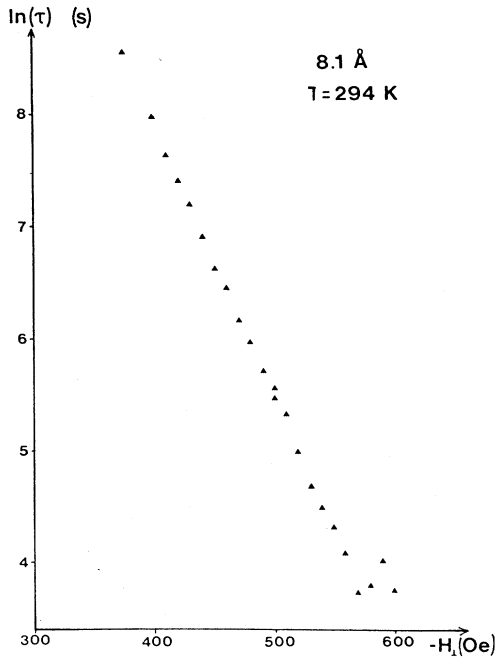

FIG. 4. Plot of $\ln(\tau)$ vs H for the 8.1-Å film.

be shown[9] that the maximum value of the slope in a M-vs-$\ln(t)$ plot yields a measure of the ratio $\Delta E_A/k_BT$, which, in the present case, is found to be $\Delta E_A/k_BT \approx 6$. However, for the early stage of the relaxation, one can always define an initial relaxation time, which corresponds to an average activation energy. We therefore chose to analyze the experiments just by measuring this initial relaxation time.

We have plotted in Fig. 4 the logarithm of the relaxation time versus the field, for the 8.1-Å film. One observes that the points can be fitted very nicely by a straight line over a wide range. Only the points corresponding to high values of the field deviate from the straight line: Actually, in these cases the relaxation is so fast that we cannot observe the early stage of the magnetization reversal. This linear behavior is observed for all of the samples.

By making a linear extrapolation down to zero applied field, we can obtain the stability time $\tau(H=0)$ of the remanent magnetization. The latter is shown in Table I. It appears that this stability time can vary by several orders of magnitude in a very narrow thickness range. This is due to the exponential dependence of $\tau(H=0)$ with respect to the activation energy: A small variation of E_A from one sample to another can therefore result in a tremendous change of $\tau(H=0)$.

TABLE I. Characteristics of the samples, stability time of resonance $\tau(H=0)$, and "Barkhausen length" l_B.

Sample	Thickness (Å)	Easy axis	$\tau(H=0)$	l_B (Å)
1	4.1	⊥	5.5 h	400
2	5.4	⊥	11.4 yrs	480
3	8.1	⊥	1.3 yrs	300
4	9.5	⊥	14 d	280
5	22.2	∥	9.3 months	310

III. DISCUSSION

From Eq. (2), we have $\ln(\tau)=\ln(\tau_0)+E_A/k_BT$; assuming that $\ln(\tau_0)$ depends only slightly on H, as compared to E_A, the above results show that the activation energy depends linearly on the field, i.e., it can be written (for $H<0$)

$$E_A=V_BM_S(H_A+H). \tag{3}$$

In order to explain this behavior, we have to discuss the mechanism of magnetization reversal. There are two possible mechanisms for magnetic reversal: (i) individual reversal of the magnetization of more or less independent particles; and (ii) magnetization reversal by wall motion.

In mechanism (i), for particles of volume V and effective anisotropy field H_K, the activation energy is (for $H<0$)[10]

$$E_A=VM_S\tfrac{1}{2}(H_K+H)^2/H_K \approx VM_S(\tfrac{1}{2}H_K+H) \tag{4}$$

for $H \ll H_K$. The activation energy therefore follows Eq. (3), with $H_A=\frac{1}{2}H_K$ and $V_B=V$.

In mechanism (ii), we have to consider the obstacles to wall motion. According to Néel,[11] these can be due to inhomogeneities of the wall energy. In that case, one can show[9] that the activation energy is of the form given in Eq. (3), where H_A is related to the propagation field without thermal activation and where V_B is the volume of the obstacles.

This shows that both mechanisms would lead to the observed behavior. In both cases, V_BM_S is the elementary magnetization reversal, i.e., it corresponds to Barkhausen jumps, and we can call V_B the "Barkhausen volume." Similar interpretations have been given for aftereffect in amorphous ferromagnets.[12,13]

The slope of the straight line in Fig. 4 yields V_BM_S/k_BT. Assuming the bulk value for M_S, we can thus evaluate V_B. We have reported in Table I the "Barkhausen length" $l_B=\sqrt{(V_B/h)}$ (h is the Co thickness).

For all the samples, l_B is in the range of 300–500 Å. The weak variation of l_B from one sample to another suggests that it is actually a relevant parameter of the problem.

From the point of view of hypothesis (i) for the magnetization reversal mechanism (single-particle reversal), l_B would be the average diameter of the "magnetic grains," and should be related to the size of the "crystallographic grains" ($D \approx 2000$ Å).

On the other hand, within hypothesis (ii) (magnetization reversal by wall motion), l_B would be related to the size of the obstacles impeding the wall displacements; the roughness of the films results in obstacles, the size of which is given by the correlation length ξ of the interfacial irregularities ($\xi \approx 200$ Å).

We therefore suggest that the magnetization reversal occurs by wall motion, and that the thermal activation is responsible for the observed aftereffect. However, a complete interpretation of the present experiments would need the direct observation of the walls, which still remains a challenge for films of a few atomic layers.

ACKNOWLEDGMENTS

We are grateful to Dr. D. Renard (Institut d'Optique, Orsay) for the preparation and structural characterization of the samples. This collaboration was financially supported by a PROCOPE grant. Institut d'Electronique Fondamentale is Unité Associée No. 22 of Centre National de la Recherche Scientifique.

[1]L. Néel, J. Phys. Radium **12**, 339 (1951).

[2]P. J. Flanders, J. Appl. Phys. **63**, 3940 (1988).

[3]Y. Yafet and E. M. Gyorgy, Phys. Rev. B **38**, 9145 (1988).

[4]D. Renard and G. Nihoul, Philos. Mag. B **55**, 75 (1987).

[5]C. Chappert, K. Le Dang, P. Beauvillain, H. Hurdequint, and D. Renard, Phys. Rev. B **34**, 3192 (1986).

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

[7]Diffusion aftereffect due to wall stabilization can be excluded by the following observation: Starting from saturation, the field is first decreased so as to reach a partially reversed magnetization, and then set at zero for a given waiting time. If stabilization of walls would occur, the field necessary to continue the magnetization cycle in the same direction (i.e., the field necessary to unpin the walls) would increase with increasing waiting time. This is definitely not observed. For a waiting time as long as 60 min, the coercive field remains unchanged.

[8]L. Néel, J. Phys. Radium **13**, 249 (1952).

[9]P. Bruno, Ph.D. thesis, Université Paris-Sud, Orsay, 1989 (unpublished).

[10]L. Néel, Ann. Géophys. **5**, 99 (1949).

[11]L. Néel, Ann. Univ. Grenoble **22**, 299 (1946).

[12]B. Barbara and M. Uehara, in *Rare Earths and Anticides—1977,* edited by W. D. Corner and B. K. Tanner, IOP Conference Proceedings No. 37 (Institute of Physics, Bristol and London, 1978), p. 203.

[13]K. Ohashi, H. Tsuji, S. Tsumashima, and S. Uchiyama, Jpn. J. Appl. Phys. **19**, 1333 (1980).