Spin-reorientation transition in ultrathin Tb/Co films

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(Received 14 July 1995)

A series of terbium (0–6 Å)/Co (16 Å) films deposited on Ru(0001) under ultrahigh vacuum conditions and covered by 40 Å Ru have been investigated *ex situ* by means of the magneto-optic Kerr effect (MOKE). Combined polar and longitudinal loops (dc-MOKE) show a reversible rotation of the remanent magnetization from out-of-plane to in-plane with increasing temperature. The reorientation is continuous and occurs without loss of long-range order. It is discussed in terms of the competition between dipolar demagnetization energy and second- and fourth-order magnetocrystalline anisotropy contributions. The switching of the easy axis of magnetization is confirmed by the susceptibility peak observed in a 10 Oe oscillating field (ac-MOKE) applied perpendicular to the film plane. The transition temperature varies from 323 K (1 Å Tb) to 277 K (6 Å Tb).

The magnetism of ultrathin films has been widely studied both theoretically and experimentally.[1] It is well known from the Mermin-Wagner theorem[2] that an isotropic two-dimensional (2D) Heisenberg system presents no long-range order at finite temperature. Nevertheless Bander and Mills[3] showed that long-range order in a 2D Heisenberg system can be stabilized by arbitrarily small magnetic anisotropies. Intrinsic anisotropies are always present in real ultrathin films and govern the magnetization direction of the films. In very thin magnetic films the surface anisotropy can induce a perpendicular easy axis of magnetization whereas for thicker films the magnetostatic anisotropy dominates and favors in-plane magnetization.[4–15] The direction of the easy axis of magnetization can also be modified by varying the temperature of the film.[11–21] The mainly discussed hypotheses in the literature on the origin of the rotation of the magnetization are the competition between the dipolar and magnetocrystalline anisotropies,[18,19] the strong fluctuations in a 2D Heisenberg system,[17] and the anisotropy in the fluctuations of the magnetization direction.[16] An interesting and still open question is what happens in the transition region where the easy axis rotates from out-of-plane toward in-plane. Some theoretical studies[17,18,21] predict that the reorientation of the magnetization as a function of the temperature occurs via a loss of long-range order. Others[19,20,22] described a continuous rotation of the magnetization from out-of-plane to in-plane with an increase of the temperature or the thickness. Experimentally, contradictory results have been obtained for the Fe/Cu(100) (Refs. 9–11, 13, and 15) and Fe/Ag(100) (Refs. 12 and 13) systems. In the reorientation region a vanishing[9,11,13,15] as well as a finite[10,12] remanence has been measured. On the other hand, the magnetic studies on the Co/Au(111) (Refs. 4 and 23) system as a function of the cobalt thickness agree, showing that the reorientation occurs within a thickness interval for which the remanence is nonzero both out-of-plane and in-plane.

Recently, it was shown that the dc (Ref. 25) and ac (Refs. 26–29) magneto-optic Kerr effect (MOKE) techniques are powerful tools to investigate ultrathin film magnetism. In particular, the ac-susceptibility MOKE measurements can determine accurately the ferro-paramagnetic transition.[26–29] The ac-MOKE technique has also been used to detect spin-reorientation transitions. An in-plane reorientation,[29] below the Curie temperature has been observed by means of longitudinal ac-MOKE. An ac-susceptibility peak observed in transverse geometry has been used to identify out-of-plane components of the magnetization for thick Gd films.[27]

In this paper we investigate the reorientation of the easy axis in ultrathin Tb/Co films as a function of temperature and terbium thickness. This class of materials, that is, rare-earth (RE)/transition-metal (TM) compounds and multilayers, are very valuable media for magneto-optic recording due to the occurrence of perpendicular anisotropy and enhanced Kerr rotation.[30] While in previous studies the main focus was the ferrimagnetic coupling mechanism at the RE/TM interface, we address the changes in the anisotropy of the Co layer due to the adsorption of Tb monolayers. Strong effects can be expected due to the crystal field splitting of the RE atomic-like $4f$ configuration. The results unambiguously show that a reversible complete and continuous spin transition takes place as a function of temperature. Moreover, we show that the spin-reorientation temperature can be finely selected by varying the thickness of the Tb cover layer.

The films are deposited by molecular beam epitaxy (MBE) on a smooth and clean hcp (0001) 150-Å-thick Ru buffer layer on freshly cleaved mica substrates below 2×10^{-10} mbar. The film growth is monitored by high-reflection energy electron diffraction and the thickness is calibrated with a quartz microbalance. The substrate temperature is held at 240 K to minimize diffusion at the Co-Ru interface and to avoid interdiffusion between terbium and cobalt.[31] The cobalt grows epitaxially on the Ru buffer with the hexagonal basal plane parallel to the surface.[32,33] Tb overlayers of up to 5 Å on Co induce a 3×3 reconstruction of the Co surface, which is in agreement with previous low-energy electron diffraction, Auger, and photoemission studies for Tb grown on Co single crystal surfaces. This indicates a two-dimensional growth of ordered 1–2 monolayers (ML)

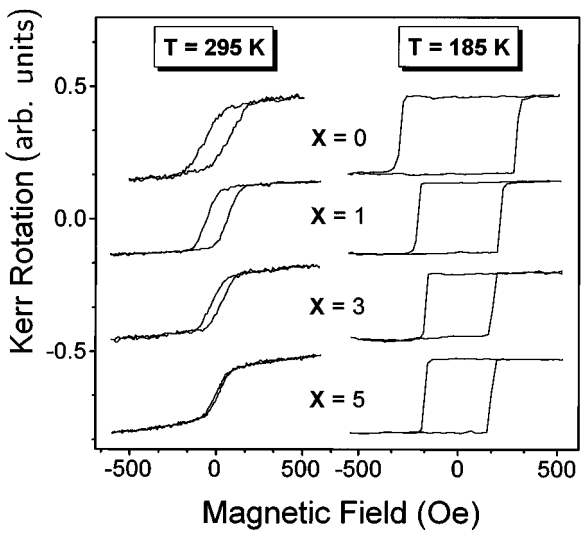

FIG. 1. Polar hysteresis loops for the 16 Å Co/X Å Tb films ($X=0,1,3,5$ Å) at 295 and 185 K. Loops are vertically offset for clarity.

of Tb. The terbium thickness in our study does not exceed 6 Å (2 ML) to avoid interdiffusion.[31] The films are covered by 40 Å Ru to prevent them from oxidation and to allow *ex situ* magnetic experiments. A Co thickness of 16 Å was chosen, because in previous studies on Co/Ru sandwiches prepared under the same conditions[31,32] it was found that the effective magnetic anisotropy decreases almost linearly with Co thickness and yields an out-of-plane magnetization below 15 Å. For Ru/Co (16 Å)/Ru the effective anisotropy is nearly equal to zero. Therefore a 16-Å-thick Co layer is a good candidate to observe a temperature-induced spin reorientation. We measure the ac susceptibility and the magnetization between 80 and 400 K by ac- and dc-MOKE in polar geometry, which is sensitive to the out-of-plane component of the magnetization only. Kerr loops are recorded in longitudinal geometry also to detect the in-plane component of the magnetization.

Figure 1 shows polar hysteresis loops obtained for different terbium depositions at 185 and 295 K. At room temperature all films except the 6 Å Tb sample (not shown here) have a remanent magnetization perpendicular to the film plane, which is not fully saturated. This can be understood if the magnetization is tilted out of the film plane. It means that the macroscopic remanent magnetization $M_r(T,d_{\mathrm{Tb}})$ lies on a cone making an angle $\theta_{\mathrm{eq}}(T,d_{\mathrm{Tb}})$ with the normal of the film such that $\cos\theta_{\mathrm{eq}}(T,d_{\mathrm{Tb}})=M_r(T,d_{\mathrm{Tb}})/M_s(T,d_{\mathrm{Tb}})$ if one considers a single-domain state only and neglects a small in-plane magnetic anisotropy. $M_s(T,d_{\mathrm{Tb}})$ is the saturation magnetization of the film, T is the temperature, and d_{Tb} is the Tb thickness. A tilted orientation of the magnetization is only obtained if one includes a fourth-order anisotropy term $K_4(T,d_{\mathrm{Tb}})$ in the free energy $E(T,d_{\mathrm{Tb}})$ of the Co(0001) hcp system:[23,24]

$$E(T,d_{\mathrm{Tb}})=\{K_2(T,d_{\mathrm{Tb}})-2\pi M_s^2(T,d_{\mathrm{Tb}})\}\sin^2\theta + K_4(T,d_{\mathrm{Tb}})\sin^4\theta,$$

θ is the angle between the normal of the film and the magnetization. The anisotropy coefficients must verify $2K_4(T,d_{\mathrm{Tb}})>-[K_2(T,d_{\mathrm{Tb}})-2\pi M_s^2(T,d_{\mathrm{Tb}})]$ for a tilted orientation of the magnetization. Both coefficients contain interface and volume contributions as discussed, for example, by Fritzsche *et al.*[22] A quantitative analysis of the anisotropy coefficients as a function of temperature and Tb thickness will be published elsewhere.

The slight increase of the remanence after 1 Å terbium deposition at 295 K (Fig. 1) indicates an increase of the perpendicular anisotropy. From 1 to 6 Å terbium the easy axis of magnetization rotates continuously toward the film plane which corresponds to a decrease of the perpendicular anisotropy. These effects can be explained by differences in nearest-neighbor coordination at the interface which changes the interface anisotropy. Indeed, x-ray magnetic circular dichroism experiments[34] showed no long-range magnetic order of the terbium for all films, which means that the dipolar anisotropy energy $2\pi M_s^2(T,d_{\mathrm{Tb}})$ remains constant for all films at a fixed temperature.

At 185 K (Fig. 1) all films present a square hysteresis loop with full remanence. For these films the easy axis of magnetization is perpendicular to the film plane, and to very good approximation the film is single domain. The origin of the perpendicular orientation is a strong magnetocrystalline anisotropy which overcomes the dipolar one. In our notation, the magnetocrystalline anisotropy includes the surface/interface, bulk, and magnetoelastic anisotropies due to spin-orbit interaction but not the magnetostatic terms. To get further insight into the temperature dependence of the orientation of the magnetization we have measured the remanence and the coercive field in polar geometry for each film in the temperature range from 80 to 400 K.

Data for the Tb(2 Å)/Co(16 Å) film are presented in Fig. 2(b). We clearly find three characteristic regions for the orientation of the magnetization. The first one (region I) goes from 80 to about 210 K: the hysteresis loop is square with full remanence. Region II extends from about 210 to 311 K where the perpendicular component of the remanence $M_R^{\perp}$ decreases gradually to approach zero. Above 311 K the polar remanence remains zero (region III). In region I the perpendicular anisotropy dominates and a quasi-single-domain magnetization perpendicular to the film plane persists. In region II the polar remanence decreases gradually in an interval of about 100 K. The remanent magnetization turns continuously from out-of-plane toward in-plane. At the same time the coercive field decreases to zero at 311 K [Fig. 2(b)], which indicates a smaller threshold for thermal activation of domain-wall nucleation and movement. The rotation of the easy axis (called spin-reorientation transition) can be explained by the competition between the shape $[-2\pi M_s^2(T,d_{\mathrm{Tb}})\sin^2\theta]$ and magnetocrystalline $[K_2(T,d_{\mathrm{Tb}})\sin^2\theta+K_4(T,d_{\mathrm{Tb}})\sin^4\theta]$ anisotropy energies, which result from long-range dipolar and short-range spin-orbit interaction, respectively. K_2 and K_4 vary differently as a function of the temperature and the direction of the magnetization can change with temperature. Below 210 K the magnetocrystalline anisotropy which favors perpendicular anisotropy dominates: that is, $K_2(T,d_{\mathrm{Tb}})>2\pi M_s^2(T,d_{\mathrm{Tb}})$ and $K_2(T,d_{\mathrm{Tb}})-2\pi M_s^2(T,d_{\mathrm{Tb}})+2K_4(T,d_{\mathrm{Tb}})>0$. From 210 K up to $T_R=311$ K the difference between the magnetocrystalline and the shape anisotropies decreases and the total

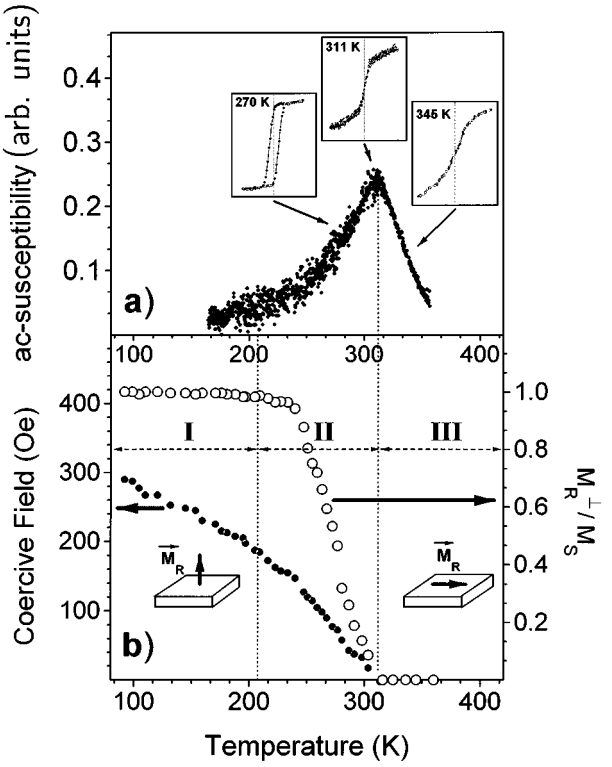

FIG. 2. (a) Remanence normalized with respect to the saturation magnetization $M_R^{\perp}/M_S$ (○) and coercive field (●) in polar geometry versus temperature for the 16 Å Co/2 Å Tb film. I, II, III represent the three regions described in the text. (b) Susceptibility recorded by ac-MOKE as a function of temperature for the 16 Å Co/2 Å Tb sample. The ac field of $f=180$ Hz and 10 Oe amplitude is applied perpendicular to the film plane. Note that the susceptibility maximum coincides with the disappearance of the polar remanence $M_R^{\perp}$ at the transition temperature $T_R=311$ K. Three polar hysteresis loops at 270, 311, and 345 K are displayed in the insets. The x-axis and y-axis lengths are 1.5 kOe and 0.4 mrad for each inset.

anisotropy of the film becomes small. It corresponds to $0<2\pi M_s^2(T,d_{\mathrm{Tb}})-K_2(T,d_{\mathrm{Tb}})<2K_4(T,d_{\mathrm{Tb}})$: θ_{eq} is given by $2K_4(T,d_{\mathrm{Tb}})\sin^2\theta_{\mathrm{eq}}=-K_2(T,d_{\mathrm{Tb}})-2\pi M_s^2(T,d_{\mathrm{Tb}})$. Above T_R the shape anisotropy overcomes the magnetocrystalline contribution and the magnetization lies in the film plane: $K_2(T,d_{\mathrm{Tb}})-2\pi M_s^2(T,d_{\mathrm{Tb}})+2K_4(T,d_{\mathrm{Tb}})<0$. By convention we define the reorientation temperature T_R as the temperature above which the perpendicular remanence vanishes and the easy axis is in-plane.

The spin-reorientation transition described above is confirmed with polar ac-MOKE. This geometry is sensitive to the out-of-plane component of the magnetization, which is the relevant parameter for the reorientation from out-of-plane to in-plane. Figure 2(a) shows the result for the 2 Å Tb film. The ac signal is proportional to the susceptibility of the film.[26–29] For a better understanding of this signal we show three hysteresis loops at 270, 311, and 345 K [insets in Fig. 2(a)]. From 180 to 311 K the susceptibility gradually increases. The nucleation field for magnetic domain reversal which roughly corresponds to the coercive field (inset at 270 K) is larger than the ac magnetic field (10 Oe). Approaching the transition temperature the coercivity decreases and the

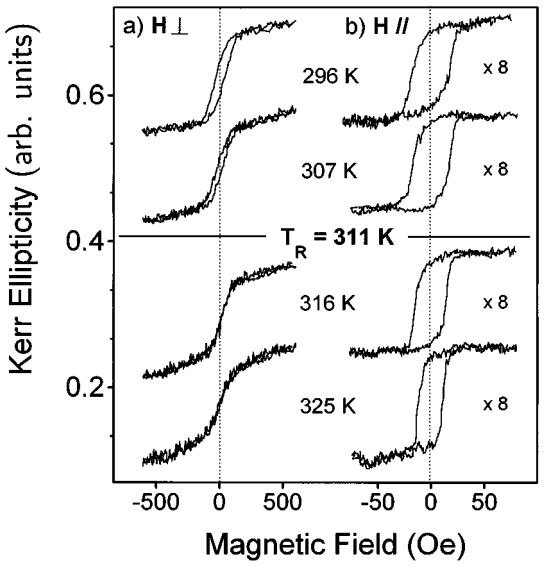

FIG. 3. Hysteresis loops of the 16 Å Co/2 Å Tb film for (a) polar and (b) longitudinal geometries above and below $T_R=311$ K. Loops are vertically offset for clarity. Note the gain for the longitudinal loops in (b).

slope of the minor magnetization loop[28] increases. The maximum of the susceptibility at T_R indicates a maximum of the slope of the magnetic loop (inset at 311 K). Above T_R the monotonous decrease of the ac signal results from the decrease of this slope: a larger field is necessary to rotate the magnetization out of the film plane for higher temperature (inset at 345 K). Our dc- and ac-MOKE measurements yield the same transition temperature. It shows that ac-MOKE is well suitable to measure the reorientation temperature. It is important to note that T_R is not the Curie temperature T_C of the film. From earlier experiments[35,36] one knows that T_C of 8 ML Co is far above 500 K. In addition, the finite in-plane remanence above T_R rules out this possibility as discussed below.

We have performed measurements in longitudinal geometry in the vicinity of T_R. Figure 3(b) shows the results for the Tb(2 Å)/Co(16 Å) sample. It is worthwhile to point out that in our longitudinal setup the magnetic field is oriented in the film plane with an accuracy better than 0.25°. The shape of the longitudinal loop does not vary when we cross the transition temperature and the amplitude of the longitudinal signal is an order of magnitude smaller than the polar one. Consequently the longitudinal hysteresis loops below T_R do not contain any polar contribution. We observe a nonvanishing in-plane remanent magnetization below and above T_R. Our results clearly show that there is no temperature range in the vicinity of the transition temperature where the remanent magnetization vanishes in both directions. *The rotation of the easy axis of magnetization as a function of temperature occurs without loss of long-range order*. The presence of both components of the remanent magnetization at the transition can again be explained by a continuous rotation of the macroscopic remanent magnetization. Allenspach *et al.*[8] observed such a rotation for ultrathin Co/Au(111) films as a function of Co thickness by spin-polarized scanning electron microscopy. The loss of long-range order is usually ex-

TABLE I. The transition temperatures of the 16 Å Co/X Å Tb ($X=0\rightarrow6$) films. The values are determined with an accuracy of ±5 K.

Terbium thickness (Å)	0	1	2	3	4	5	6
Transition temperature (K)	320	323	311	305	300	297	277

plained by the compensation of the dipolar anisotropy and the magnetocrystalline anisotropy yielding effectively an isotropic system. For ultrathin films, this state can be considered as a 2D isotropic Heisenberg system.[2] However, this argument fails if the long-range order breaks down. This is to say, if the spontaneous magnetization M_{sp} is zero, the shape anisotropy $2\pi M_{sp}^2$ (Ref. 2) is zero. Consequently the compensation is not present anymore. Based on that simple argument one can argue that a loss of long-range order cannot be achieved experimentally by choosing a thickness where long-range dipolar shape anisotropy cancels short-range spin-orbit (magnetocrystalline) terms.

In regard to previous results where 2D behavior is found only below 5 ML,[36] we expect that 8 ML Co act like a 3D system and consequently long-range order is present at any temperature below T_C, even in the absence of anisotropy. For simplicity we have considered a single-domain magnetization rotation only. However, the presence of a statistical distribution of in-plane and out-of-plane multidomain states, which cannot be ruled out, would not change our interpretation. Such a multidomain state, where long-range order is present in each domain, has been observed in the Fe/Cu(100) system.[11]

Finally, we show in Table I that the transition temperature varies as a function of the terbium thickness by almost 50 K. T_R first increases after 1 Å terbium deposition and then decreases with additional terbium. The variations of T_R can be explained by a change of the interface anisotropy as described before.

In conclusion, we have studied the spin-reorientation phase transition as a function of temperature for X ($X=0\rightarrow6$) Å Tb/16 Å Co films. A reversible rotation of the remanent magnetization from out-of-plane to in-plane is observed with increasing temperature. The switching of the easy axis occurs continuously without loss of long-range order, that is to say that a remanent magnetization is always present. To our knowledge it is the first time that a reorientation of the magnetization as a function of temperature has been observed in the cobalt-based films far below the Curie temperature and that the transition temperature varies with the cover (Tb) layer thickness from 323 K (for 1 Å Tb) to 277 K (6 Å Tb). The system may offer interesting possibilities for applications which need a tunable switching temperature of the magnetization near room temperature. Also, the new ac-MOKE technique was used in polar geometry to precisely determine the transition temperature.

G.G. would like to thank the Institut für Experimentalphysik for its hospitality. Helpful discussions with K. Baberschke and J. P. Kappler are acknowledged. This work was partly supported by the DRET under Grant No. 92 1449 and through HCM-EC Contract No. ERBCHRXCT 940473.

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