Evidence for Noncollinearity between Surface and Bulk Magnetization in Ultrathin Co Films

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The magnetization reversal of ultrathin Co films on Cu(001) has been investigated by grazing ion scattering and magneto-optical Kerr effect. Differences in the behavior of surface and bulk magnetization are found and attributed to the reduced coordination and site symmetry at the surface. The reversal behavior of the surface magnetization depends on the chemical surface composition. For pure Co films, the reversal of the bulk magnetization is preceded by a complete reversal of the surface magnetization. A particular magnetic state of the surface is suggested as a precursor for magnetization reversal.

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Magnetization reversal processes in ultrathin films are of fundamental importance for both basic research and technological applications. In magnetic memory devices, magnetization reversal has to take place in times of typically nanoseconds in order to improve currently achieved data rates. Modern techniques include spin dynamics on short time scales and current-induced switching. Depending on the geometry of the magnetic structure and the method of inducing magnetization reversal, different reversal processes dominate at different time scales [1]. The reversal processes are explained by precessional motion of magnetization for ultrashort field pulsing [2] and by spin transfer for injection of spin-polarized currents [3]. Domain wall motion and propagation are the dominant mechanisms for magnetization reversal where an external field is applied antiparallel to the direction of the average magnetization. Because the magnetic field is usually changed on a relatively long time scale, a ''static'' hysteresis loop is obtained in the latter case.

The microscopic mechanisms for magnetization reversal are still insufficiently explored. The role of reduced site coordination and symmetry at the surface, at steps, or at islands is a subject of current interest [4]. It is one of the key elements for the precise control of the domain structure and the magnetization direction.

In this Letter, we report on differences in surface and bulk magnetization in ultrathin Co films and its implications for magnetization reversal processes which are unexpected in view of common properties of the bulk. Our findings are mainly based on the capture of spin-polarized electrons into excited atomic states of fast He atoms during scattering from the surface [5,6]. For grazing scattering, trajectories of projectiles and final electron capture are localized in front of the surface (''surface channeling''). The pronounced sensitivity to the magnetic ordering of the topmost layer [7] allows one to record defined hysteresis loops of the surface magnetization.

The experiments were performed in an ultrahigh vacuum setup attached via differential pumping stages to the beam line of a small electrostatic ion accelerator. Co films were grown by molecular beam epitaxy (MBE) at a rate of 0.3 monolayers (ML)/min. Well collimated beams of 25 keV atoms were directed on a clean Cu(001) crystal at a grazing angle of incidence of about 1.6°. Protons were used for ion-induced Auger electron spectroscopy (AES). The experiments were performed close to a $\langle 110 \rangle$ direction which corresponds to an in-plane easy axis of magnetization for Co films on Cu(001). Hysteresis loops of the bulk of the films were obtained making use of the magneto-optical Kerr effect (MOKE).

For the scattering experiments on electron capture into excited levels of He atoms, the emitted polarized light of the $2s^3S - 3p^3P$ transition at $\lambda = 388.9$ nm is detected through a quartz window by means of a quarter-wave retarder plate, a narrow bandwidth interference filter, a linear polarizer, and a photomultiplier. Concepts and analysis of experiments on light emission after electron capture (EC) are described in detail in Refs. [5,6]. In brief, the spin polarization $P_s = \langle S_z \rangle / S$ of captured electrons can be deduced from the circular polarization of the fluorescence light described by the Stokes parameter $S/I = [I(\sigma^-) - I(\sigma^+)]/[I(\sigma^-) + I(\sigma^+)]$, where $I(\sigma^-)$ and $I(\sigma^+)$ are the intensities of light with negative and positive helicity (σ^- and σ^+) [8]. P_s is obtained from measurements of S/I with reversed settings of the magnetization. It is related to the long-range magnetic order (magnetization M) at the sample surface, although a quantitative relation has not been established so far.

Hysteresis loops recorded by MOKE are shown on the left side of Fig. 1 for 5 ML Co deposited at different temperatures T_{GR}. Square loops with a sudden jump close to the coercive field H_C^B (B: bulk) are observed as expected for magnetization reversal governed by domain wall motion [9]. H_C^B is enhanced if the measurement is performed at $T = 140$ K [Fig. 1(a)] instead of $T = 300$ K [Fig. 1(d)] which is likely to originate from an increase in magnetic anisotropy. In comparison with MOKE, hysteresis loops recorded by EC reveal significant differences. For $T_{\mathrm{GR}} = 140$ K [Fig. 1(b) and 1(c)], the loop shape remains the same but the coercivity decreases by 27% from $H_C^B \approx 18.5$ Oe to $H_C^S \approx 13.5$ Oe (S: surface). For $T_{\mathrm{GR}} = 300$ K [Fig. 1(e) and 1(f)], reversal starts with a sudden

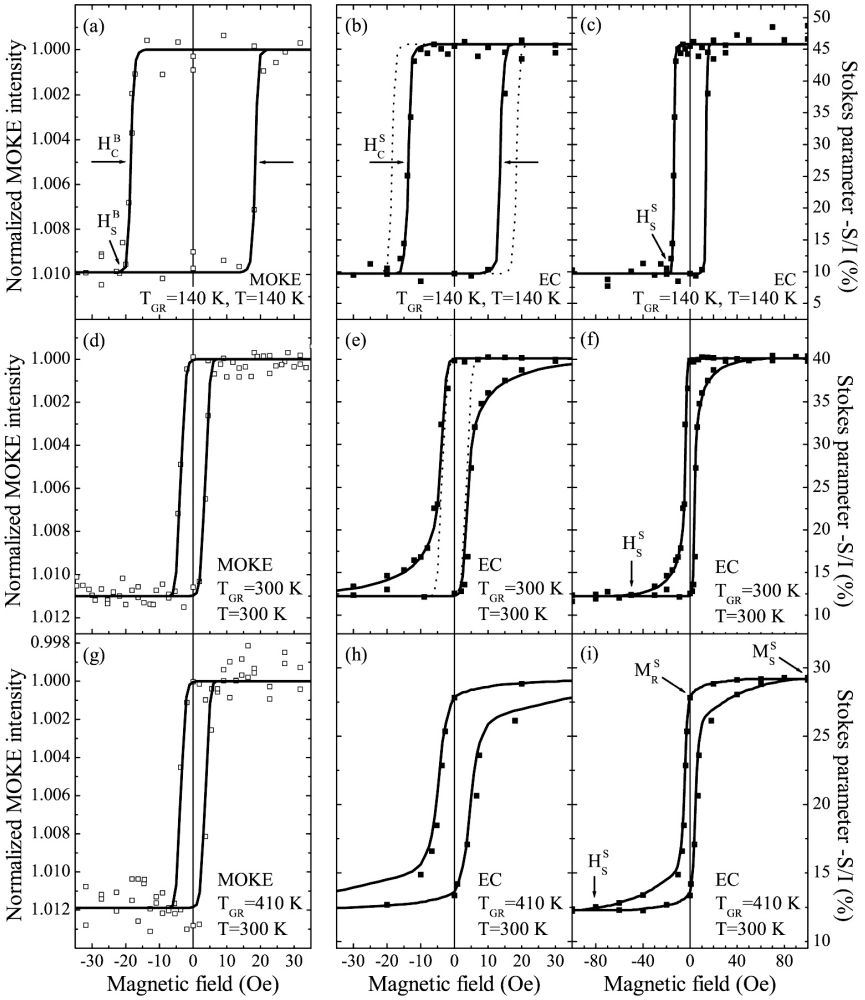

FIG. 1. Hysteresis loops for 5 ML Co obtained from MOKE (left panel) and EC (middle and right panel) for different growth temperatures: (a)–(c) $T_{\mathrm{GR}} = 140$ K, (d)–(f) $T_{\mathrm{GR}} = 300$ K, and (g)–(i) $T_{\mathrm{GR}} = 410$ K. Curves in right panel show extended field range compared to curves in middle panel. Data recorded at different sample temperatures T. Solid curves are guides to eyes; dotted curves in (b) and (e) correspond to MOKE in (a) and (d), respectively. The scales of the ordinate are chosen arbitrarily.

jump of the magnetization but saturation is only achieved at a field $H_S^S \approx 50$ Oe which is much higher than $H_C^S \approx H_C^B \approx 4$ Oe.

The differences in the data obtained by EC and MOKE are attributed to the probing depths. MOKE provides information on the entire film while EC is sensitive to the surface only. From the data shown in Fig. 1, we conclude a different behavior of surface and bulk magnetization during reversal. Additionally, characteristic features of the hysteresis loop such as H_S^S change with varying T_{GR} for the surface but not for the bulk of the film.

For Co films grown at 140 K, the reversal of the bulk magnetization [Fig. 1(a) and dotted curve in Fig. 1(b)] is preceded by the reversal of the surface magnetization [Fig. 1(b)]. A rectangular loop indicates that a relatively strong lateral exchange interaction makes a coherent motion of ensembles of spins in the surface favorable. In a small field range, a magnetic state exists where the surface magnetization differs from the bulk magnetization. For $|H_C^S| \approx |H| < |H_C^B|$, the surface magnetization is most likely switched with respect to the bulk magnetization, either by 90° or 180° in the plane due to the fourfold magnetic anisotropy of fcc(001) Co films, or by 90° out of the plane due to an increased anisotropy at the surface.

A noncollinear orientation of surface and bulk magnetization within a distance of a few lattice spacings has also been suggested for the magnetization reversal of a Fe(001) single crystal [10]. On the basis of depth profiling with spin polarization of secondary electrons, it was concluded that the surface magnetization is perpendicular to the bulk magnetization during reversal. Weak exchange coupling on a path perpendicular to the surface [11] has been assumed to be the origin of differences in surface and bulk magnetization on a ns-μs time scale investigated by the spin-polarization in time resolved photoemission [12].

While the reversal for $T_{\mathrm{GR}} = 140$ K seems to proceed by a coherent motion of ensembles of surface spins [Fig. 1(b) and 1(c)], the reversal for $T_{\mathrm{GR}} = 300$ K does not reveal a uniform behavior of the entire surface magnetization [Fig. 1(e) and 1(f)]. A part reverses by switching

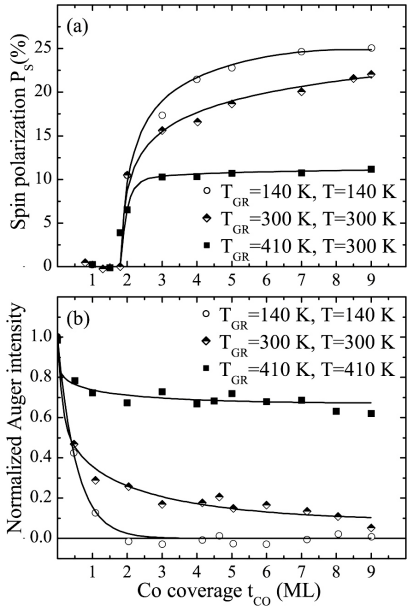

FIG. 2. (a) Remanent spin polarization P_s from EC and (b) proton-induced Cu-*MNN* Auger signal with increasing Co coverage for different T_{GR}. Data recorded at different sample temperatures T. Solid lines are guides to the eyes.

which is indicated by the sharp drop at $H_C^S \approx 4$ Oe; however, a noticeable part requires higher fields for a complete alignment ($H_S^S \approx 50$ Oe). The remaining unswitched hard magnetic entities (for $|H_C^S| < |H| < |H_S^S|$) are attributed to Cu impurities in the surface which lead to local changes in magnetic anisotropy. A low impurity concentration C_{Cu}^S causes noncollinearity between the bulk magnetization and parts of the surface spins but has no significant influence on the reversal of the bulk magnetization [Fig. 1(d)].

Detailed information on the degree of contamination is obtained from the data presented in Fig. 2. For a Co thickness up to 9 ML, the remanent spin polarization P_s for $T_{\mathrm{GR}} = 300$ K is decreased by 10%–20% compared to $T_{\mathrm{GR}} = 140$ K. The reduction can be attributed to an appreciable amount of Cu in the film surface which decreases with increasing Co thickness. The observation of Cu segregation for $T_{\mathrm{GR}} = 300$ K is supported by proton-induced AES which provides information on the chemical composition of the topmost layer [13,14]. For $T_{\mathrm{GR}} = 140$ K, the Auger signal of the Cu-*MNN* transition at 60 eV vanishes at 2 ML Co [Fig. 2(b)] which shows that Cu segregation is suppressed at low temperatures. The AES data for $T_{\mathrm{GR}} = 300$ K imply $C_{\mathrm{Cu}}^S \approx 15\%$–$20\%$ for 5 ML Co [14]. It can therefore be concluded that the difference in P_s observed for $T_{\mathrm{GR}} = 140$ K and $T_{\mathrm{GR}} = 300$ K is mainly caused by Cu incorporated in the Co film surface during deposition at higher temperature.

For $T_{\mathrm{GR}} = 410$ K, a high concentration $C_{\mathrm{Cu}}^S \approx 50\%$–$70\%$ persists in the surface which is evidenced by both the AES and P_s data. A detailed analysis reveals that two layers of Cu intermixed with Co float on top of a Co film [14]. Under this condition, the remanent magnetization of the surface deviates from the saturation magnetization with $M_R^S \approx 0.85 M_S^S$ [Fig. 1(i)] due to anisotropies different from the fourfold symmetry of pure fcc Co(001) films. These local anisotropies are induced by chemical or structural disorder and suppress a long-ranged exchange coupling which would favor a uniform alignment of the whole surface magnetization. H_S^S reaches about 80 Oe.

For both low and high Cu impurity concentrations in the surface of Co, a part of the surface spins reverses after the bulk magnetization has completely switched. This supports the assumption that exchange interaction is weakened on a path perpendicular to the surface [11,12]. The latter seems to hold especially for pure Co films [Fig. 1(a)–1(c)], but it should be noted that the real reason of the weakened exchange may be structural rather than chemical properties.

With respect to this, it is obvious to consider the characteristic oscillatory behavior in both structural and magnetic properties of Co films on Cu(001). One monolayer periodic oscillations have been observed in the surface in-plane lattice spacing [15], the step-induced magnetic anisotropy [16], and the surface magnetization [17]. The occurrence of lattice oscillations has been attributed to free edges of Co islands which are distorted owing to reduced coordination at half-integer coverage [18]. It is tempting to relate these oscillations to changes in magnetic properties such as exchange interaction or anisotropy but we do not observe any difference in reversal behavior which could be assigned to half- or full-integer coverage.

Anisotropy effects similar to the step-induced anisotropy oscillations [16], which have been speculated to originate from the oscillatory variation of film roughness or, alternatively, from oscillations in surface in-plane lattice spacing, are negligible because they are only weakly reflected in the coercivity H_C. The amplitude of the H_C oscillations amounts to only 1% [16] which is more than 1 order of magnitude smaller than the difference observed in H_C^B and H_C^S by MOKE and EC (Fig. 1).

Even if no direct relation to the oscillatory behavior in the Co/Cu(001) system can be established, the structure and morphology may still be decisive for the noncollinearity. Because the coercivity is determined by several physical mechanisms such as domain wall motion and pinning, its relation to structural irregularities is not straightforward. The reduced site symmetry at local defects or terrace steps can induce additional local anisotropies with inhomogeneous distribution of directions which destabilize the surface magnetization and influence the motion of domain walls [4] for both half- and full-integer coverage. Model calculations are very difficult owing to the intimate connection between surface morphology and reversal which has been insufficiently explored [4].

In Fig. 3, we suggest a simple microscopic mechanism for the magnetization reversal. It is based on a particular magnetic state of the surface at $H \approx H_C^S$ which provides a precursor [Fig. 3(b)] for nucleation of reversed domains in the bulk of the film [Fig. 3(c)]. The precursor state is

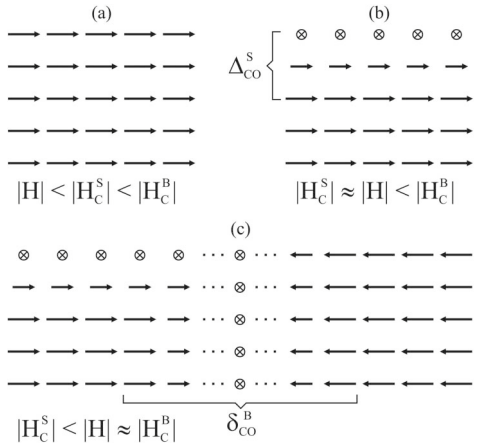

FIG. 3. Microscopic model for magnetization reversal in ultrathin Co films with increasing field H: (a) uniform film magnetization, (b) precursor state close to surface with width Δ^S_{Co}, and (c) creation of domain wall in bulk of film with width δ^B_{Co}. Arrows represent projection of spins on plane of view; crosses correspond to perpendicular orientation. Representative spin configuration of a few lattice sites is shown in side view.

assumed to exist in ensembles of surface spins which are sufficiently large to allow for strong lateral exchange interaction. The switching field of the bulk magnetization H^B_C is usually determined either by pinning of domain walls during their propagation or by the nucleation of domains with different magnetization directions.

In general, exchange interaction in ferromagnetic 3D transition metals suppresses a noncollinear spin orientation within a short distance. In the bulk, the minimum length scale over which the direction of spins can vary appreciably is given by the wall width parameter (exchange length) $\Delta = (A/K)^{1/2}$, where A is the exchange stiffness and K the magnetic anisotropy [19]. Following micromagnetic theory, the width of a 180° wall separating two oppositely magnetized domains in a uniaxial crystal can be calculated by $\delta^B_{180} = \pi(A^B/K^B)^{1/2}$. Using typical properties of Co films ($A^B = 1.3 \times 10^{-6}$ erg/cm [9] and $K^B = 2.3 \times 10^6$ erg/cm^3 [20]), Δ and δ amount to $\Delta^B_{Co} = 75$ Å and $\delta^B_{Co} = 235$ Å, respectively. Thus, the noncollinearity in the vicinity of the surface of a 9 Å Co film (within a distance $\Delta^S_{Co} \leq 9$ Å $\ll \Delta^B_{Co}$) must originate from changes in exchange interaction or magnetic anisotropy due to reduced site symmetry and coordination. For 3D transition metal films, such a behavior has, to the best of our knowledge, not been reported so far under static conditions whereas it has been suggested for dynamic magnetization reversal on a ns-μs time scale [12].

Under static conditions, noncollinearity of spins has been suggested to occur at interfaces, especially in trilayers such as Fe/M/Fe with M = Au, Al, Cu, Cr [21]. It has been explained by a model relying on "loose interfacial spins." These spins possess a weakened exchange interaction and introduce a biquadratic coupling which leads to an orthogonal alignment of spins at the interface. The special role of the interface in bi- and trilayers resembles the behavior of the surface in the Co films.

In summary, we have found differences in surface and bulk magnetization during magnetization reversal in thin Co films. For pure films, a simple microscopic model featuring a precursor state which extends over distances of only one or two atomic sites perpendicular to the surface has been suggested. The experimental finding of noncollinearity between surface and bulk spins over distances far below the exchange length of the bulk is unexpected.

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