Antiferromagnetic hysteresis in magnetoresistive multilayers investigated by x-ray resonant scattering

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We have used resonant scattering of polarized soft x rays as a direct probe of the magnetic order in a weakly coupled Co/Cu multilayer. Our field dependent results, combined with *in situ* resistance measurements, show a direct correlation between magnetoresistance and antiparallel magnetic ordering in reversible and irreversible processes. © *2002 American Institute of Physics.*
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The giant magnetoresistance (MR) observed in magnetic multilayers and spin valves strongly depends on the magnetization process and on the details of the magnetic domain structure. The broad interest in these materials for applications emphasizes the need for developing tools able to monitor the magnetic order in the presence and as a function of an external magnetic field. The Co/Cu multilayer system we have chosen to study is often considered to be a prototype for giant MR.[1] In the antiferromagnetic exchange coupling regime (Cu thickness ~ 9 Å), low and high resistance states are associated with, respectively, parallel and antiparallel alignment of the Co magnetization in adjacent layers. With increasing Cu thickness, the weak coupling (20–40 Å) and uncoupled (>50 Å) regimes are progressively reached, in which Co/Cu is known to exhibit a rather complex magnetic behavior, especially in terms of magnetic field cycling.[2–4] For convenience, throughout this letter we will refer to ferromagnetic (FM) and antiferromagnetic (AF) ordering of Co layers, although we do not necessarily imply true AF exchange.

AF order is difficult to investigate for the obvious reason that there is no macroscopic residual moment. The most direct way of observing AF order is through structural sensitive techniques such as diffraction, providing it is possible to distinguish between sites with opposite magnetizations. One such technique is polarized neutron reflectivity (PNR),[5] another is x-ray resonant magnetic scattering (XRMS). At photon energies corresponding to excitations to magnetic states, e.g., Co $2p \rightarrow 3d$ transitions, magnetic order strongly affects the optical constants, thus defining a new order parameter that can differ from the chemical one.[6] Moreover, the $2p \rightarrow 3d$ resonances in $3d$-transition metals correspond to a wavelength range (15–30 Å) that matches the order parameters of MR multilayers.

In this letter, we report on the possibility of using XRMS to measure the field dependence of the AF order in a metallic multilayer directly. We also show that it is possible to correlate the XRMS data to a concomitant change in resistivity. Our measurements also point the way to using XRMS for dynamic studies of magnetization processes in complex heterostructures such as magnetic multilayers and spin valves.

A $(\text{Co } 11.2 \text{ Å}/\text{Cu } 22.8 \text{ Å})_{20}$ multilayer was sputterdeposited on a Si(111) wafer.[7] Several samples were cut from the same wafer in order to perform a series of experiments always starting from the same as-prepared material. X-ray diffraction analysis (Cu $K\alpha$ radiation) gave an error bar of ± 0.2 Å on layer thicknesses and an interface roughness of 3 Å root-mean square. Magnetization and resistance (four-point probe) versus applied field were first measured *ex situ*. An MR of about 7% was observed in an applied field of ≈ 22 Oe. Alternating gradient field magnetometry measurements gave a saturation magnetic moment of approximately 1.5 μ_B per Co atom at 2 kOe, a remanence of 46%, and a coercive field of 23 Oe.

XRMS experiments were performed at beamline 6.3.2 at the Advanced Light Source (Berkeley) using its reflectometer endstation.[8] We selected out-of-orbit–plane emission from the bending magnet source in order to obtain elliptically polarized radiation ($\approx 60\%$ right-handed circular polarization). Resolving power was ≈ 2000 at the Co L edges. During XRMS measurements, the sample resistance was recorded *in situ* using a two-point technique. An external field (H) of up to 1 kOe was applied along the sample surface and perpendicular to the scattering plane. It is important to stress that, using circularly polarized light, the XRMS experiment is sensitive to the projection of the magnetization in the scattering plane.[9] Therefore, in the geometrical setup used here, it is the magnetic order in the direction perpendicular to the applied field which will be measured.

Figure 1(a) shows the result of a specular reflectivity scan ($\theta/2\theta$ mode) performed at a photon energy of 776.5 eV,

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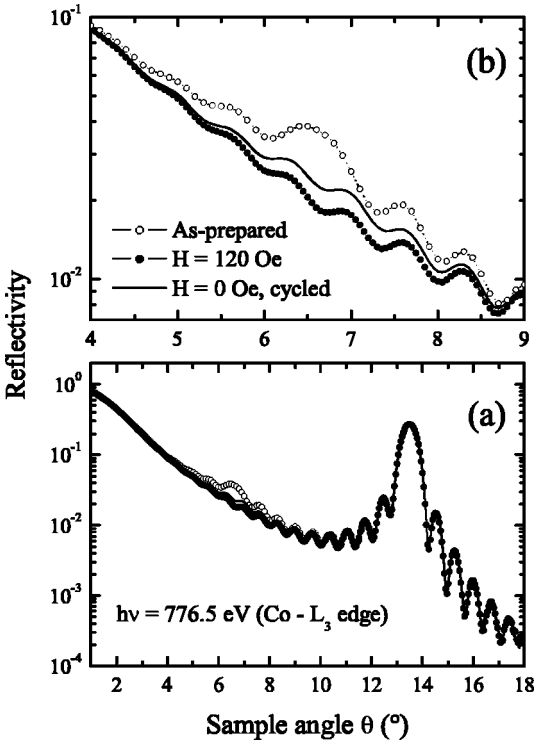

FIG. 1. (a) Specularly scattered intensity as a function of θ for the as-grown sample (open circles), for an applied field of 120 Oe (solid circles) and under remanent conditions after a magnetic cycle (full line). (b) Enlarged view of the antiferromagnetic Bragg peak region. Photon energy is $\hbar\omega = 776.5$ eV.

i.e., at the L_3 edge of Co. The strong Bragg peak at $\theta = 13.5°$ corresponds to the order parameter of the Co/Cu stacking ($2d = 68$ Å). The sharp oscillations (Kiessig fringes) visible on both sides of the Bragg peak correspond to the interference between waves scattered at the vacuum/multilayer and multilayer/substrate interfaces. The AF Bragg peak, corresponding to the order parameter imposed by the opposite sign of the magnetization in adjacent Co layers ($2d = 136$ Å), is located at $\approx 6.75°$. This angular region is enlarged in Fig. 1(b). The three curves in Fig. 1 correspond to measurements performed on the as-prepared sample (open circles), then with a field of 120 Oe applied normal to the scattering plane (filled circles), and finally at zero field after a complete magnetic cycle (full line). Our first comment is that the zero-field intensity of the AF peak of the as-prepared state is strongly reduced after the sample has been forced into its FM state by an external field equal or greater than 100 Oe.

The top panel of Fig. 2 shows the reflected intensity ($\hbar\omega = 776.5$ eV and $\theta = 6.75°$) as a function of the applied field, starting from an as-prepared sample. The MR, simultaneously recorded *in situ* and normalized to the resistance at 1 kOe, is displayed in the bottom panel of Fig. 2. The general line shapes and peak positions coincide in the MR curve and in the scattered intensity at the AF peak. The drop in remanent intensity after the field has been cycled indicates that the ordered AF stacking of Co layers across the entire sample is to a large extent lost. Our combined MR and XRMS results confirm reports by other authors[1,5,10] that antiparallel ordering and remanent sample resistivity are smaller after magnetic cycling than in the as-prepared state.

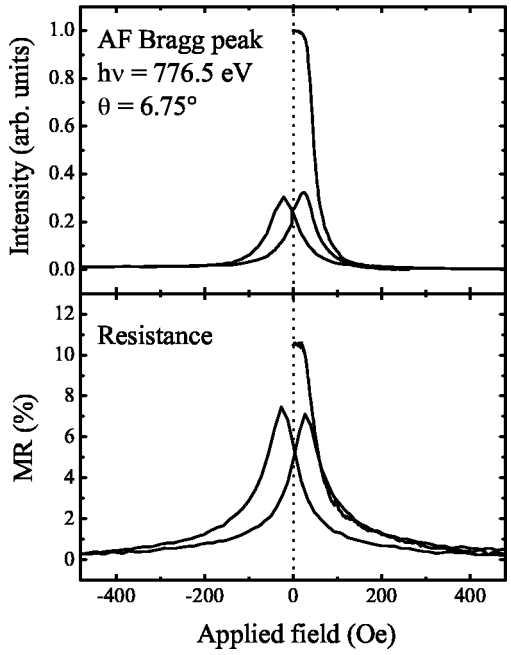

FIG. 2. MR (bottom) and scattered intensity (top) over a field loop of 500 Oe.

Finally, we show that the initial value of the zero-field resistance can be partially restored using a demagnetization-like procedure. Figure 3(a) shows the time dependence of the applied field: Starting from $H = 360$ Oe, the field is made to oscillate between positive and negative values with 1 Hz frequency and its amplitude is reduced progressively to zero ($t = 250$ s, vertical dashed line). Two further complete cycles follow, with an amplitude of 360 Oe and at a lower frequency. The results of simultaneous measurements of the

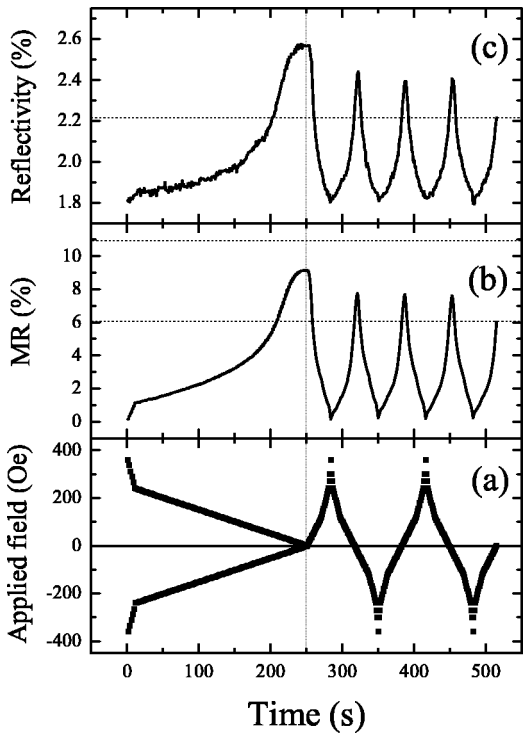

FIG. 3. (a) Time dependence of the applied magnetic field, and simultaneous acquisition of (b) sample resistance and (c) reflectivity ($\theta = 6.75°$ and $\hbar\omega = 776.5$ eV).

sample resistance and of the scattered intensity at the AF peak are shown in Figs. 3(b) and 3(c), respectively. The two horizontal dashed lines in Fig. 3(b) represent the resistance of the as-prepared sample (10.5%, top line) and its zero-field value after a magnetic cycle of 360 Oe (6%, bottom line). As the field amplitude decreases during the demagnetization cycle (0–250 s), both MR and scattered intensity increase, reaching their static remanent values for an oscillating field of about 40 Oe (205–210 s). Reducing the width of the field oscillations to zero, leads to a further increase of MR up to 9%, accompanied by an increase in the scattered intensity. This result confirms that the magnetic and electrical behavior of the sample are intimately related and strongly depend on its history. The exact magnetic state and sample resistivity in zero-field depend, for instance, on how the zero-field condition has been reached: Not only is the as-grown sample different from a cycled one, but also the magnetic state of the latter depends on the frequency used for cycling the field. These observations also illustrate how the high sensitivity of XRMS may be used to investigate the important issue of MR dynamics. Access to very rapid phenomena was beyond the scope of this work, but can be reached in principle by simply using a faster detection system.

To summarize, we have shown that resonant x-ray scattering may be used to directly probe the magnetic state of multilayers and to correlate it to their magnetoresistive properties. In the Co/Cu system investigated here, we found that both resistivity and AF ordering are at their maximum in the as-prepared state. Once a field has been applied that forces a parallel alignment between the layers, both MR and the AF intensity are irreversibly reduced. In our sample, maximum MR goes from 10%–11% in the virgin sample to 7% at 22 Oe after cycling. A demagnetization-like procedure results in a partial recovery of both MR and antiparallel magnetic ordering of adjacent Co layers.

XRMS unambiguously ties MR to AF ordering whether reversible and irreversible processes occur or even in the event of transient changes. As already mentioned, the only other technique capable of such a level of confidence and detail is PNR.[5] The similarities between PNR and XRMS using polarized x-rays hardly need to be stressed, but the element selectivity and high sensitivity of XRMS make it an exceptional tool for investigating the magnetization processes and the correlated MR in multilayer structures. In particular, the strong magnetic signals that can be obtained using polarized soft x rays provide easy access to time-dependent analysis and open the way to future studies of dynamic processes in magnetoresistive devices.

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