Hysteresis curves of ferromagnetic and antiferromagnetic order in metallic multilayers by resonant x-ray scattering

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We have measured field-dependent resonant magnetic scattering of soft x rays from a Co/Cu multilayer exhibiting giant magnetoresistance. We show that, choosing the appropriate experimental geometries, one can draw hysteresis loops of antiferromagnetic as well as ferromagnetic order.

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The use of x-ray spectroscopic tools to investigate the magnetic properties of solids is relatively recent, but it has found widespread applications over the last decade.[1] X-ray-absorption spectroscopy is certainly the most commonly used amongst these techniques, but x-ray scattering has also been successfully applied to magnetic studies. The magnetic signals in such scattering measurements are strongly enhanced if the experiment is performed under resonant conditions, i.e., if the incident photon energy is tuned to an x-ray-absorption threshold of the element of interest. This results from the magnetization dependence of the resonant matrix elements affecting the optical constants.[2] Scattering techniques are also intrinsically sensitive to the spatial modulation of the optical constants and additionally provide access to the magnetic structure of a given sample.[3–6]

In this paper we show how antiferromagnetic (AF) ordering may be measured directly as a function of the applied field **H**. Such measurements provide what it is convenient to call a hysteresis loop of the antiferromagnetic order. It is analogous to the ferromagnetic (FM) hysteresis loops already reported in the literature.[7] Given a judicious choice of experimental geometries we show that x-ray resonant magnetic scattering (XRMS) measurements can provide information on complex magnetic behavior not readily available by other means.

To demonstrate the advantages of the technique we have chosen a metallic multilayer with the structure $(Cu_{22.8}/Co_{11.2})_{20}$, deposited on a Si substrate. Co/Cu multilayers are to some extent prototypical of the systems used for magnetoresistive devices,[8,9] and have already found applications as reading heads in information storage systems. Spin-dependent transport models are used to interpret the way the electronic properties, especially resistance, respond to the presence of external magnetic fields.[9,10] The behavior of such multilayer devices is often complex but, schematically, each individual Co layer is assumed to be ferromagnetically ordered, then low- and high-resistance states are associated with parallel and antiparallel coupling between adjacent Co layers through the Cu spacer. The complexity of the coupling between Co layers is reflected in that of the magnetoresistive behavior.[9,11]

The Cu thickness that we have chosen is close to the so-called second AF peak[8,9] where, in zero field, antiparallel coupling between Co layers is favored, but complete AF ordering is not reached.

XRMS experiments were performed at the bending magnet beamline 6.3.2, at the Advanced Light Source (Lawrence Berkeley National Laboratory).[12] Part of the beam was blanked-off in order to obtain $\sim 60\%$ circular polarization. Measurements with linear *s* polarization were performed at the SuperACO storage ring of Laboratoire pour l'Utilisation du Rayonnement Electromagnétique (LURE) (Orsay), using the undulator beamline SU-7. The resolving power at the Co *L* edges was of ~ 2000 in the former setup and of ~ 1500 in the latter. Specularly reflected x-ray intensity was measured versus grazing angle θ (θ-2θ mode). An external field **H** of up to 1 kOe could be applied along the sample surface by means of an electromagnet. The field was applied either in the scattering plane (longitudinal geometry) or perpendicular to it (transverse geometry). The sample resistance was recorded *in situ* using a two-point technique during the XRMS measurements.

We first consider in detail the case of XRMS measurements performed using circularly polarized photons and start off by showing the scattered intensity as a function of angle and photon energy for an as-prepared sample with $\mathbf{H}=0$ (Fig. 1, bottom panel). The interleaved ordered structure of Co and Cu layers has a spacing of $d_{charge}=34$ Å and consists of 20 periods. It leads to a strong Bragg peak at $\theta \simeq 13°-14°$. The variation in its position and intensity as the photon energy approaches the Co L_3 maximum (778 eV) reflects the resonant variation of the diagonal elements of the dielectric tensor.[13]

In addition to Bragg diffraction and Kiessig fringes, we also observe the presence of a peak at $\theta \simeq 7°$ whose intensity increases as the Co L_3 absorption maximum is approached.

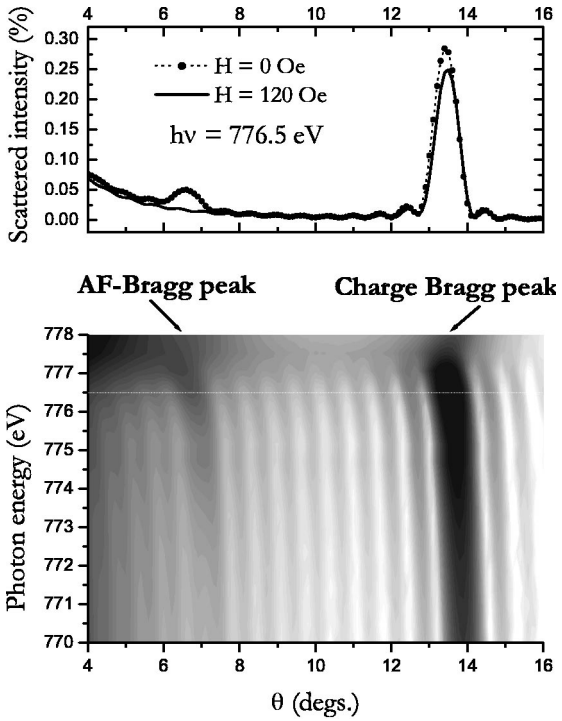

FIG. 1. Lower panel: gray-scale map of measured intensity (I) as a function of photon energy ($\hbar\varpi$) and angle (θ). Top panel: I(θ) in zero field and for $\mathbf{H}=120$ Oe ($\hbar\varpi=776.5$ eV).

Layers of Co with opposite magnetization directions ($Co^{\uparrow}$ and $Co^{\downarrow}$) behave as though they were of different materials since the optical constants at resonance depend on magnetization. Therefore, the $\theta\simeq 7°$ peak corresponds to an order parameter double that of the multilayer structure ($d_{AF}=68$ Å) and is due to a ($Co^{\uparrow}$/Cu/$Co^{\downarrow}$/Cu) stacking.[5] By performing photon energy and θ-2θ scans, we determined the best photon energy for observing the AF peak to be 776.5 eV (Fig. 1, top panel). The optimum energy is a compromise involving maximum contrast between the $Co^{\uparrow}$ and $Co^{\downarrow}$ optical constants (magnetic contributions to the real and to the imaginary parts do not peak at the same energy) and the number of layers involved in the scattering process (the number of layers that are probed decreases as absorption increases). The top panel of Fig. 1 shows the evolution of the scattered intensity when $\mathbf{H}=120$ Oe in the scattering plane (longitudinal geometry). The disappearance of the $\theta\simeq 7°$ peak is clear proof of its AF origin. In the following, we will consider three parameters that affect the scattered intensity versus applied field, namely, the scattering angle, the polarization of the light and the direction of **H**.

The scattering angle may be set to satisfy Bragg's law for the order parameter defined by either ferromagnetic ($d_{FM}\equiv d_{charge}$, $\theta\simeq 13.5°$) or antiferromagnetic ($d_{AF}, \theta\simeq 6.75°$) stacking of the Co layers.

The polarization state of the photons determines the magnetic contribution to the resonant scattering amplitude according to the coefficient $(\mathbf{e}_f^*\times\mathbf{e}_i)\cdot\mathbf{m}$, where $\mathbf{e}_i$ and $\mathbf{e}_f$ are the incoming and outgoing polarizations and **m** is the unit vector in the direction of the magnetization.[2] When the polarization is circular or linear *s* (electric vector normal to the scattering plane), as in the examples given below, the cross product $(\mathbf{e}_f^*\times\mathbf{e}_i)$ lies in the scattering plane. Therefore, measurements performed under these conditions will be sensitive to the projection of the magnetization in the scattering plane.[14] The above argument implies that sensitivity to the magnetization components of the sample along or normal to the direction of **H** will depend, respectively, on whether longitudinal or transverse geometries are used.

Figure 2(a) refers to $\theta=13.5°$ and to the use of the longitudinal geometry and of circularly polarized light. The scattered intensity mimics the total magnetization measurements performed on the same sample [see Fig. 3(a)] and provides an element-specific FM hysteresis loop. Setting $\theta=13.5°$ makes the measurement sensitive to the ordered stacking of the FM layers. However, it is worth noting that specularly reflected intensity would be sensitive to the projection of the sample's net magnetization in the scattering plane also at other angles.[15] It is clear, however, that using synchrotron radiation to obtain the FM hysteresis of a system with only one magnetic component is of limited interest.

In Fig. 2(b) the scattering angle is now 6.75° thus highlighting the periodicity imposed by antiparallel coupling between Co layers. The AF signal appears only relatively weakly, and the field dependence is dominated by the switching of the net magnetization component with respect to the photon (circular) polarization. It is important to remember that, being in longitudinal geometry, the intensity underlying the AF peak is sensitive to the net magnetization along the direction of the applied field.

The curve in Fig. 2(c) is obtained under exactly the same conditions as in Fig. 2(b) ($\theta=6.75°$), but the field is now applied normal to the scattering plane (transverse geometry). In a system characterized by AF coupling, the application of weak **H** (i.e., weaker than that required to impose FM order) favors AF domain ordering. The magnetization is aligned normal to **H**.[16] The curve shown in Fig. 2(c) is thus dominated by the AF signal (AF domains are oriented normal to the applied field, that is to say they lie in the scattering plane). The measurement is insensitive to the net magnetization direction. This is because it points along **H** and is normal to the scattering plane.

Sensitivity to AF ordering alone [Fig. 2(d)] can be attained by using linearly *s*-polarized light. As in the Kerr effect, polarization rotation occurs upon scattering from a magnetic material (see, for instance, Refs. 5, 16, and 17). The rotation angle is proportional to the magnetization component in the scattering plane. Layers with opposite magnetizations induce rotations of the polarization vector in opposite directions. An ordered stack of layers with alternate magnetization, such as ($Co^{\uparrow}$/Cu/$Co^{\downarrow}$/Cu), defines a superstructure with an associated Bragg peak. The scattered intensity will be the same for both ($Co^{\uparrow}$/Cu/$Co^{\uparrow}$/Cu) and ($Co^{\downarrow}$/Cu/$Co^{\downarrow}$/Cu) configurations. It explains the lack of sensitivity to FM order. The intensity in Fig. 2(d) is a measure of the AF coupled fraction of the multilayer with magnetization in the scattering plane, as a function of **H** applied normal to the scattering plane.

These XRMS results may be related to the macroscopic properties of the sample, such as the field dependence of its

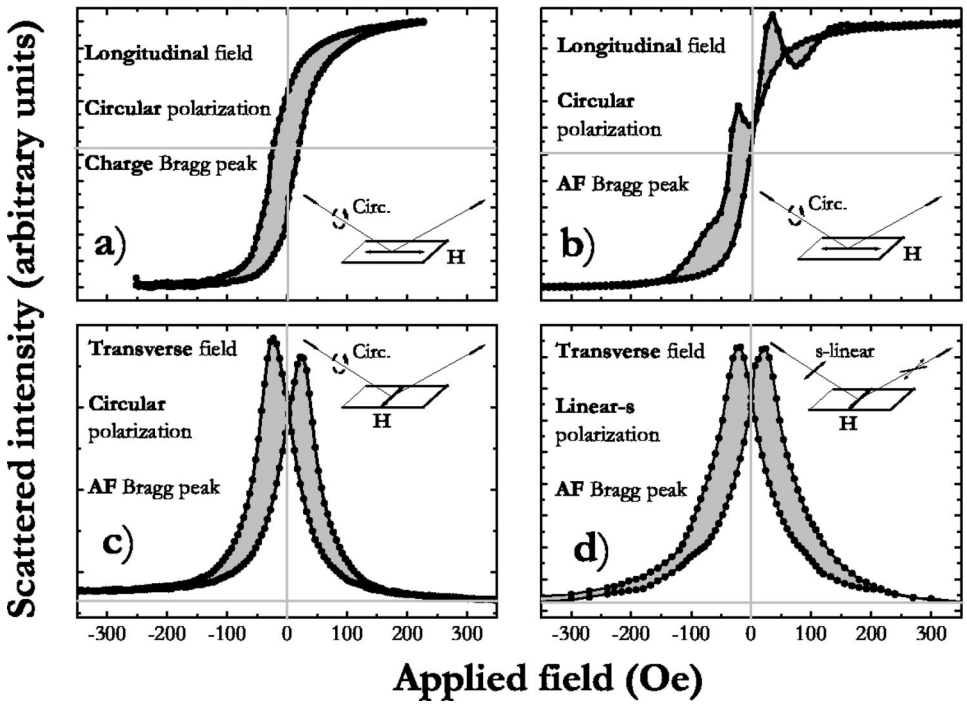

FIG. 2. Field dependence of the scattered intensity in four experimental geometries (see text). Photon energy is always 776.5 eV.

total magnetization [Fig. 3(a)] and of its resistance [Fig. 3(b)]. As mentioned before, the curve in Fig. 2(a) is sensitive to the projection of the magnetization along the field direction. It is therefore straightforward to identify it with the hysteresis of the total magnetization shown in Fig. 3(a). The percentage of magnetization at remanence ($\sim$48%), as well as the values of the field giving zero magnetization ($\sim$23 Oe), and the closure of the loop ($\sim$120 Oe), coincide in the two measurements.

Comparing Figs. 2 and 3, we observe an obvious similarity between magnetoresistance (MR) and the results of scattering experiments in transverse geometry. In particular, the value of **H** giving the peak in MR corresponds to the maximum of the AF signal [Figs. 2(b)–2(d)] and to zero total magnetization [Fig. 2(a)]. For a weak field of 20–30 Oe, the MR peak is associated with the presence of an ordered AF structure, where magnetization of each layer is dominantly oriented normal to the direction of **H** [see Figs. 2(b) and 2(c)].

The choice of the scattering angle (corresponding to e.g., a Bragg peak, Kiessig fringes, or the total reflection regime) is known to influence the exact line shape of the hysteresis, and care must be taken in drawing quantitative conclusions.[6] Nevertheless, resonant scattering in the geometry of Fig. 2(a) remains a powerful method for obtaining the field dependence of the magnetization, element selectively.

We emphasize that the scattered intensity from AF order necessarily involves tuning the photon energy to a core resonance. Here only scattering at the Co $2p$ resonance allows a clear distinction to be made between $Co^{\uparrow}$ and $Co^{\downarrow}$. Scattering at resonance is essential to this type of study. The potential of XRMS for a field-dependent analysis of AF coupling was first illustrated by Idzerda and co-workers.[6] Their results, though, were based on a model that requires quite extensive data manipulation and relies on a number of hypotheses. Our approach offers direct experimental access to antiferromagnetic hysteresis curves.

AF coupling is never easy to investigate directly, and polarized neutron reflectivity (PNR) is probably the only other technique capable of such an analysis.[18] From our experiments two advantages of XRMS over PNR emerge. XRMS offers element selectivity, of course. This remains important in the analysis of composite magnetic systems. But equally as important, the scattered intensity and its magnetic component at soft x-ray resonances allow for fast data acquisition. Time resolved experiments are still at an exploratory stage, but preliminary tests indicate that XRMS will give new opportunities to investigate magnetic dynamics with all the advantages of a resonant scattering experiment. The capability of drawing element-selective hysteresis loops of both ferro- and antiferromagnetic order opens up new perspectives in the analysis and understanding of the fundamental processes in complex magnetic systems.

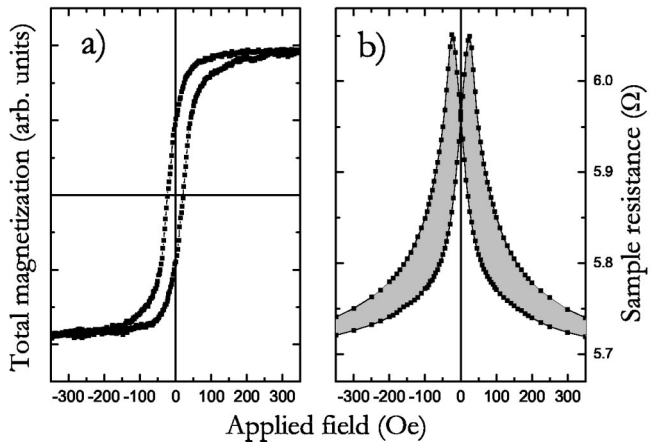

FIG. 3. (a) Total magnetization curve. (b) Sample resistance vs applied field **H** (two-point measurement, current in plane).

*Also at LURE.

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