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Study of the magnetic order in a Co/Cr multilayer by magnetic Bragg diffraction at the Co 2p resonance

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Abstract

We have measured the resonant scattering from an antiferromagnetic Co/Cr multilayer at photon energies close to the cobalt 2p → 3d transitions. The cobalt dielectric tensor has an anisotropic component, enhanced by resonance, which depends on the magnetic order and follows its modulation inside the sample. We have studied the vertical distribution of this component through the dependence of the reflectivity on the scattering angle. Using s-polarized light, we have observed the signature of the cobalt–cobalt antiferromagnetic coupling as an half-integer-order Bragg peak. Experimental results have been analyzed by numerical simulation. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

The measurement of the resonant scattering of polarized X-rays is a valuable tool to investigate the magnetic order of matter. First experiments, performed in the hard X-ray energy region measuring the Bragg diffraction at the 3d edges of actinide compounds [1] and at the 2p edges of rare earths [2], had a strong impact on the study of these systems. Soon after, large magnetic effects were observed in the resonant reflectivity from Fe [3,4], Co [5] and Ni [6] in the soft X-rays range at the $L_{2,3}$ edges. The wavelengths required to excite these resonances and in general those of the 3d transition metals (TM) $L_{2,3}$ edges, are in the order of tens of Å, preventing experiments under Bragg diffraction conditions for most crystalline materials. Enhanced magnetic signals can be recovered for artificially layered structures, where the Bragg condition depends on the chemical and magnetic modulation periods [7–10]. In these samples, the distribution of the magnetic moments can be highlighted by studying the behavior of the Bragg peaks in $\theta/2\theta$ scans. This has opened an entirely new experimental field of investigation which can

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address phenomena like oscillatory exchange-coupling, perpendicular anisotropy and enhanced interfacial magnetic moments. Recent studies on a Ce/Fe multilayer, for instance, have measured the profile of Fe-induced magnetization inside the Ce layers [11] by observation of high-order Bragg peaks.

In this work we discuss the resonant magnetic reflectivity at the cobalt $2p \rightarrow 3d$ resonance for a cobalt–chromium multilayer. This system is known to exhibit an oscillatory exchange coupling between cobalt layers as a function of the Cr thickness [12]. For the sample that we considered (10 Å Cr and 17 Å Co), cobalt layers are antiferromagnetically coupled. We have analyzed this antiferromagnetic ordering by resonant magnetic scattering at the $L_{2,3}$ edges of cobalt.

2. Experimental method

A (Co17 Å/Cr10 Å) × 30 multilayer was grown by DC magnetron sputtering in a UHV compatible sputtering system [13]. The multilayer was deposited on a 50 Å Cr buffer layer grown on a chemically etched Si(0 0 1) substrate. A 50 Å Cu overlayer was deposited to protect the sample. The Ar pressure was set to 3 mTorr and the deposition rates for Co, Cr and Cu were 3 Å/s. The multilayer was first characterized by conventional X-ray reflectometry and the multilayer period deduced from the low-angle Bragg peaks, was found in good agreement with the nominal value.

The in-plane magnetization loop at room temperature was measured with a Quantum Design MPMS Squid magnetometer. As shown in Fig. 1, the loop displays the signature of an antiferromagnetic (AF) coupling between the Co layers: a large saturation field H_s of about 1 T is necessary to overcome the AF coupling, and remanent magnetization M_r is only 0.45% of the saturation value M_s. H_s can be related to the AF interlayer exchange coupling J by the well known formula [14], $-4J = H_s M_s t_F$. For this multilayer we find $J = -0.41\,\mathrm{erg\,s\,cm^{-2}}$.

X-ray scattering measurement were performed at the 5U.1 undulator beam-line [15] of the SRS storage ring (Daresbury laboratory), delivering linearly

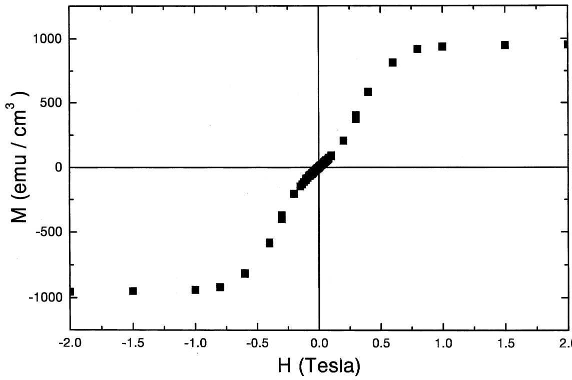

Fig. 1. Magnetization loop at room temperature for the Co/Cr multilayer, measured by SQUID magnetometry.

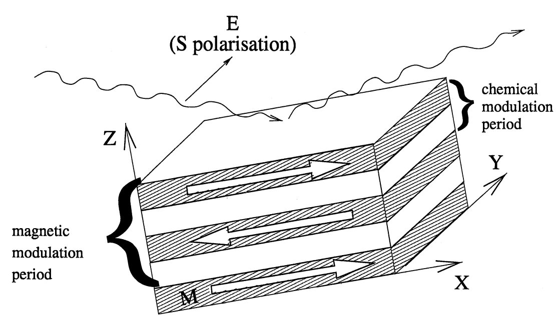

Fig. 2. Sketch of the experimental setup.

polarized photons over the 60–1000 eV range. In our experimental conditions, the plane grating monochromator gave a resolving power of about 3000 at the Co L-edges. The endstation was a vertical scattering $\theta/2\theta$ reflectometer working in vacuum ($\sim 10^{-6}$ mbar), separated from the beamline by a highly transparent window. In this scattering geometry, sketched in Fig. 2, the polarization of the incoming light is along the y-direction in the surface plane and normal to the scattering plane (S-polarized light). Measurements were performed both in remanent conditions and in the presence of a permanent magnet giving a field of about 1 kG at the sample surface. This weak field, well below the value necessary to impose a parallel orientation of all the Co layers, is sufficient to break the x–y plane symmetry of the antiferromagnetic domains. Applying the field along the x (or y) direction favors the orientation of the domains along the y (or x)

orthogonal axis. The monochromator energy calibration was fixed by recording absorption spectra at the Co 2p → 3d resonances and by comparing to cobalt absorption spectra from the literature [16].

3. Numerical method

The calculation of reflectivity from the magnetic multilayers is done taking into account the tensorial nature of the dielectric constant in magnetic materials. The first step of the procedure is to find, for given values of photon energy and incidence angle, the electromagnetic (EM) eigenmodes inside a uniform tensorial medium. These eigenmodes correspond to the solution of a second-order differential equation [17]. The four eigenmodes found correspond to the two opposite propagation directions (ingoing and outgoing) and the two photon polarizations which are given by the solution of the eigenproblem may vary between linear and circular.

To calculate a multilayer stack, the eigenmodes for all the layers of the stack must be found first, then Fresnel's equations have to be solved for the interfaces between layers.

Fresnel's equations impose four conditions of continuity on the parallel components of the EM field across the interfaces (two parallel components for E and two for B). When the EM field across the interface is decomposed in the eigenmodes basis, the resulting 4×4 matrix describes the linear dependence between the eigenmode coefficients on both sides of the interface. The interface roughness (which can be due to either interdiffusion or corrugation) is taken into account by introducing Debye–Waller-like factors [18].

The propagation of the modes across the thickness of a layer is described by a 4×4 diagonal matrix whose diagonal elements are the propagation factors for the eigenmodes (which are the exponential of the eigenvalue times the layer thickness).

At the end of the procedure the whole stack is described by a 4×4 matrix, obtained by multiplying the interface and the propagation matrices together from one side to the other side of the multilayer. Reflectivity is obtained by imposing the condition of outgoing waves in the substrate.

For chromium, we used a scalar dielectric constant obtained from tabulated values [19], since we are far from the chromium resonances. For cobalt, we started from the absorption spectra reported in the literature [16] of bulk magnetized metallic cobalt measured for positive and negative helicity of the circularly polarized photons. The absorption data were rescaled to join the tabulated values [19] for cobalt on the two sides of the 2p → 3d edges to obtain the imaginary part of the index, β^+ and β^-. The decrement to the real part of the optical index, $\delta^\pm$, was obtained by Kramers-Kronig transformation of $\beta^\pm$. The dielectric tensor was constructed on the basis of the optical indexes $n_\pm = 1 - \delta^\pm - i\beta^\pm$, neglecting linear dichroism [17]. In the (xyz) frame of Fig. 2, and for a magnetization along the x-axis ($M = m\hat{e}_x$), the cobalt dielectric tensor is

$$\varepsilon_{M=m\hat{e}_x} = \begin{pmatrix} (\varepsilon_+ + \varepsilon_-)/2 & 0 & 0 \\ 0 & (\varepsilon_+ + \varepsilon_-)/2 & \pm i(\varepsilon_+ - \varepsilon_-)/2 \\ 0 & \mp i(\varepsilon_+ - \varepsilon_-)/2 & (\varepsilon_+ + \varepsilon_-)/2 \end{pmatrix}, \quad (1)$$

where $\varepsilon_\pm = n_\pm^2$, the $\pm$ sign depends on the sign of the magnetization (it alternates over the cobalt layers for antiferromagnetic coupling). The dielectric tensor for y magnetization is written similarly as

$$\varepsilon_{M=m\hat{e}_y} = \begin{pmatrix} (\varepsilon_+ + \varepsilon_-)/2 & 0 & \pm i(\varepsilon_+ - \varepsilon_-)/2 \\ 0 & (\varepsilon_+ + \varepsilon_-)/2 & 0 \\ \mp i(\varepsilon_+ - \varepsilon_-)/2 & 0 & (\varepsilon_+ + \varepsilon_-)/2 \end{pmatrix} \quad (2)$$

4. Results and discussion

We show in Fig. 3 the reflectivity versus grazing angle for x and y magnetization (M) at a photon energy of 773.5 eV. This energy lies immediately

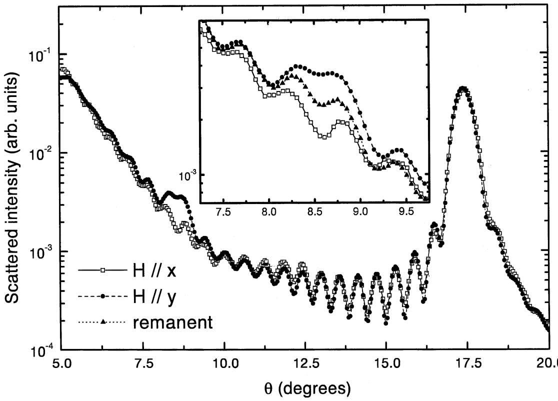

Fig. 3. Experimental results of $\theta/2\theta$ scans at 773.5 eV photon energy. Open squares refer to measurements performed in an external field H applied in the scattering plane (x-direction), which orients the sample magnetization along y. Filled circles refer to $H \| y$ and magnetization along x. The main peak at about 17.5° is due to first-order Bragg diffraction from the chemical modulation of the multilayer. The half-order Bragg peak due to antiferromagnetic order is around $\theta = 8.5°$. The inset shows the detail of the magnetic peak, with in the addition the spectrum is measured in remanent conditions with an isotropic distribution of the magnetic domain directions in the sample (no external field).

before the cobalt $2p \rightarrow 3d$ transition so that the absorption is still low but the dichroism in the real part of the index is strong. The reflectivity oscillations of the period about 1° arise from interference between the signals from the top and the bottom of the multilayer (the whole stack is probed at this energy). This phenomenon is well known in the literature on X–UV multilayers [20] and the oscillations are called Kiessig fringes. The number of fringes between two Bragg peaks is related to the multilayer total thickness. The curve of Fig. 3 shows a peak at an angle of about 8.5° that is half that of the first-order Bragg peak (about 17.5°). This signal appears when the magnetic polarization is parallel to x and disappears completely when it is parallel to the y-axis. We show in the inset of Fig. 3 the details of the magnetic peak, together with the reflectivity curve measured in remanent conditions ($H = 0$). This latter curve corresponds to disordered magnetic domains and gives a lower magnetic peak than for the x-polarized case. The origin of the magnetic signal can be understood considering that in order to observe a magnetic diffraction peak of the product $M(E_{in} \times E_{out})$, where $E_{in(out)}$ indicates the polarization of the ingoing (outgoing) wave, must be non-zero [15]. Assuming E_{in} along the y-direction (S-polarization), this implies that the scattering process induces a polarization rotation and E_{out} has a component in the scattering plane ($S \rightarrow P$ scattering channel). When the external field orients the magnetization along the y-axis, E_{in} is parallel to M and the product vanishes. For M along x, on the contrary, $M(E_{in} \times E_{out})$ is non-zero and is proportional to the P component of E_{out}. Moreover each cobalt layer contributes to the $S \rightarrow P$ channel with a phase that is the sum of an optical-path-dependent term plus a $\pm \pi$ term that depends on the sign of the magnetization. As a consequence the $S \rightarrow P$ channel is sensitive to the magnetization modulation. The magnetic signal appearing at an angle equal to half of the first Bragg order angle indicates that the magnetic modulation has a period double than the chemical modulation (see Fig. 2) and is a clear signature of antiferromagnetic coupling between cobalt layers.

When the magnetization is turned from x to y the S to P channel is closed because $\varepsilon_{M=m\hat{e}_x}$, given by Eq. (2), has no off-diagonal element connected to the y-direction. As a conseguence the $M \| y$ curve has no magnetic peak around 8.5°.

The magnetic peak brings us information on both the magnetic polarization intensity and its distribution inside the sample. Regarding the distribution, the width of the magnetic signal is sensitive to the coherence of antiferromagnetic coupling. In the case of perfect antiferromagnetic ordering of the cobalt layers the width is inversely proportional to the number of layers but it is expected to get larger in case of imperfect antiferromagnetic alignment. The peak height is sensitive to magnetization intensity and distribution inside the sample.

We show in Figs. 4 and 5 the numerical fits at 773.5 and 787.5 eV for the x-polarized and y-polarized case, respectively. The simulation is done considering a 16.7 and 10.4 A thicknesses for cobalt and chromium, respectively, and perfect antiferromagnetic ordering between all the cobalt layers. We consider a $\sigma = 6$ Å roughness for the cobalt-crome interface.

We also considered a slight drift of the layer thicknesses of about 0.1 A from top to the bottom

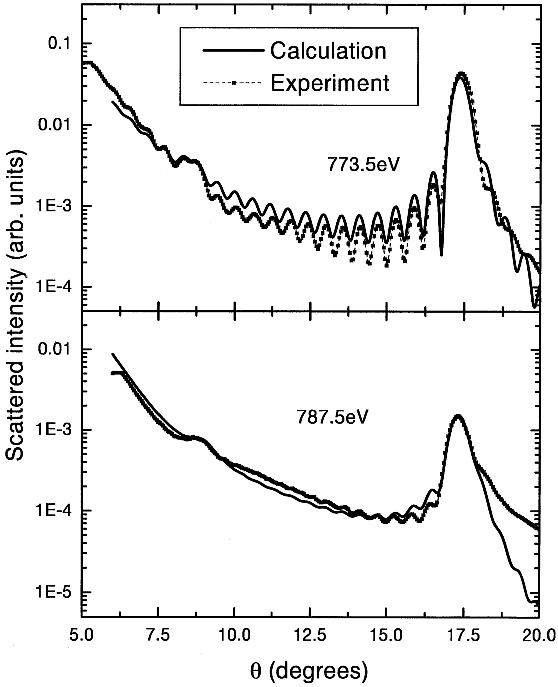

Fig. 4. Calculated and experimental scattered intensity at 773.5 and 787.5 eV photon energy for a magnetization orientation along x. The simulation is done considering a 16.7 and 10.4 A thicknesses for cobalt and chromium, respectively, and perfect antiferromagnetic ordering between all the cobalt layers. In order to fit the height of the half-order Bragg peak, the magnetic part of the refractive index has been reduced by a factor 0.6 with respect to bulk values.

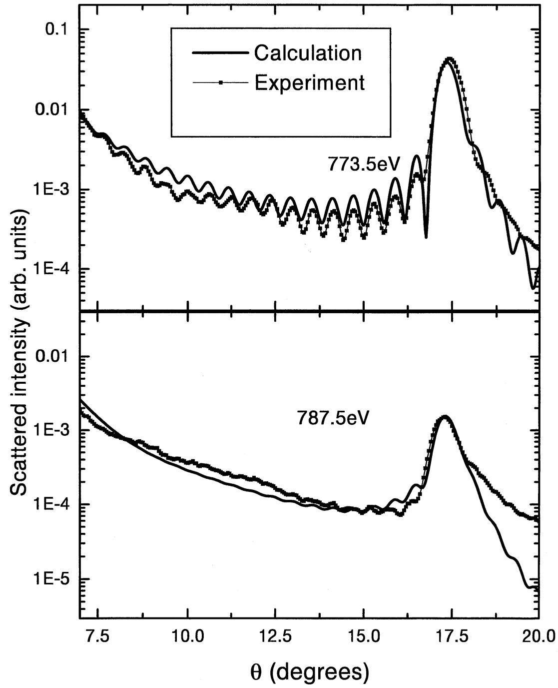

Fig. 5. As in Fig. 4, for a magnetization oriented along y.

of the multilayers. This drift has been chosen in order to fit correctly the positions of the Kiessig fringes which are very sensitive to the thickness drift, even when they are too small to affect the Bragg or the magnetic peaks. In order to fit the height of the magnetic peak the asymmetry ratio between the two helicities has been rescaled with respect to the bulk values [16] by a factor 0.6. This reduction might simulate an interface effect.

We also simulated a non-uniform distribution of the magnetic-scattering amplitude within the Co layer splitting it in to three layers: a central one having bulk constants and two interface layers characterized by a reduced magnetic moment. The reduction factor was taken as a function of the interface layers thickness in order to keep the total magnetization constant. We find that the calculated magnetic peak strongly depends on the magnetization distribution and we obtain the best agreement when the central layer has zero thickness, which corresponds to a 0.6 magnetic moment reduction factor, uniform over the entire cobalt layer.

The y-polarized case fits well to the calculated reflectivity assuming complete y-polarization and does not show any magnetic peak. An incomplete polarization could explain the height of the magnetic peak without the need to scale the asymmetry for cobalt, but then a magnetic peak should be visible even in the y-polarized case.

In order to check the sensitivity of the magnetic order we compare in Figs. 6 and 7 the experimental reflectivity to a simulation done assuming the same structural parameters as those used in Figs. 4 and 5 and a ‘slip’ in the antiferromagnetic alignment at a given point in the stack.

In particular, we considered two consecutive cobalt layers having parallel magnetization. Figs. 6 and 7 show the magnetic peak at 773.5 and 787.5 eV

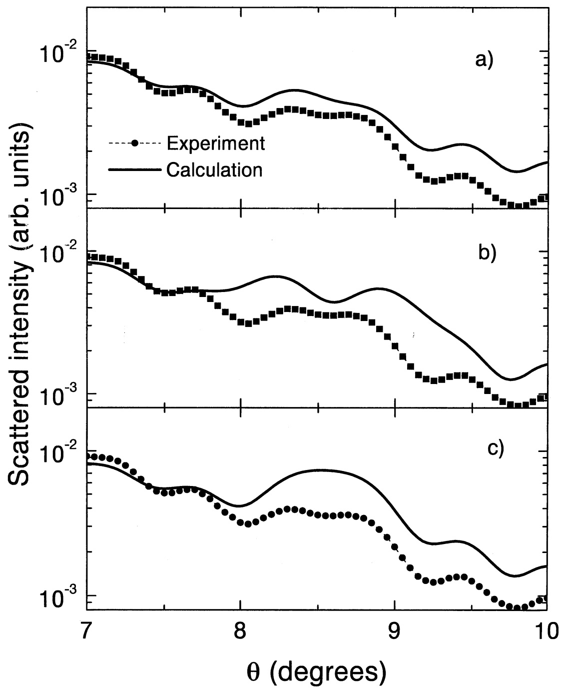

Fig. 6. Calculated and experimental reflectivity at 773.5 eV photon energy for x-oriented magnetization. The simulation is done considering the same structural parameters as in Fig. 3 but imperfect antiferromagnetic ordering in the stack and using unscaled magneto–optical constants. In the simulation a slip in the antiferromagnetic alignment is introduced at the top (a), middle (b) and bottom (c) of the multilayer. The slip consists of two consecutive cobalt layers ferromagnetically coupled.

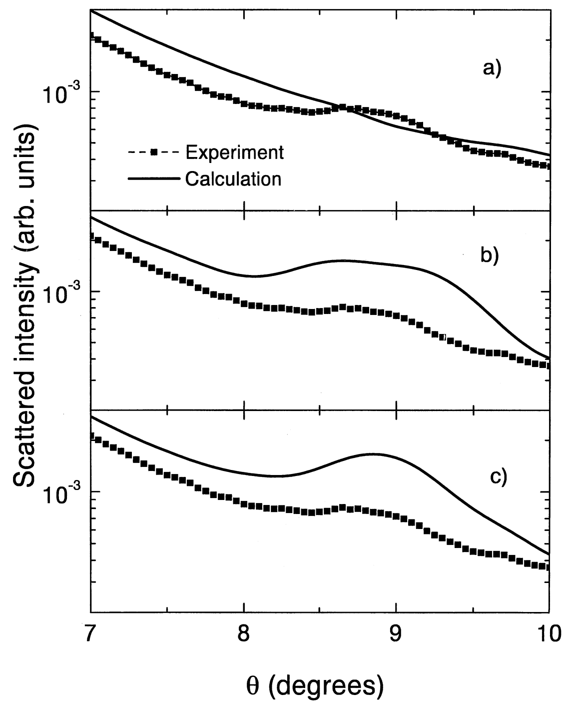

Fig. 7. Same as Fig. 5, for a photon energy of 787.5 eV.

respectively for three different positions of the slip, using x-polarization. The three positions are at the bottom, in the middle and at the top of the multilayer. The simulation is done assuming bulk optical constants for cobalt.

The height of the simulated magnetic peak is lowered by destructive interference in all three cases. This reduction is large when the slip is at the top, and lower when the slip is at the bottom because of absorption in the sample. In the 787.5 eV case the slip destroys completely the magnetic peak.

In the 773.5 eV case a slip in the antiferromagnetic alignment at the top of the multilayer could explain the magnetic peak height without the need of the asymmetry reduction hypothesis but this would give no magnetic peak at 787.5 eV.

5. Conclusion

We have studied the antiferromagnetic coupling between cobalt layers in a Co/Cr multilayer by reflectivity measurements versus incidence angle at photon energies close to the cobalt $2p \rightarrow 3d$ transitions. The cobalt dielectric tensor has an anisotropic component, enhanced by resonance, which depends on the magnetic order and follows its modulation inside the sample. We have studied the vertical distribution of this component through the dependence of the reflectivity on the scattering angle. We have polarized the cobalt layers in the tangential (sagittal) direction by a weak sagittal (tangential) magnetic field and using S-polarized light, we have observed the signature of the cobalt–cobalt antiferromagnetic coupling as an half-integer-order Bragg peak. By numerical simulation of the experimental results we have found that the height of this peak cannot be explained assuming bulk optical constants for cobalt. By introducing a reduction of 60% of the asymmetry ratio the magnetic peak can be reproduced with good agreement. On the other hand, the hypothesis of incomplete magnetic alignment, or imperfect antiferromagnetic coupling within the stack, could explain the lowering of the magnetic peak in certain cases but cannot give a good agreement with the whole set of experimental results. Within the limits imposed by available experimental data, the analysis of the peak height seems to indicate a reduction, compared to the bulk value, of the magnetic dependent part of the cobalt dielectric tensor. In terms of Co magnetic moment, this reduction of about 60% leads to an estimated magnetic moment per Co atom of the order of $1\,\mu_B$. From the SQUID measurements we find an average magnetic moment of $1.16 \pm 0.06\mu_\beta$ per Co atom which is not far from the estimated moment deduced from reflectivity. The slight difference can be understood in terms of sensitivity of the two methods to the interfacial magnetism. It is highly probable that the magnetization is more strongly reduced at the Co/Cr interfaces, due to chemical intermixing for example, than in the interior of the Co layers.

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