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Resonant magnetic x-ray and neutron diffuse studies of transition metal multilayers

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Electron scattering mechanisms within metallic multilayers are affected by both structural and magnetic disorders. Off-specular x-ray scattering has long been used to probe the structural interfaces, and it is only recently that it has been applied to the study of magnetic disorder. We compare the resonant magnetic x-ray scattering with off-specular neutron studies from magnetron-sputtered Co/Cu and Co/Ru multilayers grown at the second antiferromagnetic coupling peak. Both techniques yield similar results for the Cu system, and a simple domain model can be applied to extract the magnetic interface morphological parameters. For the Cu system, the in-plane correlation length is field dependent and is 880±20 Å after saturation along the hard axis, but increases to 7000±100 Å after saturation along the orthogonal easy axis. Both systems show strong out-of-plane correlations in both the structural and magnetic disorders. In all cases, the out-of-plane correlation length for the structural interfaces is 200–250 Å, but the ratio of the magnetic to structural correlations length is dependent on the magnitude of the exchange coupling and ranges from 0.4 to 1.4. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1543876]

In recent years, there has been considerable interest in the magnetotransport properties of thin magnetic films such as spin valves and multilayers. Their magnetoresistive properties have been exploited in the fields of magnetic read heads and magnetic memory storage.[1,2] Use of a wide variety of characterization techniques, such as high-resolution transmission electron microscopy as well as x-ray and neutron scattering, have resulted in significant progress in correlating structure with transport properties, thereby improving the efficiency of such devices. However, in order to understand completely the magnetotransport properties of spin valves and magnetic multilayers, a full description and thus characterization of the magnetic structure is required. The giant magnetoresistance properties resulting from spin-dependent scattering is often hypothesized to occur at the interface. The role of the chemical interface is now becoming better understood, but the magnetic interface is also a key component in understanding both spin accumulation and electron scattering.

Initial characterization techniques concentrated on the chemical structure, with particular emphasis being placed on the role of the interface morphology with respect to the transport. Modeling the distribution of scattered intensity of either neutrons or x-rays around the origin of reciprocal space allows the chemical interface morphology to be deduced, and to be separated from interdiffusion effects.[3] The high flux of synchrotron sources has greatly advanced diffuse x-ray scattering studies, and both experimental ideas and theoretical concepts are well developed. Although x-ray scattering has proved to be immensely beneficial in elucidating structural parameters, the technique has little sensitivity to the magnetization profile. Away from resonance, the magnetic contribution is many orders of magnitude weaker than the Thompson charge scattering. In the past, the characterization of the magnetic structure has been confined to polarized neutron reflectometry.[4] This technique probes the atomic moment directly, with the neutron–spin interaction being well understood. Advances in detector design and increases in source flux have enabled off-specular and other diffuse neutron studies to be performed routinely.

However, recent synchrotron experiments,[5,6] in which the incident x-ray energy is tuned close to an atomic transition, directly probe the magnetic band structure and provide a magnetic scattering amplitude comparable to that of the charge scatter. For transition metals, it is the empty states in the $3d$ band that give rise to the magnetization, thus experiments need to be conducted at the L_{III} and L_{II} edges, which for cobalt are at 778.1 and 793.2 eV, respectively.[7] This provides an alternative magnetic characterization technique that has the advantage of high synchrotron flux, coupled with element specificity. However, since the interaction between the photon and the magnetic moment is indirect, it cannot be separated uniquely from the charge scattering.

Multilayers of Co/Cu and Co/Ru were prepared by magnetron sputtering in a system with a base pressure better than 10^{-8} Torr. The thickness of the spacer layer was adjusted to correspond to the second antiferromagnetic (AF) coupling maxima. The samples were deposited on Si (001) substrates without removal of the native oxide. The interlayer exchange coupling in the Co/Ru system is significantly greater than that in the Co/Cu case.

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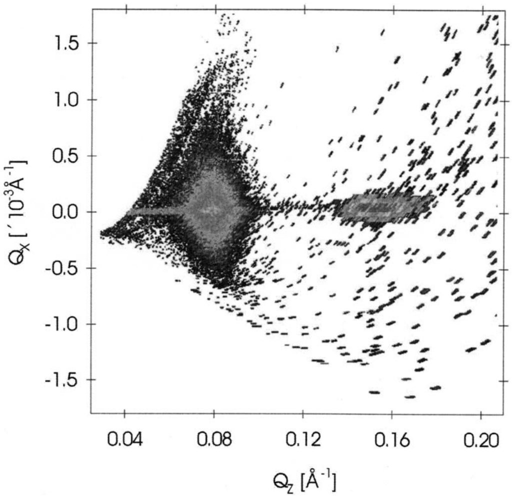

FIG. 1. Full reciprocal space map of the neutron scattering from an AF-coupled Cu/Co multilayer. The specular data are seen in the data as a streak at $q_x = 0$.

Neutron reflectometry was conducted at room temperature on the CRISP reflectometer at the time-of-flight ISIS facility at the Rutherford Appleton Laboratory as a function of applied field. Two-dimensional maps in reciprocal space were recorded using a one-dimensional detector and neutrons with wavelengths in the range 1.2–6.5 Å.

Resonant magnetic x-ray scattering was recorded on beamline 5U1 at the SRS, Daresbury Laboratory. The sample was mounted in an in-vacuum, two-circle diffractometer. Specular and diffuse scatter were recorded in the conventional manner using linearly polarized x-rays. Experiments were conducted at remanence, but prior to sample alignment they were saturated *ex situ* by the application of a 140 mT field along the hard or easy axis.

Figure 1 shows the neutron data recorded at remanence for a sample with nominal structure Co(20 Å)/Cu(22 Å). The Bragg peak associated with the repeat structure occurs at an out-of-plane momentum transfer of $q_z = 0.155$ Å^{-1}. No diffuse intensity associated with the structural interfaces could be observed, and the apparent broadening at the structural Bragg peak is resolution limited. However, this is not the case at the magnetic Bragg peak, centered on $q_z = 0.075$ Å^{-1} where a broad peak in both q_x and q_z can be seen.

The width of the peak in q_z was used to estimate the extent of the out-of-plane correlation of the magnetic interfaces. We observed clear differences between Co/Cu and Co/Ru multilayers.[8] For the Cu system, the magnetic and structural out-of-plane correlation lengths were approximately 600 Å. In the Ru system, the structural correlation length was similar, but that associated with the magnetic disorder was larger and approached 2000 Å, the total thickness of the film.

Resonant magnetic scattering was performed on samples deposited in the same growth run as those used in the neutron experiments. Longitudinal diffuse scans that probe the diffuse scatter as a function of q_z were measured from samples from both the Cu and Ru systems. In addition to those samples deposited with spacer layers corresponding to the second. AF coupling maximum, we also measured those grown at the first AF peak.

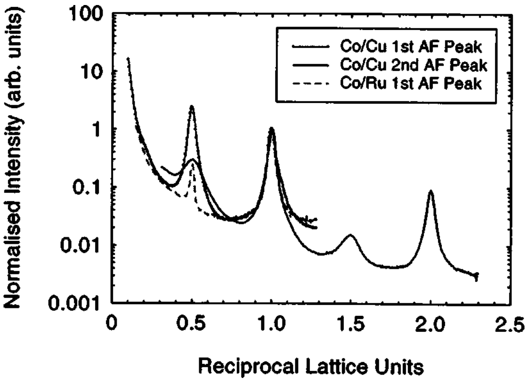

FIG. 2. Longitudinal x-ray diffuse scans for a series of AF-coupled Co multilayers with different spacer layers. Magnetic peaks occur at (0 0 0.5) and (0 0 1.5).

Unlike the neutron data, the x-ray scans showed strong diffuse intensity at both the charge and magnetic Bragg peaks (Fig. 2). On application of a saturating field, the magnetic peaks disappeared. The peaks were fitted to Lorentzians, and we find that for all samples, the out-of-plane correlation length for the structural roughness was similar, and in the range from 200 to 250 ± 20 Å. We have defined the out-of-plane correlation length as being equal to the inverse of the half width at half maximum of the peak in q_z, which corresponds to the length scale at which the out-of-plane correlation has reduced to a value of $1/e$. The greater signal-to-noise ratio in the x-ray data, compared to the neutron data, enabled this more explicit formulism to be used. The width of the magnetic peak varied; in the Ru system the associated magnetic correlation length was always greater than the structural correlation length, the effect being greater for samples prepared at the first AF coupling peak, the ratio of magnetic to structural being 1.4. In the Cu system, the ratio for samples grown with Cu = 9 Å, corresponding to the first AF peak, was 1. Samples prepared at the second coupling maximum had much broader magnetic peaks (Fig. 2), giving a out-of-plane magnetic correlation length of only 90 ± 15 Å and a ratio of 0.4. This length scale was not dependent on magnetization direction. We note that there is a trend towards higher exchange coupling, resulting in larger out-of-plane coherence of the magnetic disorder.

The distribution of the neutron data along the in-plane direction q_x has been modeled assuming a Gaussian distribution of domains, characterized by an in-plane domain correlation function given by:

$$C(R) = \sigma_m^2 \exp\left(-\frac{R}{\xi_m} \right). \tag{1}$$

Here, σ_m is the magnitude of the magnetic domain disorder and ξ_m is the lateral correlation length, defined as the length scale of the magnetic disorder.[9] Equation (1) is a special case of the general correlation function introduced by Sinha *et al.* used to model the charge interface, with the fractal parameter h set to 0.5.[10] Line scans were extracted at the magnetic Bragg peak and the in-plane correlation length found to be 15 000 ± 4000 Å.[8] This value is significantly less

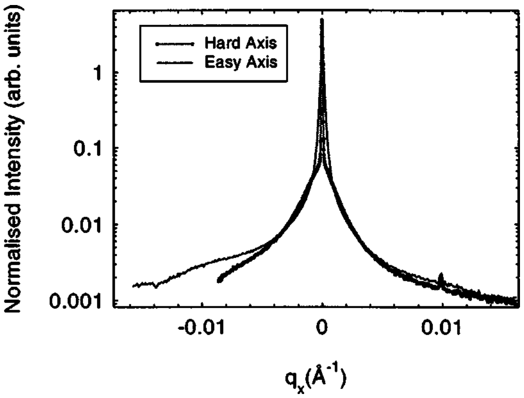

FIG. 3. Transverse x-ray scans at fixed $q_z = 0.1128$ Å taken through the first magnetic Bragg peak for an AF-coupled Cu/Co multilayer for two magnetization directions. The full width at half maximum of the sharp specular peak ($1.103 \pm 0.001 \times 10^{-4}$ Å^{-1}) is the instrument resolution and corresponds to an in-plane length scale of over 5 μm.

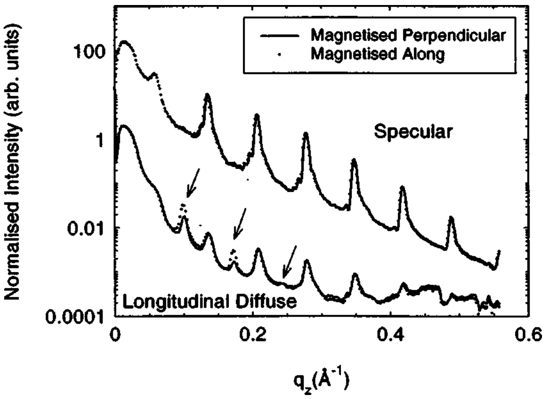

FIG. 4. Specular and diffuse x-ray scans from a Co/Ru multilayer. The magnetic Bragg peaks in the diffuse data are marked by arrows.

than the maximum measurable correlation length at the antiferromagnetic peak of 8 μm set by the instrument resolution.

Similar x-ray data were obtained by conducting transverse scans through the Bragg peaks shown in Fig. 2. Figure 3 shows such transverse scans taken through the magnetic Bragg peak for both magnetization directions.

The data could be fitted to a Lorentzian line shape that corresponds to the Fourier transform of the correlation function given in Eq. (1). We note that the higher flux and extended q range of the synchrotron experiments enables the data to be recorded over a wider intensity range than that possible in the comparable neutron case. The Lorentzian line shape found here confirms the correlation function that was used to fit the neutron data, and that the magnetic disorder can be described with a scaling, or fractal parameter of 0.5. In the special case of $h = 0.5$, the full width at half maximum of the diffuse peak in reciprocal space is simply $2/\xi_m$.[11] After magnetization along the hard axis, the in-plane correlation length was 880 ± 20 Å, compared to the much larger 7000 ± 100 Å length scale observed after magnetization along the easy axis. Both length scales are much smaller than the largest resolvable length scale, which is of the order of 5 μm.

As expected, no change was observed in the transverse scans taken through the structural Bragg peak upon field application. The peak shape, similar to the magnetic Bragg peak, corresponded to a correlation length of 48 Å. We note that, as was the case in our previous study on Cu/Co multilayers grown at the first AF coupling maximum, the length scale of the structural roughness is substantially shorter than that defining the magnetic interface.[7]

The resonant x-ray and neutron off-specular data show remarkably good agreement, however, the specular data are less consistent. Figure 4 shows the specular and longitudinal diffuse scans from a 20-period Co/Ru multilayer grown at the second AF coupling peak. The quality of the sample is high, with structural Bragg peaks being seen in the specular data out to many orders. Neutron data on similar samples show clear magnetic and structural Bragg peaks, but in the x-ray data the specular magnetic peaks cannot be seen, it is only in the diffuse scan that the magnetic peaks appear. Their magnetic origin is confirmed by their field sensitivity and energy resonance. The variation in the AF peaks with direction of magnetization is consistent with the predominant sensitivity of x-ray scattering to the component of magnetization in the scattering plane.[12]

We have successfully applied two different scattering mechanisms to the study of magnetic disorder in multilayer systems. The agreement in the off-specular data is extremely good for the Co/Cu system, but we find in the Co/Ru system that the two techniques do not give comparable results. The difference in sensitivity could arise from the different interaction potentials of the two probes, one being primarily sensitive to the point-like nuclei, and the other interacting with the electron density profile. However, the high flux of the x-ray source does allow for rapid determination of the element-specific interface morphological parameters, but how these data relate to the better-understood neutron interactions remains unclear.

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