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Polarized neutron reflectivity characterization of weakly coupled Co/Cu multilayers

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Abstract

Room temperature resistivity studies on (1 1 1) Co(6 nm)/Cu(6 nm) multilayers with weak interlayer coupling yield a giant magnetoresistance (GMR) of several percent for the as-prepared state relative to the saturated (i.e., large magnetic field) state. After application of a magnetic field, the magnetoresistance for the coercive state is only half to two-thirds as large. Using specular and off-specular polarized neutron reflectivity, we have determined the magnetic structure of these multilayers in the as-prepared and coercive states. Measurements of as-prepared samples show a strong antiparallel correlation of in-plane ferromagnetic Co domains across the Cu interlayers. At the coercive field, the Co domains are uncorrelated along the growth direction. Thus, the larger GMR for the as-prepared state arises from long-range antiparallel magnetic order along the growth-axis direction that is destroyed upon application of a magnetic field. For both the as-prepared and coercive states, the size of the in-plane ferromagnetic domains is approximately 0.5–1.5 μm. These domains give rise to pronounced diffuse scattering in rocking curves through the antiferromagnetic peak position.

Keywords: Giant magnetoresistance; Magnetic multilayer; Off-specular scattering; Magnetic domains; Polarized neutron reflectivity

Giant magnetoresistance (GMR) has been observed in magnetic multilayers [1–3] with alternating ferromagnetic and non-magnetic metallic layers. In these materials, a maximum in the magnetoresistance (MR), is associated with an antiparallel alignment of adjacent ferromagnetic layers. The resistance decreases when an external magnetic field aligns the magnetization of the ferromagnetic layers parallel to each other. In multilayers with thick non-magnetic layers ($t_n > 4$–5 nm), the interlayer magnetic coupling is weak. In this limit, the magnetoresistance, (MR(0)) for the as-prepared multilayer is often larger than the maximum value obtained at the coercive field MR(H_C) after saturation [4,5]. The initial MR(0) is not recovered after field cycling [6,7]. For proper analysis of MR data, it is important to know whether the magnetic order associated with MR(0) or MR(H_C) is closer to the antiparallel state.

Here we describe specular and diffuse polarized neutron reflectivity (PNR) studies of Co/Cu multilayers with $t_{Cu} = 6$ nm. In the as-prepared state, we observe a distinct magnetic reflection at the half-order position. MR(0) thus originates from antiparallel correlations among the ferromagnetic Co layers. Application of a small magnetic field irreversibly destroys this reflection. We observe that the peak in the magnetoresistance at the coercive field MR(H_C) arises from a randomization of the Co layer domains along the growth direction. For both states, the presence of diffuse magnetic scattering reveals that the Co moments, which preferentially lie in the growth plane, are ordered in small domains with an average in-plane size of 0.5–1.5 μm. (This domain size has been confirmed by scanning electron microscopy with polarization analysis [SEMPA] measurements [5]). These results demonstrate that the combination of

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specular and diffuse PNR provides a full profile of the magnetization along the growth direction and within the sample plane.

For this study, we examined several multilayers of nominal composition $[Co(6\ nm)|Cu(6\ nm)]_N$ with $N = 10$ or 20 bilayers. The samples were sputtered onto Si substrates as described elsewhere [4]. The field dependence of the magnetization and magnetoresistance was measured at room temperature for comparable samples grown at the same time. The current-in-plane MR(0) was typically 4–7%, and MR(H_C) was half to two-thirds as large [4,5].

Fits to specular X-ray reflectivity data confirm that the Co and Cu interlayers are well modulated. Typical full-widths of the interfacial roughness (averaged over the sample plane) were ≈ 1.5 nm for our original samples, compared to 4.0 nm for samples that were prepared later. To characterize the structural disorder within the sample plane, we probed the diffuse scattering by performing rocking curves at various Q_z positions [8,9]. For samples with interfacial-roughness widths of 1.5 nm, the diffuse scattering is insignificant relative to the specular. Thus, the interfaces are relatively smooth, and the limited interfacial "mixing" is due to interlayer interdiffusion or atomic-scale roughness. In contrast, the samples with interfacial widths of 4.0 nm exhibit pronounced diffuse scattering. The measured interfacial widths probably originate from larger structures such as islands, corrugation or steps within the growth plane. In the rest of this paper we focus on a $[Co(6\ nm)|Cu(6\ nm)]_{20}$ multilayer that shows some diffuse scattering.

We performed the specular and diffuse PNR studies on the NG-1 reflectometer at the NIST Center for Neutron Research at room temperature. Fits to the specular data yield a profile of the magnetization as a function of depth for regions of the multilayer with ferromagnetic domains having in-plane dimensions larger than the coherence length of the neutrons, $\approx 100\ \mu m$ [10]. Diffuse measurements are sensitive to the in-plane length scale of the structural and magnetic roughness [8,9,11] and of the magnetic domains [11]. They also provide information about the interlayer correlations among ferromagnetic domains having in-plane dimensions smaller than $\approx 100\ \mu m$. For specular and diffuse experiments, we measured all four cross sections, $(--)$, $(++)$, $(+-)$ and $(-+)$. (The $+$ and $-$ signs indicate polarizations of the incident and scattered neutrons parallel or antiparallel to the guide field.) The $(--)$ and $(++)$ non-spin-flip (NSF) data depend on the chemical structure, as well as the projection of the in-plane magnetization parallel to the guide field. The $(+-)$ and $(-+)$ spin-flip (SF) cross sections originate from the projection of the in-plane magnetization perpendicular to the field [10]. The instrumental polarization efficiency is $>95\%$.

We first characterized the magnetic structure of each sample in the as-prepared state. Fig. 1 shows specular

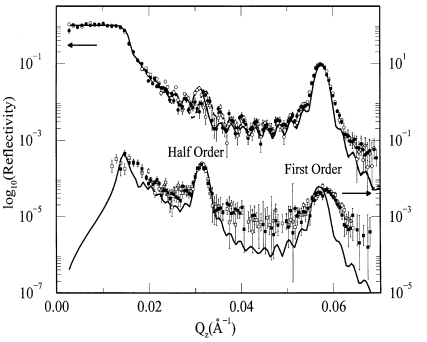

Fig. 1. Specular PNR as a function of $Q_z = (4\pi/\lambda)\sin\theta$ for $[Co(6\ nm)|Cu(6\ nm)]_{20}$ in the as-prepared state in a 1.5 Oe field. The diffuse scattering has been subtracted, and the data have been corrected for the polarization efficiencies. The open and shaded circles correspond to the $(--)$ and $(++)$ data respectively. The open and shaded squares designate the $(+-)$ and $(-+)$ data. The arrows point to the appropriate vertical axis for the NSF and SF data.

PNR scans[1] along the Q_z direction for the $[Co(6\ nm)|Cu(6\ nm)]_{20}$ sample. As expected, the NSF cross sections show a first-order structural superlattice peak at $Q_z = 0.057\ \text{Å}^{-1} \approx 2\pi/d$ where $d = 11.4$ nm is the bilayer repeat distance. A pronounced peak is also evident at the half-order position ($Q_z = 0.031\ \text{Å}^{-1} \approx 2\pi/2d$). A fit to the data indicates that $\approx 1.5\%$ of the Co moments are ordered in large ferromagnetic domains (in-plane size $> 100\ \mu m$) that are aligned antiparallel across the Cu. The orientation of these domains in the sample plane is likely random since the magnetic intensity is evenly distributed in all four cross sections. The magnetic SF scattering at the first-order position indicates that at least an additional 2.5% of the Co moments are aligned parallel across the Cu. Along the growth axis, sets of distinct domains aligned parallel and antiparallel may coexist or there may be a collection of non-collinear domains [12]. The latter possibility is unlikely because the first-order SF magnetic peak is broader than the half-order peak, suggesting a difference in the growth-axis coherence length of the parallel and antiparallel magnetic structures. (We note that the coherence of the parallel magnetic structure is also shorter than the structural coherence length as indicated by the difference between the full-widths of the SF and NSF first-order reflections).

This analysis of the specular PNR data accounts for only about 4% of the total Co magnetization. Because specular reflectivity averages over features with in-plane

[1] Diffuse data have been subtracted from the total reflectivity to give the specular reflectivity. This is exact only if the sample has regions with large and small domains that scatter respectively in the specular and diffuse.

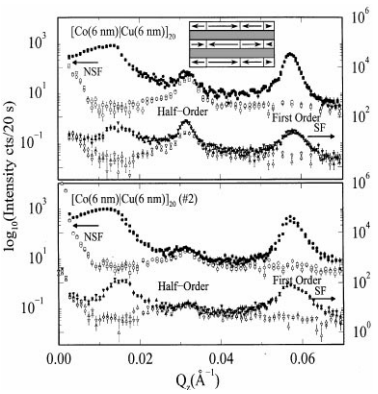

Fig. 2. Total PNR (shaded symbols) relative to the diffuse scattering (open symbols) for $[Co(6\ nm)|Cu(6\ nm)]_{20}$ and $[Co(6\ nm)|Cu(6\ nm)]_{20}$ (#2) in the as-prepared state. The diffuse scattering was measured by offsetting the angle θ by a small amount and then scanning Q_z. The circles and squares correspond to $(--)$ and $(++)$ data, respectively. The up and down triangles mark $(+-)$ and $(-+)$. No data corrections have been made. The arrows designate the vertical axis for each cross section. The inset shows the magnetic structure.

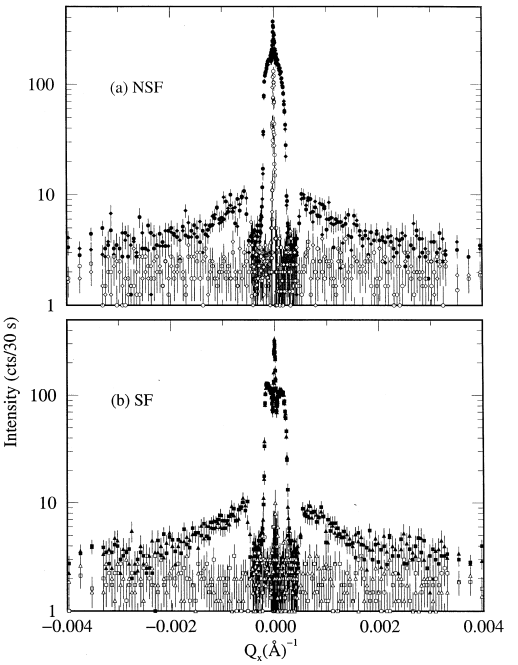

Fig. 3. Transverse Q_x scans at the half-order position ($Q_z = 0.314$ Å^{-1}) for $[Co(6\ nm)|Cu(6\ nm)]_{20}$ in the as-prepared (shaded symbols) and saturated ($H = 400$ Oe) states (open symbols). The $(++)$ and $(--)$ NSF cross sections (circles and diamonds respectively) are shown in (a) and the $(-+)$ and $(+-)$ SF cross sections (squares and triangles) are shown in (b).

dimensions less than ≈ 100 μm, the remaining moments appear to be ordered in small, discrete domains spread over the growth plane.[2] For the as-prepared state, a comparison of the diffuse scattering (i.e., Q_z scan with θ offset by 0.1°) to the total reflectivity [Fig. 2(a)] shows that the total scattering is dominated by diffuse at the half-order position. These results indicate that most of the small domains in these samples are oriented antiparallel [inset Fig. 2(a)] along the growth direction. The narrow Q_z width of the diffuse reflection indicates that the antiparallel order is correlated through the entire multilayer thickness. For a second $[Co(6\ nm)|Cu\ (6\ nm)]_{20}$ sample, the half-order reflection shown in Fig. 2(b) is entirely of diffuse character, and the Q_z width is greater. We note that the data for most of our samples resembled those in Fig. 2(b). In these multilayers the growth-axis coherence is limited to a few bilayers. The as-prepared state responsible for the maximum GMR is thus dominated by small, ferromagnetic Co domains that are aligned antiparallel across the Cu layers.

Consistent with the magnetoresistance data, this antiparallel order is irreversibly destroyed by the application of a field. After saturation in a large field, the half-order reflection in the PNR data for all of the samples considered is absent at the coercive field ($H_C \approx 50$ Oe) in both the total reflectivity and diffuse data. No other magnetic feature is evident. In the coercive state, the Co moments in all of the samples seem thus to order in domains with small in-plane dimensions (<100 μm) that are randomly oriented across the Cu layers [5]. As a consequence, the GMR is not as large as that measured for the as-prepared antiparallel state. This irreversible loss of antiparallel magnetic order is analogous to the magnetic structure reported for weakly coupled Fe/Nb multilayers [13].

In the as-prepared and coercive magnetic states, additional details of the in-plane magnetic structure can be gained from transverse Q_x scans centered at the half-order position ($Q_z = 0.0314$ Å^{-1}). Fig. 3(a) and (b) show NSF and SF data for $[Co(6\ nm)|Cu(6\ nm)]_{20}$ in the as-prepared and saturated states. Pronounced dips are centered at the sample angles $\Omega = 0$ and $\Omega = 2\theta$ where either the incident or scattered beam is parallel to the sample face and is thus refracted away from the detector [14]. The as-prepared data in Fig. 3 are otherwise composed of a sharp specular reflection at $Q_x = 0$ Å^{-1} on top of a broad, diffuse peak. The coercive state data look similar to the as-prepared data [5]. We conclude that the diffuse scattering in the as-prepared and coercive states originates entirely from magnetic features within the sample plane because it disappears when the Co moments are aligned in a saturating field of 400 Oe (Fig. 3). In addition, much of the diffuse scattering is SF, which has no chemical contributions.

[2] Specular PNR is also insensitive to spins perpendicular to the growth plane, but magnetization measurements indicate that the Co moments lie in-plane.

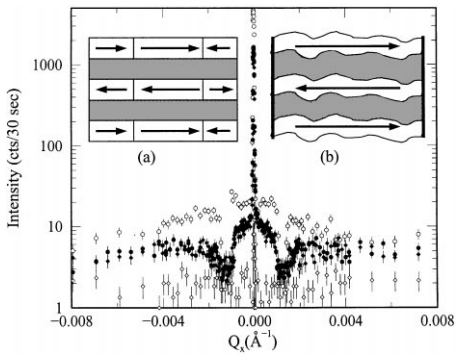

Fig. 4. Transverse Q_x scans at the first-order position ($Q_z = 0.575$ Å^{-1}) for $[\mathrm{Co(6\ nm)|Cu(6\ nm)}]_{20}$ in the as-prepared state (shaded symbols) relative to the saturated state (open symbols) in $H = 400$ Oe. Only the NSF cross sections are shown. The 400 Oe ($--$) data (circles) and ($++$) data (squares) are split because the Co moments are aligned parallel to the field. Insets (a) and (b) show multilayers with in-plane domains and magnetic roughness, respectively.

An estimate of the in-plane magnetic correlation length can be obtained from the transverse full-width of the half-order diffuse peak. This value corresponds either to the size of the ferromagnetic domains and/or to the correlation length of the magnetic roughness [11]. Lorentz fits of as-prepared and coercive data similar to those shown in Fig. 3(a) and (b) give correlation lengths of 0.5–1.5 μm. (A direct analysis of the transverse data using a kinematic or dynamical formalism [8,9,11] would yield a more accurate estimate, but this work is still in progress.) SEMPA imaging of the first and second Co layers in the $[\mathrm{Co(6\ nm)|Cu(6\ nm)}]_{20}$ show clearly that the Co moments are ordered in irregularly shaped micron-sized domains for the as-prepared state [5]. The observation of strong magnetic diffuse scattering in the PNR data (Fig. 3) indicates that these domains are distributed throughout the entire sample.

Transverse scans through the first-order Q_z position provide information about other in-plane magnetic structures that may contribute to the diffuse magnetic scattering. By comparing the field dependence of the diffuse peak at first- and half-order, we can differentiate between in-plane domain formation and magnetic roughness (i.e., structural roughness between the Cu and ferromagnetic Co layers). If the Co moments order completely in small domains at low fields [Fig. 4 inset (a)], any diffuse intensity at the half- and first-order positions should disappear when the Co domains are aligned parallel in a saturating field. If instead the interfaces are rough across the growth plane [Fig. 4 inset (b)], any low-field diffuse scattering at half-order should transfer to the first-order position as the Co spins align in high-fields [11]. In fact, the NSF ($--$) diffuse scattering for the $[\mathrm{Co(6\ nm)|Cu(6\ nm)}]_{20}$ sample at the first-order position does increase in a 400 Oe field, as shown in Fig. 4. At least part of the diffuse intensity for this sample can thus be attributed to magnetic interfacial roughness. This result agrees with the observation of structural diffuse scattering in transverse X-ray scans. However, other samples do not show substantial chemical roughness within the sample plane, and the diffuse magnetic scattering at the first-order position subsequently disappears in high fields. Most of the magnetic diffuse scattering from our samples is apparently characteristic of the micron-size in-plane magnetic domains directly observed by SEMPA [5].

Our PNR measurements have shown that the larger MR(0) for as-prepared $[\mathrm{Co(6\ nm)|Cu(6\ nm)}]_N$ multilayers arises from antiparallel ordering between ferromagnetic Co domains across the Cu layers. This antiparallel state occurs only after growth and cannot be restored upon application of a field. The smaller MR(H_C) at the coercive field is associated with random Co domains. In both the as-prepared and coercive states, the typical in-plane domain size is ≈ 0.5–1.5 μm as determined from the full-width of the magnetic diffuse scattering. Our measurements of the magnetic diffuse scattering demonstrate the power of PNR as a probe of domains in buried magnetic layers and are a complement to other neutron and X-ray resonant scattering results for related thin film systems [15–18].

Acknowledgements

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