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A neutron study of magnetic domain correlations in antiferromagnetically coupled multilayers

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The observed magnetotransport properties of magnetically coupled multilayers depends not only on the nature of the magnetic coupling but also the magnetic domain correlations and disorder. Neutron scattering gives access to the magnetic coupling through the specular reflectivity and the domain correlations through the diffuse scattering. Sputtered multilayers of Co/Cu and Co/Ru have been investigated as a function of the applied magnetic field. A simple domain model relates the observed scattering to the domain correlation length and the magnetic disorder. In both systems highly vertically correlated magnetic domains are observed with in-plane correlation lengths, at remanence, of 1.5 and 7 μm for the Cu and Ru systems, respectively. In both systems the Co domains order antiferromagnetically across the nonmagnetic spacer. The remanent vertically correlated state is recovered after saturating the sample. © *2000 American Institute of Physics.*

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I. INTRODUCTION

Following the observation of the giant magnetoresistance effect (GMR) in 1988[1] there have been considerable advances in understanding the origins of the change in resistivity arising from the transition from antiferromagnetic (AF) coupling to ferromagnetic (*F*) coupling between ferromagnetic layers (typically $3d$ metals) separated by a nonmagnetic spacer. To fully understand the observed GMR knowledge of both the interlayer magnetic coupling and the in-plane magnetic domains is required. For thin film and multilayer structures, polarized neutron reflectivity (PNR)[2] allows an *absolute* measure of the *vector* in-plane magnetization with a depth dependent sensitivity (normal to the sample surface) and is ideally suited to studying magnetic interlayer coupling.[3] Until recently in-plane inhomogeneities such as magnetic domains have not been accessible through neutron reflectivity measurements due to the weak nature of the scattering relative to the specular reflectivity and the flux limited nature of the neutron technique. This is not the case for synchrotron x-ray sources and studies of structurally rough interfaces are well advanced in both experimental and theoretical studies.[4,5] By using the huge photon flux and the resonant enhancement in the x-ray magnetic scattering cross section, magnetically rough ferromagnetic thin films have been studied.[6,7] The neutron technique[8] complements these measurements in three important aspects. First, the much larger lateral coherence length of the neutron beam (~30 μm) ensures measurements sample many magnetic domains even when the domain size becomes large. Second, the direct nature of the neutron-magnetization density interaction is well understood and allows a simple connection between the neutron observations and the in-plane magnetic disorder as outlined in Sec. III. Third, the low neutron absorption cross section ensures that the whole multilayer system is sampled. In this article we compare results from Co/Cu and the more strongly coupled system Co/Ru and present clear evidence for a remanent field vertically correlated domain structure that is not destroyed by magnetizing and demagnetizing the system.

II. EXPERIMENT

Samples of Co/Cu and Co/Ru were prepared with spacer thicknesses corresponding to the second AF maxima of the coupling oscillation. The samples were deposited by dc magnetron sputtering in a custom vacuum system with a base pressure of 2×10^{-8} Torr. The multilayers were grown on Si (001) wafers without removing the native oxide layer. To optimize the neutron reflectivity signal while maintaining a good bilayer thickness homogeneity, the largest attainable sample size was 25×20 mm. For perfect conformal roughness (the roughness profile is correlated between interfaces) the diffuse intensity is the product of the perfect multilayer reflectivity and the scattering from a single rough surface and so to maximize the scattering from any conformal roughness 50 bilayer repeats were deposited. Smaller samples were grown in the same run for magnetotransport measurements. PNR measurements were made at the Rutherford Appleton Laboratory on the polarized beam time of flight reflectometer, CRISP.[9,10] By utilizing the incident wavelength range (1.2–6.5 Å) and a one dimensional detector, a large region of reciprocal space can be accessed in a single measurement (see Fig. 1). For the diffuse scattering measurements presented, the reflectometer was run in a nonpolarized mode to increase the incident sample neutron flux. The reciprocal space maps are acquired in typically 2 h.

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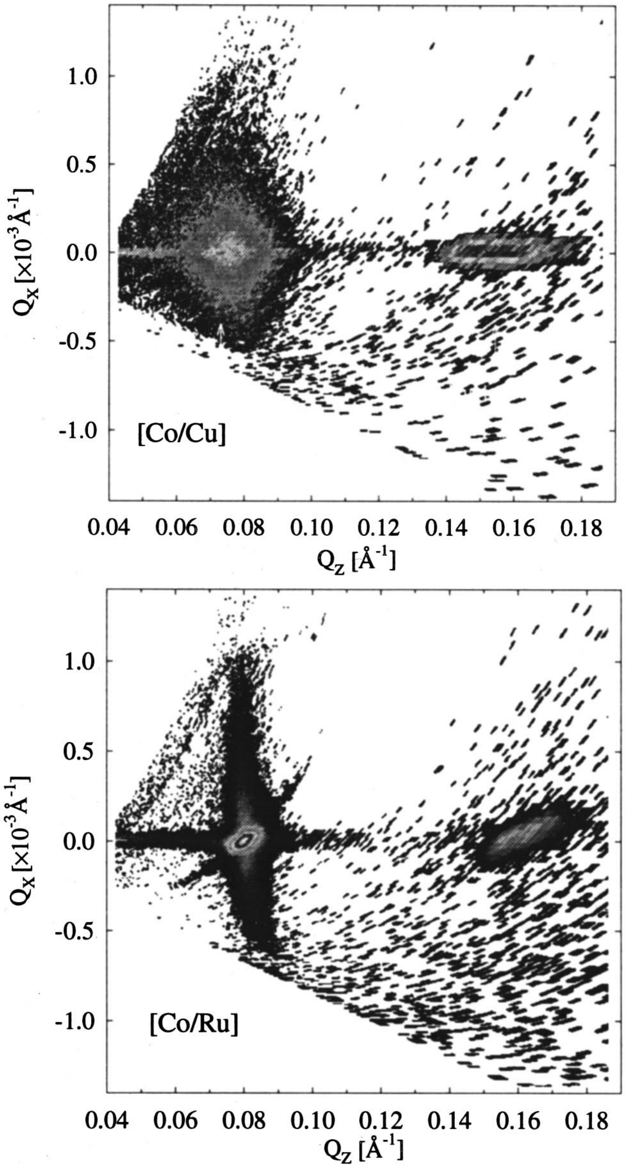

FIG. 1. The observed neutron scattering from a nominal sample of $[(\mathrm{Co}(20\,\text{Å})/\mathrm{Cu}(22\,\text{Å})]_{50}$ and $[(\mathrm{Co}(20\,\text{Å})/\mathrm{Ru}(18\,\text{Å})]_{50}$ at remanence. The cutoffs evident in the data represent the kinematical limits in the measurement.

III. RESULTS AND DISCUSSION

Figure 1 shows the scattering observed for samples with a nominal structure of $[(\mathrm{Co}(20\,\text{Å})/\mathrm{Cu}(22\,\text{Å})]_{50}$ and $[(\mathrm{Co}(20\,\text{Å})/\mathrm{Ru}(18\,\text{Å})]_{50}$ at remanence. The specular reflectivity corresponds to a section along the longitudinal wave vector transfer Q_Z at zero perpendicular wave vector transfer $Q_X=0$. The nuclear first order Bragg peak is clearly visible and does not show any significant scattering away from the specular direction. The peak at half this wave vector (twice the real-space bilayer repeat) indicates that both samples are AF coupled. The narrow width in Q_Z implies that the AF ordering is vertically coherent throughout the multilayer. Concentrating on the AF Bragg peaks, there are clear differences between the Cu and Ru spacers. For the Cu system the magnetic and structural vertical coherence lengths are ~600 Å. For the Ru system the structural coherence length is again ~600 Å while the magnetic vertical coherence length is ~2000 Å, the total film thickness. The AF peak has significant diffuse scattering, manifested as a stripe in Q_X at the AF ordering wave vector. This scattering is entirely magnetic in origin and we attribute it to magnetic domain formation within the Co layers, as discussed below. The stripe of scattering implies a coherent interference of the scattered neutrons such as would be produced by a vertically coherent domain structure analogous to a structural conformal roughness. A detailed search was made for nonconformal roughness which would be manifested as intensity uniformly distributed in the Q_X,Q_Z plane. No intensity was observed. This is not surprising given that for an equivalent conformal and nonconformal roughness we expect the nonconformal intensity to be reduced relative to the conformal intensity by a factor of 50.[11] In addition, the Ru data show a weak diagonal stripe at the AF ordering wave vector. This scattering may arise from a magnetic analog of ''Newton's fringes.''[12] To extract quantitative information the sections through the AF Bragg peaks are presented in Fig. 2. The scattering from the Co/Cu is dominated by the broad diffuse component with little evidence for a specular component to the scattering. Conversely the Co/Ru data are dominated by the specular scattering with a weaker diffuse component.

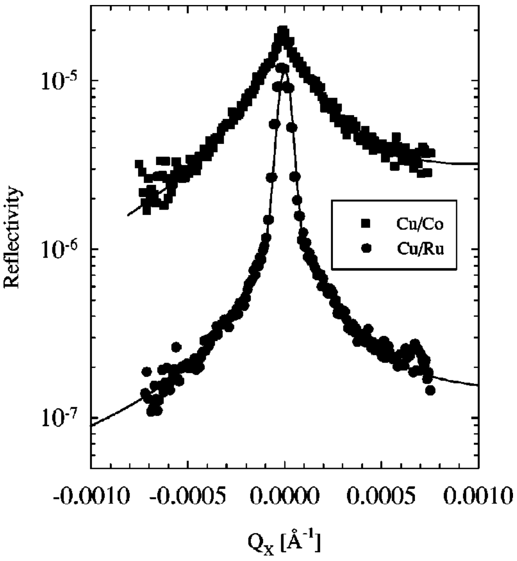

FIG. 2. Transverse momentum transfer cuts through the AF peak of the Co/Cu and Co/Ru samples at remanence. The solid lines are least squares refinements to the data.

To understand these results we consider a simple model in which the Co layers consist of in-plane magnetic domains, the lateral extent of which we associate with a correlation length ξ and in which the distribution of the individual domain magnetization vectors around the applied field direction is treated as a random variable with a Gaussian distribution. This is analogous to the Gaussian height distribution employed in the analysis of structural roughness.[4] Given that the scattering is in the weakly interacting regime, we can consider the interpretation within the Born approximation, significantly reducing the complexity of the calculation which will be discussed elsewhere.[13] The solid lines in Fig. 2 are least square refinements of this model to the data. The analysis of the Co/Cu data indicate an average domain size at

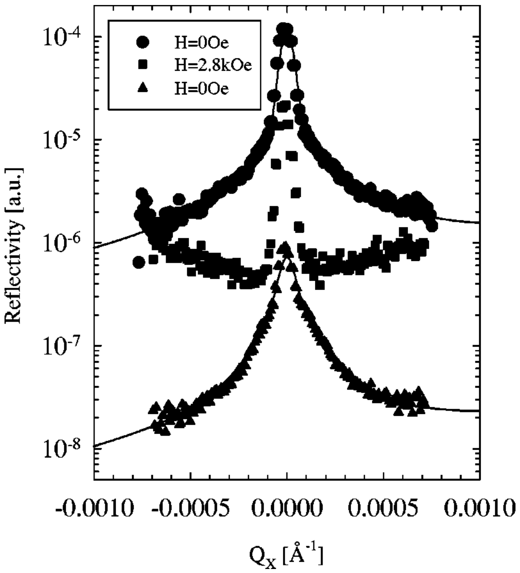

FIG. 3. Transverse momentum transfer cuts through the AF peak from the Co/Ru sample at remanence (circles), close to saturation and back to remanence (triangles). The hysteresis is clearly visible in the data.

remanence of $1.5 \pm 0.4\,\mu$m and to within the experimental resolution the magnetic domain magnetization vectors are randomly distributed. For the more strongly coupled Ru the correlation length is $7 \pm 3\ \mu$m. The domain distribution is no longer random but corresponds to Gaussian distribution with a full width at half maximum of $200° \pm 40°$.

Figure 3 presents the remanent field data, at an applied magnetic field of 2.8 kOe and back at remanence. As the multilayer approaches saturation the diffuse scattering collapses and only the nuclear specular ridge of scattering remains. We can understand this result in terms of changes to the domain distribution. As the applied field is increased, the domain size increases and the domain magnetization vector angular distribution focuses around the applied field direction. This leads to a reduction in the intensity of the magnetic diffuse component and also the specular AF Bragg peak. At saturation the ferromagnetic alignment results in additional scattering at the first order Bragg peak. It is interesting to note that when the sample has a large ferromagnetic component, no diffuse scattering (either structural or magnetic) is observed around the first order Bragg peak. This is in contrast to samples produced at the first ferromagnetic coupling peak which show clear magnetic diffuse scattering around the first order Bragg peak.[13] This observation gives some insight into the nature of the diffuse scattering. The scattering from either uniformly magnetized layers with structurally rough interfaces or layers with domain formation both give rise to a stripe of diffuse intensity at the AF Bragg peak.[14] The total structural rms roughness of our samples (correlated and uncorrelated) as determined by x-ray reflectivity is ~5 Å. This is not observable within our neutron data and as such the absence of diffuse scattering at the first order Bragg peak when the magnetic coupling is ferromagnetic (or for fields between remanence and saturation) suggests that the dominant mechanism for the diffuse scattering is the magnetic domain structure, not structurally rough interfaces. In fact, Lorentz transmission electron microscopy measurements reveal direct evidence of domain formation.[15] A description of the field dependence of the diffuse scattering will be postponed.[13] However we note that in both the Cu and Ru systems a comparison of the as-prepared and coercive states both show vertically correlated Co domains in contrast to the weakly coupled Co/Cu system (with a much thicker Cu layer to reduce coupling to a minimum), where a loss of coherence has been used to explain the reduction in GMR between these magnetic states.[16]

IV. CONCLUSIONS

We have studied the field dependence of the magnetic domain distribution in Cu/Co and Ru/Co multilayers. A high degree of vertical coherence is observed for the Co domains. A simple model allows us to relate the diffusely scattered neutron intensity to the domain distribution. Our results suggest that it is the in-plane domain distribution rather than uniformly magnetized layers with structurally rough interfaces thats gives rise to the diffuse intensity. The increased coupling strength of the Ru system (saturation field $H \sim 3000$ Oe) relative to the Cu system ($H \sim 300$ Oe) gives rise to a nonrandom domain distribution with a large in-plane correlation length and a vertical correlation length greater than the structural coherence and equivalent to the multilayer thickness.

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