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Understanding correlations between chemical and magnetic interfacial roughness

J. W. Freeland
Experimental Facilities Division, Argonne National Laboratory, Argonne, Illinois 60439

K. Bussmann and Y. U. Idzerda
Materials Science Division, Naval Research Laboratory, Washington, DC 20375

C.-C. Kao
National Synchrotron Light Source, Brookhaven National Laboratory, Upton, New York 11973

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By studying the variation in magnetic interfacial structure using diffuse x-ray resonant magnetic scattering, underlying connections between the chemical and magnetic interfaces are uncovered and utilized to understand how intrinsic magnetic properties influence the behavior of magnetic interface. By considering the effect of competing magnetic energies (dipolar, anisotropy, and exchange) at the rough interface, it becomes clear how magnetic forces can lead to a reduction of the magnetic interfacial disorder. [S0163-1829(99)51638-X]

With the rapidly expanding number of applications for magnetic thin-films structures,[1] a proper understanding of the connection between magnetic properties and interfacial disorder is extremely important. In spin transport effects, such as giant magnetoresistance (GMR),[2] roughness alters the amount of spin-dependent scattering events causing a modification of the observed GMR.[3–6] In the case of exchange bias[7] there is evidence that in some systems the underlying physical mechanism is related to uncompensated spins present at the ferromagnetic-antiferromagnetic interface due to roughness.[8] In both cases, it is clear that the important factor is not the chemical disorder present at the interface but rather the magnetic disorder.

Evidence for differing chemical and magnetic interfaces was first observed qualitatively with neutron scattering, where the damping of the reflectivity was found to be smaller for the magnetic scattering than for the chemical scattering,[9,10] indicating a smaller magnetic interfacial disorder. Using the strongly enhanced magnetic component due to soft x-ray resonant magnetic scattering (XRMS),[11–15] similar qualitative evidence of smaller magnetic roughness was also seen using specular XRMS.[16] Quantitative determinations of the magnetic roughness were recently achieved with measurements of soft x-ray diffuse XRMS,[17,18] but due to a lack of theory the analyzed magnetic roughness was not properly extracted and in the end was overvalued. In this paper, proper extractions of magnetic roughness are presented and used to uncover correlations between the chemical and magnetic interfaces that can be modeled to give insight into how the magnetic disorder is related to intrinsic structural and magnetic properties.

In order to study the relationships between magnetic and chemical interfaces, two sets of CoFe alloy thin films were utilized in which chemical roughness could be accurately tailored. The samples were prepared by triode sputtering on Si substrates coated with 2500 Å of atomically flat insulating Si_3N_4 (roughness ~1.5 Å rms)[19] with the following structure: Cu(30 Å)/$Co_{95}Fe_5$(50 Å)/Cu(x Å)/Si_3N_4. Due to growth dynamics, the rms roughness of Cu grown on Si_3N_4 increases dramatically as the thickness of the Cu buffer layer, x, is increased. In both sets, the perpendicular roughness of the CoFe layer showed the same variation from 4 to 28 Å rms as the Cu buffer thickness ranged from 200 to 1600 Å,[18] but differing growth processes resulted in a different range of grain sizes.[20]

The interface structure was probed by soft x-ray reflectivity measurements conducted at the NRL/NSLS magnetic circular dichroism facility[16] located at beamline U4B of the National Synchrotron Light Source. For these measurements the degree of circular polarization was set to 75% and, since the resonant scattering can vary significantly with only a few eV change in incident photon energy,[12,13] the energy resolution was 0.8 eV. The sample was mounted in the gap of an electromagnet inside a vacuum-compatible $\theta - 2\theta$ diffractometer that has been described elsewhere.[21] Samples were oriented with the field along the magnetic easy axis, and data were taken in the presence of a ± 100 G field, which was sufficient to achieve magnetic saturation. All measurements were taken at a photon energy equivalent to the Co L_3 absorption edge (778 eV), which, in addition to making this measurement element specific, leads to strong attenuation of x-rays in the CoFe layer[22] and introduces a depth selectivity. For these samples, the measurements were made at grazing incidence ($\omega = 0 \rightarrow 10^0$) and thus probe predominately the top CoFe/Cu interface.

The magnetic and chemical roughness information in these structures was quantitatively determined through helicity-dependent sample rocking curves (see inset Fig. 1). In this geometry, the detector is placed at a fixed angle 2θ and the incident angle, ω, is varied from 0 to 2θ. In terms of the transferred wave vector ($\vec{q} = \vec{k}_{Out} - \vec{k}_{In}$), at these photon wavelengths, the normal component, q_z, remains approximately constant while the in-plane component, q_x, varies. Figure 1 shows the results for the measurements taken with an incident light of negative helicity and with the sample magnetically saturated so that the helicity and magnetic moment are aligned (I^+) and antialigned (I^-). Of interest to the study at hand is the separation and correlation of the

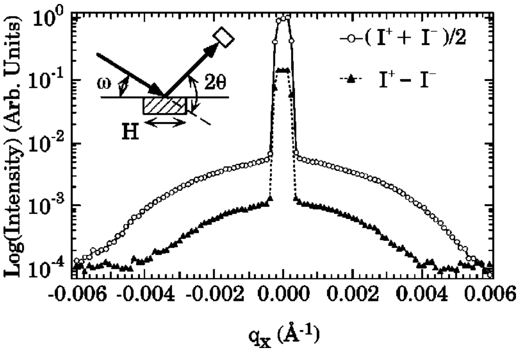

FIG. 1. The sum $(I^+ + I^-)$ and difference $(I^+ - I^-)$ rocking curves constructed from the helicity-dependent data measured at the Co L_3 edge (778 eV) with $q_z = 0.0686$ Å $(2\theta = 10)$. The inset displays the measurement geometry.

chemical and magnetic contributions of the helicity-dependent scattering. As will be seen below, although the average of the two scattering intensities $(I^+ + I^-)/2$ gives the chemical scattering, the difference $(I^+ - I^-)$ is not purely magnetic in origin.[23] From these scans, the roughness perpendicular to the plane of the film, σ, is extracted from the specular vs diffuse intensities, while the in-plane correlation length, ξ, is extracted from the half width of the diffuse portion.[18,24]

To properly understand and analyze the scattering from a rough magnetic interface, it is necessary to utilize a theory of scattering from a rough surface for the resonant magnetic case (i.e., off-diagonal terms in the dielectric tensor),[23] which for this study involves a longitudinal scattering geometry. This result provides a form for the scattering factor that can be utilized to determine the proper extraction of the parameters describing the magnetic disorder. The first result of this theory is that the difference measurement, $\Delta I = I^+ - I^-$, is not purely magnetic in nature and contains both chemical and magnetic contributions. Within this theory the scattered diffuse intensity is given by

$$\Delta I(\vec{q}) \sim \frac{A(E)}{q_z^2}[\cos(2\theta-\omega)+\cos(\omega)\cos(2\theta)]e^{-q_z^2(\sigma_c^2+\sigma_m^2)/2} \times \int_c \int_m dx\,dy\,(e^{q_z^2 C_{cm}(x,y)}-1)e^{-i\vec{q}_{\|}\cdot\vec{\rho}}, \qquad (1)$$

where $A(E)$ is related to the energy dependent scattering factors, σ_c and σ_m denote chemical and magnetic roughness, c and m represent integration over the chemical and magnetic surfaces $[z(x,y)]$, $C_{cm}(x,y)$ is the chemical-magnetic correlation function defined as $\langle z_m(x,y) z_c(0,0)\rangle$ (see Fig. 2), and $\vec{\rho}$ is a distance vector in the plane of the film. $C_{cm}(x,y)$ is an important factor in the scattering because it indicates that only if the chemical and magnetic interfaces are correlated do they give rise to diffuse intensity in the ΔI measurement. Because the chemical roughness (σ_c) is already known from the $(I^+ + I^-)$ measurement, the magnetic roughness (σ_m) can be extracted from the integrated difference spectra.

To illustrate correlations between the magnetic and chemical perpendicular roughness, σ_m is plotted as a function of σ_c in Fig. 3. On this plot in which σ_c is a line of slope 1, the magnetic roughness on average clearly has a distinctly smaller slope (0.73) indicating that, as the interface becomes chemically rougher, the magnetic interface tends to smooth more significantly. To understand this connection and uncover what factors might be important in influencing the magnetic interface, consider how the roughness might be connected to the magnetic energies in the system. The presence of magnetic interfacial spins adds a new dimensionality to the problem because moments are vector quantities, which can possess disorder both in magnitude and orientation; so it is first worthwhile to discuss the meaning of magnetic roughness.

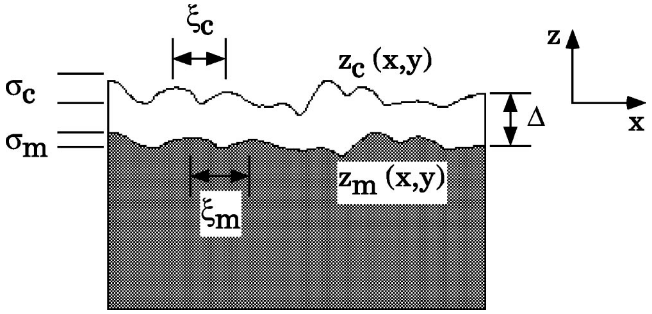

FIG. 2. Diagram of chemical and magnetic interface that gives rise to the diffuse scattering in Eq. (1). The quantities in the figure are defined in the text.

In the end, all of the interfacial disorder probed by the scattering is contained in the structure of the interface, $z_m(x,y)$ (see Fig. 2), where the $\sigma_m^2 = \langle z_m(x,y)^2\rangle$ and ξ_m is determined by the length scale of magnetic correlations in the plane. However, magnetic scattering is related not only to the magnitude of magnetic moment but also to $\vec{\Sigma}\cdot\vec{M}$, where Σ is the photon helicity and $\vec{M}$ is the magnetic moment, which implies there are orientational as well as spatial contributions to the shape of the magnetic interface. Unfortunately the theory above deals only with a scalar quantity, $z_m(x,y)$, and is not capable of calculating the scattering from a distribution of $\vec{M}$ directions.[25] It is also worth noting that since the measurement of magnetic roughness requires the reorientation of the magnetic moment, if some spins always remain fixed they are not included in the ΔI measurement. However, in the CoFe system the exchange cost is significantly large so it would be difficult for interfacial spins to

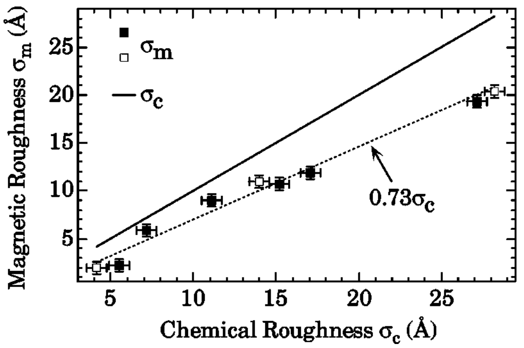

FIG. 3. Magnetic perpendicular roughness, σ_m, as a function of chemical roughness, σ_c. The linear fit (dashed line) demonstrates how the magnetic interface tends to smooth as the chemical interface grows rougher (solid line). Open and closed symbols represent the two separate sets of CoFe thin films described in the text.

remain fixed upon reversal of the bulk moment. Because in this system the interfacial roughness is due to the underlying substrate and not to intermixing at the interface,[26] very little quenching of the moment at the boundary should occur and the disorder can be considered as a first approximation as being purely orientational in origin. Roughness in terms of interfacial spins would indicate that aligned spins correspond to less disorder and would lower the values for the roughness.

With this in mind, consider two limiting cases within a simple model that can aid in understanding how magnetic energies can alter the magnetic interface: 1) The spins are all aligned with the bulk moment satisfying exchange and anisotropy, but induced magnetic charge at the rough interface results in a dipolar energy contribution; 2) this case can quench this induced surface charge by randomizing the spin directions at the expense of exchange and anisotropy. In this way the dipolar energy will compete with the exchange and anistropy to determine the lowest energy configuration of the interfacial spins. This implies that material parameters, such as exchange strength, A, anisotropy, K, and the saturation moment, M_s, should be important parameters in determining σ_m. The exchange cost will always be the dominant energy in the system and will force an alignment or partial alignment of the spins resulting in a decreasing disorder. The relation between σ_m and σ_c stems from the fact that increasing chemcial disorder will result directly in an increase in surface charge that must be compensated to balance the total energy of the system.

Utilizing this simple idea, one can consider general inferences concerning the connection between the magnetic and chemical disorder. Even if the chemical interface remains constant, altering the material parameters (A, K, M_s, etc.) either through the environmental conditions (e.g., temperature) or types of materials will result in a variation in σ_m. As a conceptual example consider the comparison of Co and Ni interfaces that have identical chemical interfaces. If we assume that Co and Ni possess the same in-plane anisotropy, the lower Ni exchange constant should cause the Ni interface to be magnetically rougher than that of the Co. This demonstrates clearly the assertion that characterization of only the chemical interface is insufficient to understand fully the influence of disorder on the magnetic properties.

In the case of in-plane magnetic disorder, extraction of the true magnetic correlation length, ξ_m, is difficult due to the mixture of chemical and magnetic scattering,[23] but differences between the chemical and chemical-magnetic ξ represent differences caused by the magnetic interface. Because interest lies in the connection between the chemical and magnetic disorder, consider ξ_{cm} as a function of ξ_c (see Fig. 4), extracted from the width of the diffuse portion of the sum and difference measurements.[18] On average, the two correlation lengths track together with a constant separation. Because ξ_c gives a measure of the grain size, the enhanced ξ_{cm} can be understood from the point that the ferromagnetic interaction extends beyond the grain boundary, which is expected since the grains are magnetically interacting. If the length scale of the magnetic coupling is independent of roughness, then ξ_m will track directly with the grain size. With this picture, the behavior of the data can be matched quite well by a line representing $\xi_c + 260$ Å (the dashed line in Fig. 4). In reality, because ξ_{cm} represents a combination of the true magnetic and chemical correlation lengths, the true difference will correspond to a larger value (~500 Å), which is of the order of the ferromagnetic interaction length.[27]

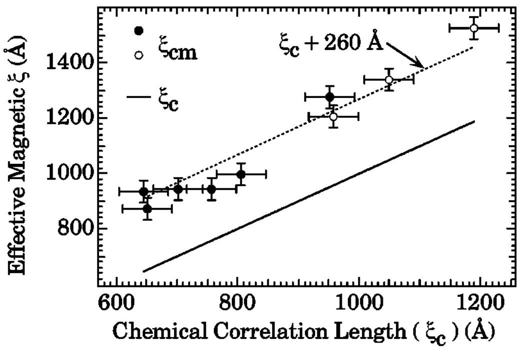

FIG. 4. Chemical-magnetic ξ_{cm} as a function of the chemical correlation length, ξ_c. The magnetic in-plane roughness tends to track with ξ_c (solid line) as indicated by the dashed line representing $\xi_c + 260$ Å. The open and closed symbols represent the two separate sets of CoFe thin films described in the text.

In conclusion, this work has illustrated the connection between the disorder for the chemical and magnetic interfaces and given insight into how magnetic energies can influence the magnetic interfacial disorder. Such a connection opens the door to a better understanding of how interfacial disorder is connected to intrinsic magnetic quantities.

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