Element Specific Magnetization of Buried Interfaces Probed by Diffuse X-Ray Resonant Magnetic Scattering

J. F. MacKay, C. Teichert, D. E. Savage, and M. G. Lagally
University of Wisconsin, Madison, Wisconsin 53706

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The magnetization of buried interfaces and its relationship to interfacial roughness is probed for Co films and Co/Cu multilayers using diffuse x-ray resonant magnetic scattering, a method in which the average diffusely scattered x-ray intensity is compared with the component that reflects magnetic scattering. The comparison demonstrates that the boundary between magnetic and nonmagnetic layers is smoother than the interfacial roughness, with short-wavelength roughness less effective in magnetic scattering than longer-wavelength roughness. [S0031-9007(96)01572-4]

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Much of the recent interest in thin-film magnetism is driven by the potential application of magnetic multilayers as magnetoresistive recording heads. Such multilayers are typically devised from alternating layers of a transition element (Fe, Co, Ni) and a noble-metal spacer layer (Ag, Au, Cu), These systems exhibit so-called giant magnetoresistance, associated with spin dependent scattering of conduction electrons from antiferromagnetically aligned magnetic layers [1]. Interlayer exchange coupling causes the magnetic layers to align antiferromagnetically for specific spacer layer thicknesses [2]. As with any multilayer structure, the quality of the interfaces between the different layers affects performance. For magnetic multilayers, interfacial roughness is of particular importance because it is believed to reduce the interlayer exchange coupling and thus the magnetoresistance [3,4].

Although the surface morphology of a multilayer may be probed with one of several imaging microscopies or spectroscopies, these techniques give only very indirect information about buried interfaces. Diffuse x-ray scattering can yield information on the roughness of buried interfaces [5,6]. Conventional x-ray scattering techniques, however, are not sensitive to the magnetic properties of a film, making it difficult to quantify the relationship between interfacial roughness, interlayer exchange coupling, and magnetoresistance. In recent years, x-ray resonant magnetic scattering (XRMS) has been developed to investigate the interaction of polarized x rays and magnetic materials [7,8]. XRMS is due to electronic transitions of inner-shell electrons into empty electronic states, with the magnetic field dependence of the scattered intensity arising from the spin-orbit interaction in the core level and the spin polarization of the conduction band [8]. Away from the absorption edge, pure magnetic scattering is observed but is very much weaker than the usual charge scattering [9]. Near the absorption edge of a magnetic element the magnetic-field dependence of the scattered intensity can be quite large [10–17]. By using circularly polarized photons tuned to an absorption edge of a magnetic material, the element specific resonant magnetic component of the scattered intensity may be extracted [10]. Although early XRMS measurements used Bragg reflections from the crystal lattice, XRMS is also observed in the specular reflection of soft x rays from a film surface [18,19].

To our knowledge, XRMS has not been used to investigate the diffuse intensity scattered away from the specular direction, in either a thin film or a multilayer. In this Letter, we show that the magnetization of buried interfaces can be probed directly by diffuse XRMS associated with the specular reflection from a magnetic film to provide new information that cannot b obtained in other ways. We measure the diffuse XRMS in Co films and Co/Cu/Co sandwiches using synchrotron radiation tuned to the L_3 absorption edge of Co. We compare diffuse XRMS to the average diffuse scattering, and demonstrate that the diffuse XRMS intensity displays a dependence on the length scale of the interfacial roughness different from that displayed by the average scattered intensity. We conclude that the magnetization associated with interfacial features depends on the size of the feature.

We measure surface and interface morphology using two methods, atomic force microscopy (AFM) for the surface, and soft-x-ray scattering, which probes both the surface and buried interfaces. AFM images of the surface of our films show there is short-wavelength roughness in the 10–100 nm scale, but on larger scales the films are very smooth. This kind of morphology gives rise to a characteristic two-component profile in the x-ray scattering measurements, an instrument-limited central peak, and a diffuse component [5,6]. The scattered intensity as a function of detector position Ω out of the scattering plane is given by

$$I(\Omega) = I(\Omega)_{\text{specular}} + I(\Omega)_{\text{diffuse}}\,, \qquad (1)$$

with the incident angle θ held fixed as illustrated in Fig. 1. We refer to a scan in Ω as an azimuthal transverse scan. For a surface or multilayer with no interfacial roughness, only a specular peak would be observed. Roughness has the effect of scattering x rays away from the specular

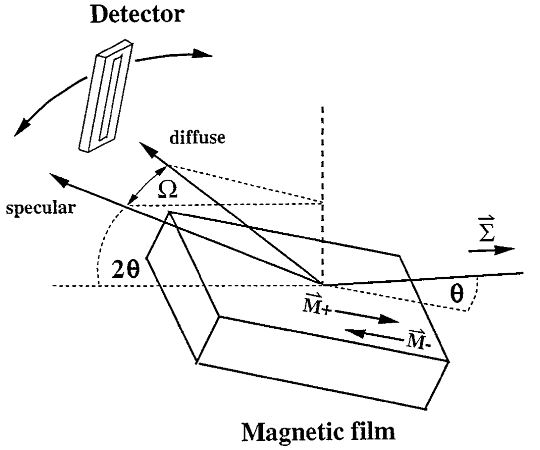

FIG. 1. Scattering geometry for azimuthal transverse scans from a magnetic film. θ is the angle of incidence and Ω is the azimuthal angle out of the scattering plane. The photon helicity Σ is shown with the magnetization parallel ($\mathbf{M}_+$) and antiparallel ($\mathbf{M}_-$) to the photon spin.

direction, creating a diffuse background. Both the specular and diffuse components are observed for $\Omega = 0$, although the specular component is typically very much larger. The angular width of the specular component is determined by beam size and the detector aperture. Modeling the intensity distribution allows one to extract the mean square roughness from the relative intensities of the specular and diffuse components, the lateral length scale of the roughness from the width of the diffuse component, and the power spectrum from the shape of the diffuse component [5,6].

The scattering amplitude for circularly polarized x rays at the L_2 or L_3 absorption edge of a transition element such as Co contains the usual charge scattering term plus a resonant magnetic scattering term that depends on the magnetization $\mathbf{M}$ [8]. The scattered intensity contains the product of magnetic and nonmagnetic amplitudes and is changed by reversing the magnetization. For the scattering geometry in Fig. 1 with a photon helicity parallel (+) or antiparallel (−) to the magnetization, the scattered intensity can be written as

$$I_\pm(\Omega) = I_{\text{charge}}(\Omega) + I_{\text{resonant}}(\Omega, \mathbf{M}_\pm)\,, \qquad (2)$$

where $I_{\text{resonant}}(\Omega, \mathbf{M}_\pm)$ refers to the component of the intensity that changes with reversal of the magnetization. The asymmetry ratio $(I_+ - I_-)/(1/2)(I_+ + I_-)$ is usually defined to quantify the change in the scattered intensity with the change of direction of the magnetization [14]. The denominator is simply the average intensity $I_{\text{ave}}(\Omega)$. The magnitude of the asymmetry ratio depends strongly on the angle of incidence [8,19]. However by holding the angle of incidence fixed and making azimuthal transverse scans in the plane normal to the scattering plane, the specular and diffuse intensities may be probed for a constant asymmetry ratio (see Fig. 1) [20]. We are interested in the angular dependence of the difference $[I_+(\Omega) - I_-(\Omega)] = \Delta I(\mathbf{M}, \Omega)$, as the difference isolates the element specific resonant contribution to the scattered intensity. $\Delta I(\mathbf{M})$ is element specific because it is large only for scattering from magnetic atoms with an absorption edge corresponding to the selected photon energy. $\Delta I(\mathbf{M}, \Omega)$ will have a specular and diffuse component as in Eq. (1),

$$\Delta I(\mathbf{M}, \Omega) = \Delta I(\mathbf{M}, \Omega)_{\text{specular}} + \Delta I(\mathbf{M}, \Omega)_{\text{diffuse}}\,. \qquad (3)$$

The morphology of the boundaries between the magnetic film and the nonmagnetic layer or surface is reflected in the shape and relative intensity of the diffuse component $\Delta I(\mathbf{M}, \Omega)_{\text{diffuse}}$. If the roughness of the magnetic boundary is identical to the interfacial roughness, then $\Delta I(\mathbf{M}, \Omega)$ will have the same relationship between the specular and diffuse components as the average intensity $I_{\text{ave}}(\Omega)$. In addition $I_{\text{ave}}(\Omega)_{\text{diffuse}}$ and $\Delta I(\mathbf{M}, \Omega)_{\text{diffuse}}$ will have the same shape and width. It is, however, possible that $\Delta I(\mathbf{M}, \Omega)_{\text{diffuse}}$ can differ from $I_{\text{ave}}(\Omega)_{\text{diffuse}}$. If the magnetic moments at the interface do not respond to the reversal of the magnetic field in the same way as those in the bulk of the film, then there will be a difference between the ratio of the specular and diffuse components of $\Delta I(\mathbf{M}, \Omega)$ when compared to $I_{\text{ave}}(\Omega)$. It is also possible that $\Delta I(\mathbf{M}, \Omega)_{\text{diffuse}}$ can have a shape different from that of $I_{\text{ave}}(\Omega)_{\text{diffuse}}$, which would indicate that the roughness of the magnetic boundary does not have the same length scale as the interfacial roughness.

We deposited Co films and Co/Cu/Co sandwiches by dc magnetron sputtering in a system with a base pressure of 5×10^{-8} Torr. Films were grown at a pressure of 2.0 mTorr on Si(100) and capped with 2.0 nm of Al to prevent oxidation. AFM in air was used to image the surface before and after growth. Diffuse x-ray scattering measurements were performed using a W/C multilayer monochromator (beamline 051) at the Synchrotron Radiation Center. The monochromator delivers 6×10^8 elliptically polarized photons/sec to the sample at ~780 eV, near the L_3 edge of Co. Azimuthal transverse scans are made with a three-circle diffractometer, holding the angle of incidence fixed at 3 degrees. The scattered intensity is measured using a channel electron multiplier and a Si photodiode as the photocathode. The detector is masked by a slit 0.7 mm wide by 10 mm high, so that the intensity in the scattering plane is integrated (see Fig. 1). An electromagnet delivers a maximum field of 700 G either parallel or antiparallel to the photon helicity. Hysteresis curves were taken using the XRMS specular reflection to determine the coercive field and magnetic quality of each sample. All of the samples have coercive fields less than 50 G, and nearly square hysteresis loops indicating that the films are single domain. We determine the difference $\Delta I(\mathbf{M}, \Omega)$ and the sum $I_{\text{ave}}(\Omega)$ by measuring I^+ and I^- point by point for the magnetic field, respectively, parallel and antiparallel to the photon helicity. $\Delta I(\mathbf{M}, \Omega)$ and

$I_{\rm ave}(\Omega)$ are normalized to one at the specular-peak maximum $\Omega = 0$. Even though we cannot compare the absolute intensities, normalization allows us to compare the relative shapes of the specular and diffuse intensities of $\Delta I(\mathbf{M},\Omega)$ and $I_{\rm ave}(\Omega)$.

Figure 2 compares $\Delta I(\mathbf{M},\Omega)$ and $I_{\rm ave}(\Omega)$ transverse scans for a 7.0 nm thick Co film grown on Si(100) and capped with 2.0 nm of Al. From fits to $I_{\rm ave}(\Omega)$ we obtain an rms roughness of the Co film of $\sigma = 0.29$ nm and a correlation length of $\xi = 12.5 \pm 1.5$ nm, in good agreement with AFM measurements. These are typical values for sputter deposited films [5,6]. The normalized $I_{\rm ave}(\Omega)$ specular peak is reproduced by the normalized $\Delta I(\mathbf{M},\Omega)$ specular peak, as expected, because the latter reflects the magnetic scattering from all the atoms. In contrast, the diffuse background for the normalized $\Delta I(\mathbf{M},\Omega)$ is smaller, with less intensity scattered away from the specular peak, demonstrating that the magnetic-boundary roughness is less than the interfacial roughness. In addition, the shape of the diffuse background for the normalized $\Delta I(\mathbf{M},\Omega)$ is different from that of $I_{\rm ave}(\Omega)$. The narrowing of $\Delta I(\mathbf{M},\Omega)_{\rm diffuse}$ compared to $I_{\rm ave}(\Omega)_{\rm diffuse}$ implies a longer effective lateral correlation length, and demonstrates that Co atoms constituting the short-range structural roughness at the interfaces of this film do not contribute as well to magnetic scattering. Fits to $\Delta I(\mathbf{M},\Omega)$ given an rms roughness for the magnetic boundary of $\sigma = 0.145$ nm with a correlation length $\xi = 20 \pm 5$ nm.

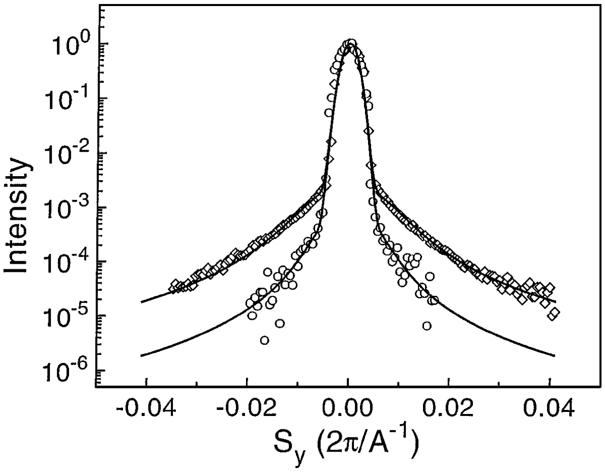

FIG. 2. $\Delta I(\mathbf{M},\Omega)$ and $I_{\rm ave}(\Omega)$ transverse scans normalized to their respective specular peaks at $\Omega = 0$ for a 7.0 nm Co film capped with 2.0 nm of Al grown on Si(100). The curves are plotted as a function of S_y, the momentum transfer perpendicular to the scattering plane ($S_y = (2\pi/\lambda)\cos\theta\sin\Omega$, where λ is the photon wavelength). The diamonds are $I_{\rm ave}(\Omega)$ and the circles are $\Delta I(\mathbf{M},\Omega)$. The best fit to $I_{\rm ave}(\Omega)$ gives an rms roughness of $\sigma = 0.29$ nm and a correlation length $\xi = 12.5 \pm 1.5$ nm. The best fit to $\Delta I(\mathbf{M},\Omega)$ gives an rms roughness of the magnetic boundary of $\sigma = 0.15$ nm and a correlation length $\xi = 20 \pm 5$ nm.

Figure 3 compares $\Delta I(\mathbf{M},\Omega)$ and $I_{\rm ave}(\Omega)$ azimuthal scans normalized at $\Omega = 0$ for an Al capped Co(2.0 nm)/Cu(1.0 nm)/Co(2.0 nm) sandwich grown on Si(100). The results are similar to those displayed by the single Co film. From $I_{\rm ave}(\Omega)$ we obtain an rms roughness of 0.30 nm with $\xi = 13 \pm 1$ nm, in agreement with AFM, while $\Delta I(\mathbf{M},\Omega)$ gives an rms roughness of $\sigma = 0.15$ nm and a correlation length of $\xi = 19 \pm 6$ nm. The trend illustrated by the films in Figs. 2 and 3 of decreased $\Delta I(\mathbf{M},\Omega)_{\rm diffuse}$, giving lower rms roughness and somewhat increased correlation length compared to $I_{\rm ave}(\Omega)$, continues for rougher films grown by increasing the sputtering pressure. In contrast, similar films grown on substrates deliberately prepared with periodic laterally correlated roughness of long correlation length are very different. For these films $\Delta I(\mathbf{M},\Omega)$ and $I_{\rm ave}(\Omega)$ are identical, indicating that the roughness and correlation length of the magnetic boundary are the same as the interfacial roughness [21].

To summarize, diffuse XRMS intensity measurements demonstrate that the magnetic scattering at the interfaces differs from charge scattering for these samples. The magnetic boundary roughness appears to be smaller than the interfacial roughness. Several explanations for this difference in response at the magnetic boundary are possible. The Co moments at the interface could be pinned by the surrounding atoms, either Al, Cu, or Si. We cannot discount the possibility that the coercive field was not sufficient to align the magnetic moment of Co atoms at the interface with those of the bulk film. Increasing the applied magnetic field to 700 G (~10 times the coercive field) did not change $\Delta I(\mathbf{M},\Omega)_{\rm diffuse}$. It would be

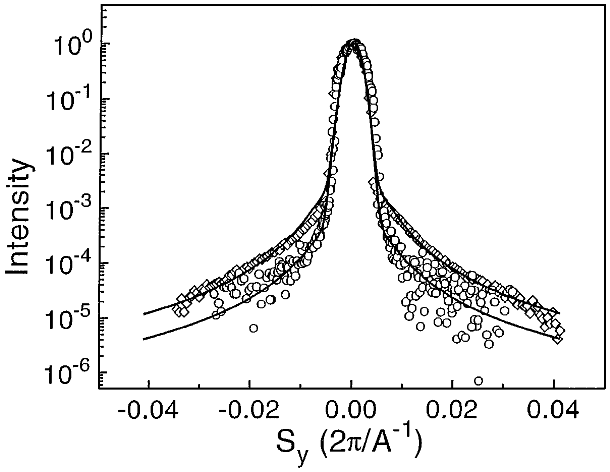

FIG. 3. $\Delta I(\mathbf{M},\Omega)$ and $I_{\rm ave}(\Omega)$ transverse scans of a Co(2.0 nm)/Cu(1.0 nm)/Co(2.0 nm) sandwich capped with 2.0 nm of Al grown on Si(100). The diamonds are $I_{\rm ave}(\Omega)$ and the circles are $\Delta I(\mathbf{M},\Omega)$. The best fit to $I_{\rm ave}(\Omega)$ gives an rms roughness of $\sigma = 0.30$ nm and a correlation length $\xi = 13 \pm 1$ nm. The best fit to $\Delta I(\mathbf{M},\Omega)$ gives an rms roughness of the magnetic boundary of $\sigma = 0.15$ nm and a correlation length $\xi = 19 \pm 6$ nm.

difficult to confirm by conventional experiments such as the magneto-optic Kerr effect or x-ray magnetic circular dichroism that the field was too weak, because the contribution to the signal from the interface is 10^3 smaller than that from the bulk. A second explanation for the different response of magnetic moments at the interface is an enhanced orbital moment for atoms there [22]. An enhanced moment seems unlikely for the following reasons. Recent x-ray magnetic circular dichroism experiments indicate that the asymmetry ratio for atoms at the interface is $1/2$ that for atoms in the bulk of the film, whereas the asymmetry ratio for magnetic scattering at the interface of our films is less than one-tenth that of atoms in the bulk film. Additionally, the asymmetry ratio is not constant over the diffuse XRMS intensity, inconsistent with a single value for an enhanced orbital moment. Hence, we do not believe that an enhanced orbital moment can be responsible for the effect that we observe. We suggest instead a different explanation. We believe that roughness causes weaker coupling of the magnetic moments of interface atoms to the bulk magnetization of the film, with the sharpest structural asperities at the interface most poorly coupled to the bulk magnetization, while smoother variations are better coupled. Moments at the interface could fluctuate in the plane about the direction of the external magnetic field. Recent calculations support this picture [21]. The coupling would then depend on temperature: Thermally assisted fluctuation of the magnetic moments would be more effective in decoupling spins of atoms at sites where the number of magnetic neighbors is most reduced. The fact that the interfaces for charge and magnetic scattering are similar at long length scales supports this view. A measurement of the temperature dependence of the diffuse XRMS intensity can confirm this picture as well as differentiate between pinned magnetic moments and reduced coupling.

In conclusion, we have shown that with a high-flux, tunable, and stable source of soft x rays, the interfaces a magnetic film or a magnetic multilayer makes with other elements or the vacuum can be probed by measurements of the x-ray-magnetic-scattering diffuse intensity. For sputter deposited Co and Co/Cu/Co films with typical interface roughness, we find that the magnetic boundary roughness differs from the structural interface roughness. The magnetic boundary appears to be smoother. We suggest that the magnetic moments of atoms that “stick out,” producing the short-wavelength roughness, are not able to follow the magnetization reversal and thus effectively do not contribute to the resonant magnetic scattering. We are presently measuring the temperature dependence of the diffuse magnetic scattering, and working to explain both theoretically and experimentally the dependence of magnetic scattering on the size of interfacial features.

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[1] *Magnetic Ultrathin Films,* edited by B.T. Jonker *et al.*, MRS Symposia Proceedings No. 313 (Materials Research Society, Pittsburgh, 1993).

[2] S.S.P. Parkin, N. More, and K.P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).

[3] Zhu-Pei Shi, P.M. Levy, and J.L. Fry, Phys. Rev. B **49**, 15 159 (1994).

[4] P. Beliën, R. Schad, C.D. Potter, G. Verbanck, V.V. Moshchalkov, and Y. Bruynseraede, Phys. Rev. B **50**, 9957 (1994).

[5] S.K. Sinha, E.B. Sirota, S. Garoff, and H.B. Stanley, Phys. Rev. B **38**, 2297 (1988).

[6] D.E. Savage, J. Kleiner, N. Schimke, Y.-H. Phang, T. Jankowski, J. Jacobs, R. Kariotis, and M.G. Lagally, J. Appl. Phys. **69**, 1411 (1991).

[7] Doon Gibbs, D.R. Harshman, E.D. Isaacs, D.B. McWhan, D. Mills, and C. Vettier, Phys. Rev. Lett. **61**, 1241 (1988).

[8] J.P. Hannon, G.T. Trammell, M. Blume, and Doon Gibbs, Phys. Rev. Lett. **61**, 1245 (1988).

[9] F. de Bergevin and M. Brunel, Acta Crystallogr. Sect. A **37**, 314 (1981).

[10] J.M. Tonnerre, L. Sève, D. Raoux, G. Soullié, B. Rodmacq, and P. wolfers, Phys. Rev. Lett. **75**, 740 (1995).

[11] E.D. Isaacs, D.B. McWhan, C. Peters, G.E. Ice, D.P. Siddons, J.B. Hastings, C. Vettier, and O. Vogt, Phys. Rev. Lett. **62**, 1671 (1989).

[12] D.B. McWhan, C. Vettier, E.D. Isaacs, G.E. Ice, D.P. Siddons, J.B. Hastings, C. Peters, and O. Vogt, Phys. Rev. B **42**, 6007 (1990).

[13] C.C. Tang, W.G. Stirling, G.H. Lander, Doon Gibbs, W. Herzog, Paolo Carra, B.T. Thole, K. Mattenberger, and O. Vogt, Phys. Rev. B **46**, 5287 (1992).

[14] F. de Bergevin, M. Brunel, R.M. Galéra, C. Vettier, E. Elkàim, M. Bessière, and S. Lefèbvre, Phys. Rev. B **46**, 10 772 (1992).

[15] D.B. McWhan, E.D. Issacs, Paolo Carra, S.M. Shapiro, B.T. Thole, and S. Hoshino, Phys. Rev. B **47**, 8630 (1993).

[16] B.A. Everitt, M.B. Salamon, B.J. Park, C.P. Flynn, T. Thurston, and Doon Gibbs, Phys. Rev. Lett. **75**, 3182 (1995).

[17] T.R. Thurston, G. Helgesen, Doon Gibbs, J.P. Hill, B.D. Gaulin, and G. Shirane, Phys. Rev. Lett. **70**, 3151 (1993).

[18] C. Kao, J.B. Hastings, E.D. Johnson, D.P. Siddons, G.C. Smith, and G.A. Prinz, Phys. Rev. Lett. **65**, 373 (1990).

[19] C.-C. Kao, C.T. Chen, E.D. Johnson, J.B. Hastings, H.J. Lin, G.H. Ho, G. Meigs, J.-M. Brot,S.L. Hulbert, Y.U. Idzerda, and C. Vettier, Phys. Rev. B **50**, 9599 (1994).

[20] The asymmetry ratio is proportional to $\cos\theta(1 + \cos\Omega)$.

[21] D. Zhao, F. Liu, J.F. MacKay, D.L. Huber, and M.G. Lagally (to be published).

[22] M. Ticher, O. Hjortstam, D. Arvanitis, J.Hunter Dunn, F. May, K. Baberchke, J. Trygg, J.M. Willis, B. Johansson, and O. Eriksson, Phys. Rev. Lett. **75**, 1602 (1995).