Magnetic profile as a function of structural disorder in Fe/Cr superlattices

Michael J. Pechan
Department of Physics, Miami University, Oxford, Ohio 45056

J. F. Ankner and C. F. Majkrazk
NIST, Gaithersburg, Maryland 20899

David M. Kelly and Ivan K. Schuller
Department of Physics, UC San Diego, La Jolla, California 92093

Structural disorder has been systematically introduced in an $[Fe(30\ Å)/Cr(17\ Å)]_{10}$ superlattice to investigate its effects on the magnetization profile. Low angle x-ray and neutron scattering results indicate progressive structural degradation in samples produced with increased argon pressure (3, 6, and 12 mTorr). The x-ray spectra from the more disordered samples are characterized by a decreased intensity and increased linewidth of the superlattice Bragg peaks, along with a systematic disappearance of the finite size intensity oscillations. Elastic, polarized, low angle, neutron reflectivity data exhibit decreasing superlattice Bragg intensities and increasing linewidths, indicating increased disorder in agreement with x-ray data. Antiferromagnetic alignment of adjacent Fe layers is inferred by the appearance of magnetic peaks between the structural peaks and by the lack of magnetic contribution to the structural superlattice peaks. Interestingly, the magnetic superlattice peak does not exhibit appreciable degradation with increasing disorder in the structure, indicating a magnetic coherence length significantly greater than that of the chemical superlattice structure.

INTRODUCTION

Magnetic multilayer materials are proving fruitful in both technological and fundamental arenas. This is perhaps most readily exemplified in magnetic/nonmagnetic multilayers in which large magnetotransport effects are observed in a wide range of systems, which also exhibit magnetic interlayer coupling.[1] An understanding of the role played by superlattice structure in determining the magnetic and transport properties is essential, and is the subject of considerable theoretical and experimental investigation. Degree of epitaxy and varying crystalline orientation are examples of parameters varied in probing structural dependences. Neutron scattering has been utilized by others in structural investigations of Fe/Cr,[2] but not to probe systems wherein roughness is a controlled parameter. Recently, a significant correlation was shown between the size of magnetoresistance and structural disorder, by systematically increasing and characterizing the superlattice disorder in Fe/Cr multilayers.[3] As an extension of this approach, the present investigation characterizes the magnetization profile as a function of structural disorder in the Fe/Cr system. Results indicate a magnetic multilayer profile that is essentially independent of structural disorder over the ranges investigated.

EXPERIMENTAL DETAILS AND ANALYSIS TECHNIQUES

Fe(30 Å)/Cr(17 Å) multilayer samples were prepared on [100] Si substrates by dc sputtering in an Ar atmosphere. System base pressure was less than 2×10^{-7} Torr. First Fe, then Cr was deposited, forming a bilayer structure that was repeated ten times for each sample. Structural disorder was introduced by increasing the Ar pressure during sputtering from 3 to 12 mTorr. Both high and low angle x-ray (Cu, K_α, $\lambda=1.54$ Å) measurements were performed on each sample. Low angle diffuse scattering contributions were accounted for by repeating the specular scans with the sample rotated 0.1° away from the superlattice Bragg condition and subtracting from the specular scans. Superlattice structural information is obtained by fitting the specular low angle spectra to a classical optics model,[4] where each layer is assigned a thickness and scattering density, and has an associated interfacially mixed region. Polarized, low angle neutron ($\lambda=2.37$ Å) reflectivity measurements were also performed, providing additional structural information and characterization of the multilayer magnetic profile. Polarized neutrons have historically been used, at the atomic level (and therefore high scattering angles), to extract the magnetic contribution from a combined nuclear/magnetic scattering amplitude. Recently,[5,6] the technique has been applied to layered structures, operating in the low angle regime, to characterize the superlattice magnetization profile and chemical modulation. A small quantization field, $\mathbf{H}=14$ Oe is applied in the plane of the film, following *ex situ* application of 200 Oe in the same direction. The scattering vector is normal to the film plane. Assuming an in-plane magnetization (appropriate for Fe/Cr), one can realize two scattering processes–spin flip (SF) and non-spin flip (NSF)—each with two channels: $(-+)$ and $(+-)$ in the SF and $(++)$ and $(--)$ in the NSF, where the states refer to the spin polarization of the incoming and reflected neutrons, respectively. The SF process refers to scattering from a component of the magnetization M normal to $\mathbf{H}$, whereas NSF scattering arises from components of M along $\mathbf{H}$ and from the nuclei. The scattering intensities from the two SF channels will be identical, but the NSF $(++)$ intensity is derived from the sum of the magnetic and nuclear scattering amplitudes, whereas the $(--)$ intensity arises from their difference. Low angle neutron spectra are also analyzed according to a classical optics model, utilizing the same fitting approach as for the x rays.

RESULTS AND DISCUSSION

Low angle x-ray spectra for samples prepared with increasing Ar pressure are shown in Fig. 1. The sample sput-

 0021-8979/94/75(10)/6178/3/$6.00

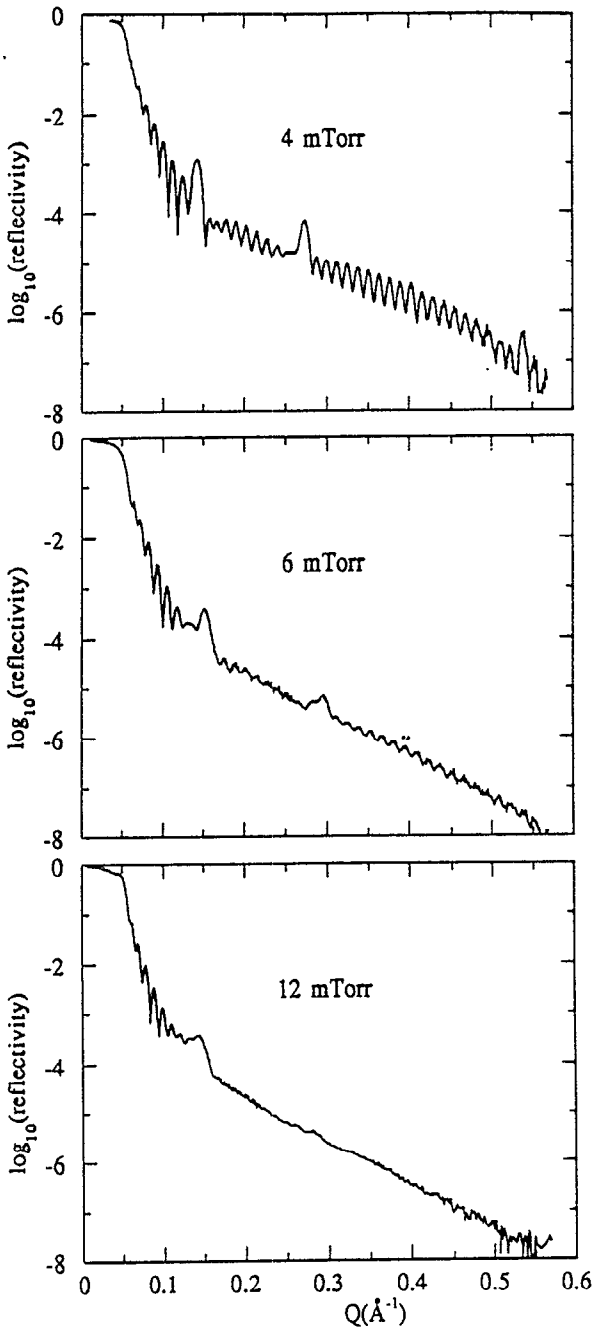

FIG. 1. Low angle x-ray spectra for $[Fe(30\ Å)/Cr(17\ Å)]_{10}$ samples prepared at various Ar sputtering pressures.

tered at 4 mTorr exhibits superlattice Bragg peaks out to fourth order (the third order is suppressed, owing to the approximate 2:1 thickness ratio of the bilayer components), indicating square wave chemical modulation. In addition, short period, finite size peaks, (interference from the top and bottom of the aggregate bilayer stack) are evident throughout the spectrum, attesting to uniform film thickness. These spectral features degrade with increasing Ar sputtering pressure. The finite size peaks become much less pronounced and less extended in Q space, indicating decreasing uniformity in the overall film thickness. The superlattice Bragg peaks exhibit progressively weaker higher-order amplitudes and increasing linewidths. One may be tempted to conclude, based upon the disappearance of higher-order amplitudes, that the samples are increasingly characterized by interfacial interdiffusion. However, interdiffusion alone will *not* increase linewidths *nor* depress the finite size peak structure. Rather, these latter effects are known to arise from cumulative random variations in layer thickness,[7] which can superimpose wavelike modulations on the superlattice structure. Such fluctuations would also give rise to *apparent* interdiffusion as they are averaged over the lateral coherence length (>1000 Å) of the low angle x rays, producing the observed broadening of the higher-order Bragg peaks. The present optic model used to extract structural information treats thickness fluctuations in the simplest sense—layers are assumed flat but their thicknesses are adjustable. In order to produce the increase in linewidth observed in going from the 4 to 12 mTorr sample, the Cr layer thicknesses went from 100% t_{Cr}=17 Å (and Fe/Cr intermixing of 4 Å) to 50% t_{Cr}=18.6 Å, 25% each of t_{Cr}=17.4 and 20.1 Å (again Fe/Cr intermixing of 4 Å). A more complex model of the layer thickness fluctuations may be useful for explaining details of the spectra, however, this simple approach reproduces the measured data adequately for the present discussion.

The *high* angle x-ray spectra are essentially indistinguishable for the samples shown in Fig. 1. All films exhibit a predominant bcc (110) orientation with linewidths, indicative of approximately 130 Å grain sizes. Owing to the similar scattering strengths of Fe and Cr, only weak superlattice satellites are observed at high angle, however, these provide some indication of the interfacial mixing on the atomic scale. The fact that the "smoothest" through the "roughest" samples show the same high angle pattern, yet differ so greatly in low angle, indicates that the roughness induced is on a length scale much greater than the interatomic spacing, but less than the coherence length of the x-ray beam.

Low angle polarized neutron spectra are shown in Fig. 2. The NSF peaks seen at approximately 0.14 $Å^{-1}$ correspond to the first-order superlattice Bragg peaks seen in the x-ray spectra. No difference is observed in these (++) and (−−) intensities, indicating no magnetic component to the scattering amplitude, and therefore ruling out ferromagnetic alignment of adjacent Fe layers. The half-order (0.07 $Å^{-1}$) NSF peaks again show no difference in the (++) and (−−) intensities. However, based upon the x-ray spectra (no Bragg peak), one assumes *only* magnetic scattering, which arises from antiferromagnetically aligned adjacent Fe layers. Half-order peaks are also observed in SF scattering, indicating that either the Fe sublattice magnetization makes an angle with respect to the applied quantizing field, **H**, or that there exist AF domains parallel and perpendicular to **H**. As with the x rays, the neutrons show a degradation of the chemical superlattice structure with increasing Ar pressure. The NSF AF peak also decreases in intensity with increasing sample roughness, however, the SF AF peak changes very little during the roughening process.

How can we understand a system where the magnetic structure persists in the face of degrading chemical superlattice structure? As evidenced by the decrease, with roughness, in NSF magnetic scattering, while the SF remains essentially

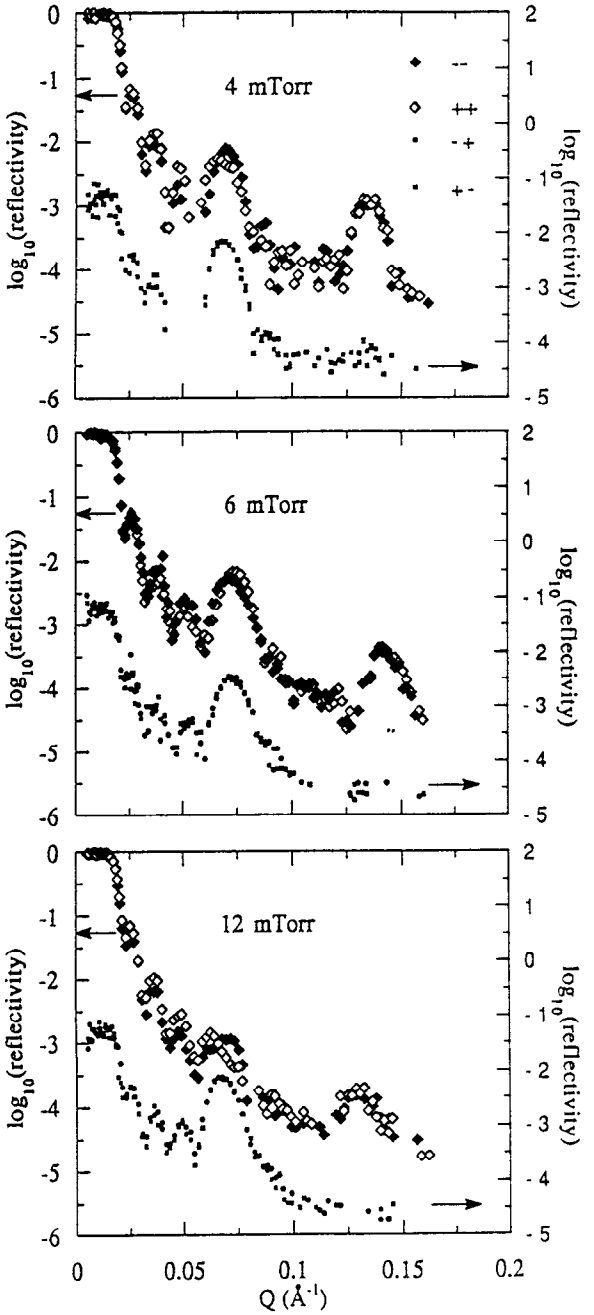

FIG. 2. Low angle polarized neutron spectra for $[Fe(30\ Å)/Cr(17\ Å)]_{10}$ samples prepared at various Ar sputtering pressures. Open diamonds and closed diamonds represent the two NSF (++) and (−−) scattering channels, respectively, whereas both SF (−+) and (+−) channels are represented by closed squares.

constant, the Fe layer superlattice becomes less ordered. This is consistent with magnetization measurements,[2] where the remnant moment is much larger in the rougher sample, indicating a decrease in the AF coupled component, but would require alignment without spatial coherence among the ferromagnetic component comprising the remnant. It is not hard to imagine random layer thickness fluctuations producing such a remnant. Why is the half-order magnetic peak retained in the SF component, but not the NSF? In the absence of in-plane anisotropy, the initial response of a simple AF structure is for each sublattice to align perpendicular to an applied field. This arrangement would give rise to half-order peaks in the SF but not the NSF spectra. Therefore one possibility here is that an in-plane ansotropy exists in the smoother sample, which disappears in the rougher. On the other hand, if the NSF and SF components in the smoother sample arise from AF domains with distributed orientations, then the coercive field must be reduced with increasing roughness. Finally, why is the SF AF peak affected so little by roughening? In the smooth samples, an AF component is pinned parallel to H, presumably by some in-plane anisotropy. With roughening, the spacially coherent AF volume fraction decreases (declining total AF scattered intensity), but this is accompanied by an apparent decrease in pinning strength (disappearance of the NSF AF scattering). Therefore the data would suggest a compensation process, wherein the AF component normal to the field is degraded by the decreasing coherent AF volume fraction, but is enhanced as a larger portion of that fraction orients normal to that field.

Addressing more precisely the relationship between the magnetic multilayer profile and structural disorder will require careful analysis of diffuse as well as specular scattering components of both x rays and neutrons.[8]

However, the most striking feature of the neutron data is the persistance of a coherent magnetic scattering in the face of greatly degraded chemical superlattice scattering. This, interestingly, implies a magnetic coherence length significantly greater than that of the chemical superlattice.

ACKNOWLEDGMENTS

This work was supported in part by the U.S. Department of Energy Contract No. DE-FG02-86ER45281 (Miami) and National Science Foundation Contract No. NSF DMR 9201698 (UCSD).

[1] A. Fert, A. Barthelemy, P. Lequien, R. Loloee, D. K. Lottis, D. H. Mosca, F. Petroff, W. P. Pratt, and P. A. Schroeder, J. Magn. Magn. Mater. **104–7**, 1712 (1992).

[2] A. Barthelemy, A. Fert, M. N. Babich, S. Hadjoudj, F. Petroff, P. Etienne, R. Cabanel, S. Lequien, F. Nguyen Van Dau, and G. Creuzet, J. Appl. Phys. **67**, 5908 (1990); N. Hosoito, S Araki, K. Mibu, and T. Shinjo, J. Phys. Soc. Jpn. **59**, 1925 (1990); S. S. Parkin, A. Mansour, and G. P. Felcher, Appl. Phys. Lett. **58**, 1473 (1991); J. A. C. Bland, A. D. Johnson, H. J. Lauter, R. D. Bateson, S. J. Blundell, C. Shackleton, and J. Penfold, J. Magn. Magn. Mater. **93**, 513 (1991). N. Hosito, K. Mibu, S. Araki, T. Shinjo, S. Itoh, and Y. Endoh, J. Phys. Soc. Jpn. **61**, 300 (1992); M. Takeda, Y. Endoh, H. Yasuda, K. Yamada, A. Kamijo, and J. Mizuki, J. Phys. Soc. Jpn. **62**, 3015 (1993). M. Lowenhaupt, W. Hahn, Y. Y. Huang, G. P. Felcher, and S. S. Parkin, to appear in Int. J. Phys. B.

[3] Eric E. Fullerton, D. M. Kelly, J. Guimpel, and I. K. Schuller, Phys. Rev. Lett. **68**, 859 (1992).

[4] J. F. Ankner and C. F. Majkrzak, in *Neutron Optical Devices and Applications*, edited by C. F. Majkrzak and J. L. Wood, Proc. SPIE, Bellingham, WA, 1992, Vol. 1738, p. 260.

[5] G. P. Felcher, R. O. Killeke, R. K. Crawford, J. Haumann, R. Kleb, and G. Ostrowski, Rev. Sci. Instrum. **58**, 609 (1987).

[6] C. F. Majkrzak, Physica B **173**, 75 (1991).

[7] E. E. Fullerton, I. K. Schuller, H. Vanderstraeten, and Y. Bruynseraede, Phys. Rev. B **45**, 9292 (1992). Also, as an example of cumulative random layer thickness fluctuations, see E. E. Fullerton, I. K. Schuller, and Y. Bruynseraede, MRS Bulletin, Dec. 1992, p. 33.

[8] S. K. Sinha, Physica B **173**, 25 (1991).