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Interfacial structure and giant magnetoresistance in Fe/Cr superlattices

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A correlation between interfacial structure and giant magnetoresistance (GMR) has been established for Fe/Cr superlattices [1-8]. However, previous studies were qualitative in that the interfacial structural disorder was expressed in terms of growth temperature, sputtering pressure or annealing temperature. In order to understand more quantitatively the effects of interfacial structure on GMR, we have undertaken [9] parallel measurements of x-ray diffraction (XRD) and magnetoresistance for two Fe/Cr samples subjected to a series of anneals at successively higher temperatures, both samples having initial GMR > 50 %. Such anneals are known to increase the thickness of the Fe/Cr interface through atomic diffusion, a structural change which can be characterized quantitatively by XRD. The advantages of high intensity and anomalous dispersion provided by synchrotron radiation has greatly contributed to the quality of our x-ray data, for which the use of simulation programs has allowed a robust and precise extraction of several structural parameters for the two samples, including the Fe/Cr interfacial thicknesses.

1. DESCRIPTION OF EXPERIMENTS

The two $(\text{Cr}_{11}\text{A}/\text{Fe}_{28}\text{A})_{10}/\text{MgO}(001)$ superlattice samples were produced (K.U. Leuven) using molecular beam epitaxy (MBE) at two different temperatures: 150 °C for sample B5 and 18 °C for sample B6, having overall sizes of about $5 \times 5 \times 0.5 \text{ mm}^3$ and $3 \times 5 \times 0.5 \text{ mm}^3$, respectively. The samples have been stored at ambient temperature and pressure since their growth in Sept 1991.

The x-ray diffraction measurements were carried out using a 4-circle goniometer at the D23A anomalous dispersion beamline at the DCI synchrotron (LURE, Orsay). A Si(111) double-crystal sagittal-focusing monochromator gave sufficient energy resolution ($\Delta E/E \approx 10^{-4}$) to choose an x-ray energy only 15 eV below the K-absorption edge of Cr. This choice allowed a decrease of $\sim 6 e^-$ for each Cr atom, resulting in an electron density contrast ($\rho_{\text{Fe}} - \rho_{\text{Cr}}$) enhancement of a factor of ~ 3 and hence an intensity enhancement of ~ 10 . The technique of anomalous dispersion was therefore a great advantage to the feasibility of these experiments, the inelastic scattering signal being blocked by a Ge(111) analyser crystal before the NaI detector.

The magnetoresistance (MR) of the samples was measured (Lab. Phys. Sol., Univ. Paris-Sud) with a 4-probe (in-line) low frequency (22Hz) AC lock-in technique. The four electrical contacts were effected using silver paint applied directly to the sample surface, which was subsequently cleaned with ethanol followed by an isopropanol rinse. All the MR data presented here were taken at $T=4.2 \text{ K}$ in magnetic fields up to 40 kOe (4 tesla) applied in the plane of the Fe/Cr layers and parallel to the current. The magnetization curves $M(H)$ were measured at $T=20 \text{ K}$ using a Quantum Design SQUID magnetometer.

The anneals took place in a vacuum oven having a pressure of $\sim 10^{-6}$ Torr at the annealing temperatures T_{ann} of 295, 330 and 370 °C. The temperature trajectory for all 3 anneals consisted of a 1 hr ramp to T_{ann} followed by 1 hr at T_{ann} and then an immediate descent to room temperature.

We emphasize that the anneals have thus been carried out *successively* on the *same* samples, which eliminates critical sample-dependent effects such as the the thicknesses of the Cr layers [10].

2. RESULTS AND DISCUSSION

For structural characterization, x-ray diffraction has two principal advantages over other techniques (such as TEM): it is a non-destructive probe, and it measures structural parameters as averaged over the entire illuminated volume of the sample. We performed a variety of x-ray diffraction measurements both at small (SAXS) and large (LAXS) angles in order to probe both the mesoscopic and atomic structures, respectively.

Figures 1 and 2 show the SAXS data for the two samples, and Figs. 3 and 4 the LAXS data, as a function of T_{ann} . The data for the anneals have been shifted vertically for clarity.

Figure 1: SAXS data for sample B5

Figure 2: SAXS data for sample B6

Figure 3: LAXS data for sample B5

Figure 4: LAXS data for sample B6

The solid lines for the 370 °C anneal data in Figs. 1 and 2 are examples of our SAXS data simulations using the matrix method formalism [11] of calculating the x-ray reflectivity of a multilayer, wherein the interfacial region was modeled as a series of fine steps in electron density. The simulations provided a precise determination of the Fe and Cr layer thicknesses for the two samples: $\text{Cr}_{10.5\text{\AA}}/\text{Fe}_{28\text{\AA}}$ for B5 and $\text{Cr}_{11\text{\AA}}/\text{Fe}_{27\text{\AA}}$ for B6. The only simulation parameter varied between anneals was the average interfacial thickness (or roughness amplitude) between the Fe and Cr layers, which we quote as a standard deviation σ for the interface profile (the results appear later in Tables I and II). Qualitatively however, one can see by the appearance of 3 superlattice peaks in Fig. 1 (at $2\theta \approx 3, 6$ and 9 degrees) as opposed to only 2 peaks in Fig. 2 that sample B5 has less disordered interfaces (smaller σ) than sample B6.

The LAXS data of Figs. 3 and 4 show the clear presence of satellite peaks on either side the central peak, attesting to the atomic structural coherence across Fe and Cr layers. However, changes on annealing for the LAXS data are less apparent than for the SAXS data, owing to poorer counting statistics at large angles (in spite of the intensity enhancement from anomalous dispersion and a counting time of ~ 6 hours for each curve).

Both the SAXS and LAXS data represent *specular* diffraction scans (equal incident and reflected angles). Since a perfect sample would not scatter any photons in a non-specular direction, we can obtain more direct information about structural imperfections by performing *off-specular* (also called diffuse-scattering) measurements. In addition, whereas specular x-ray scans probe the electron density of the superlattice in the vertical direction only (*i.e.* $\perp$ to the interfaces), *off-specular scans also give structural information in the horizontal (x) direction*, such as the correlation length ξ_x of the interfacial roughness [12].

For our Fe/Cr samples we performed two types of off-specular scans, both at small angles: i) ω -rocks (or rocking curves), where ω is the angle between the sample's surface and the incident beam, and ii) 2θ -rocks (or detector scans), where 2θ is the angle between the detector and the incident beam. Each scan contains one specular peak, taking place when $2\theta = 2\omega$.

Figures 5 and 6 show ω -rocks and 2θ -rocks for sample B6 around the same specular peak (we used the 4th Kiessig fringe at $2\theta = 2\omega = 1.92^\circ$), which appears as the sharpest and most intense peak in the two figures. The off-specular signal is produced by a variety of (elastic) diffuse scattering processes in the sample, which for these small-angle data take place mostly at the Fe/Cr interface and the outer sample surface. The most evident feature in these data is the continued relative decrease (on annealing) in the intensity of the specular peak with respect to the off-specular intensity, serving to confirm the accumulation of structural disorder.

Figure 5: ω -rock data for sample B6

Figure 6: 2θ -rock data for sample B6

The data to the right of the specular peak in Fig. 6 show the presence of several broad peaks. These peaks exist because of a non-zero *vertical* correlation length ξ_z for the Fe/Cr interfacial roughness [13]; that is, neighboring interface profiles are at least partially correlated in the growth direction (*i.e.* they track each other). If adjacent interfaces were completely uncorrelated, the off-specular scattering in the right of Fig. 6 would have the form of a smooth monotonic decay to zero, without peaks. The slope of this background decay, in turn, is related the average *horizontal* correlation length ξ_x of the Fe/Cr interfacial roughness, a weaker slope implying a smaller ξ_x . A very small $\xi_x \lesssim 15$ Å would correspond to interfacial diffusion rather than interfacial roughness, having a possibly different effect on GMR.

The roughness parameter ξ_x (which is very approximately the distance between roughness “bumps” along the interface) can also be extracted from the overall intensity of the off-specular scattering once the roughness amplitude σ has been determined from the specular data, since a value of $\xi_x = 0$ (corresponding to perfectly smooth interdiffusion, which is in reality always limited by atomic size) would produce no off-specular signal even for large σ . We have developed a simulation program for off-specular scattering from single surfaces, and we will soon generalize it to the case of multiple interfaces in order to extract values of ξ_x and ξ_z for our Fe/Cr samples.

We also performed ω -rocks at large angles, which allow a measure of the crystallinity or average domain size D_x parallel to the interfaces, obtaining values of ~ 90 Å for sample B5 and ~ 70 Å for sample B6, both showing no changes on annealing.

Figures 7, 8, 9 and 10 show some of our magnetoresistance (MR) data and magnetization curves ($M(H)$) for the two samples. The MR is defined as the ratio of the spin-dependent electrical resistivity $\Delta\rho = (\rho_o - \rho_s)$ to the spin-independent resistivity ρ_s , with ρ_o and ρ_s being measured at $H = 0$ and $H > H_s$, respectively. The saturation field H_s , obtained from the MR data from the intersection of the high and low-field slopes, is a measure of the *strength* of anti-ferromagnetic (AF) interlayer coupling.

Figure 7: MR data for sample B5 (T=4.2K)

Figure 8: MR data for sample B6 (T=4.2K)

Figure 9: M(H) data for sample B5 (T=20K)

Figure 10: M(H) data for sample B6 (T=20K)

Tables I and II summarize our magnetic measurement results and interfacial thickness σ values for the pre-anneal and annealed states of both samples. The values of M_r and M_s are the residual and saturation magnetizations obtained from the $M(H)$ curves of Figs. 9 and 10, with $1 - M_r/M_s$ being a measure of the *fraction* of AF order (as opposed to ferromagnetic order) in the samples.

Considering first the pre-anneal results in the tables, we note that the fraction of AF order is nearly the same for both samples, but that the strength of this coupling (given by H_s) is clearly lower for sample B6. This effect is well explained by the greater interfacial thickness σ of B6, since interfacial disorder has been shown to reduce the strength of AF coupling [14]. Samples grown at lower temperatures (such as B6) are known to have interfaces which tend to be rough (longer correlation lengths ξ_x) rather than interdiffused. It is therefore plausible that B6's interfaces are as abrupt as B5's, but simply more corrugated.

On annealing, both samples showed increases in their interfacial thicknesses σ . In fact, the annealing-induced increases in interfacial thickness $\Delta\sigma_{\text{ann}}$ are the same for both samples within our uncertainty of ± 0.2 Å, which is consistent with the annealing producing an equal amount of atomic interdiffusion for both samples. Moreover, since both samples B5 and B6 had nearly no pre-anneal Fe/Cr interdiffusion (the first because of a very small σ and the second because of a very low growth temperature), we can conclude that $\Delta\sigma_{\text{ann}}$ is an approximate, but quantitative, measure of the absolute amount of interfacial *diffusion* for the two samples.

Both samples show expected decreases in H_s with increasing interfacial diffusion $\Delta\sigma_{\text{ann}}$, with the greater percent decrease for sample B5 being likely due to a larger percent increase in σ during

the anneals. Likewise, the fraction of AF order $1 - M_r/M_s$ for sample B5 decreases by a slightly larger percentage.

The MR data show slightly different behavior for the two samples on annealing, possibly due to their having different pre-anneal interfacial structures. The MR of sample B5 remained mostly constant until decreasing after the last anneal, while sample B6 showed a small increase in MR after the first anneal, followed by a continued decrease. Although uncertainties in measurement geometry hindered our obtaining accurate values for the absolute resistivities $\Delta\rho$ (spin-dependent) and ρ_s (spin-independent), their ratio (the MR) is geometry independent and was also confirmed through additional MR measurements on the same samples using a different technique.

Table I. X-ray and magnetic measurement results for sample B5.

T_{ann} ($^{\circ}\text{C}$)	σ ($\text{\AA}$)	$\Delta\sigma_{\text{ann}}$ ($\text{\AA}$)	$\text{MR}^{\text{def}} \triangleq \Delta\rho/\rho_s$	$1 - M_r/M_s$	H_s (kOe)
pre-anneal	0.5	0.0	66.7 %	0.857	11.91
295	1.9	1.4	65.4 %	0.805	11.40
330	2.1	1.6	66.5 %	0.768	10.47
370	2.5	2.0	56.2 %	0.701	10.05

Table II. X-ray and magnetic measurement results for sample B6.

T_{ann} ($^{\circ}\text{C}$)	σ ($\text{\AA}$)	$\Delta\sigma_{\text{ann}}$ ($\text{\AA}$)	$\text{MR}^{\text{def}} \triangleq \Delta\rho/\rho_s$	$1 - M_r/M_s$	H_s (kOe)
pre-anneal	2.1	0.0	52.2 %	0.886	8.45
295	3.3	1.2	57.6 %	0.874	8.33
330	3.6	1.5	51.2 %	0.832	8.29
370	4.2	2.1	46.6 %	0.771	8.06

3. CONCLUSIONS

At present we see no clear correlation for either sample between the evolution of the magnetoresistance data on annealing and the other measured properties, such as the annealing-induced interfacial diffusion thickness $\Delta\sigma_{\text{ann}}$. We intend to perform a second Fe/Cr annealing study in the near future, which will include absolute measurements of the resistivities $\Delta\rho$ and ρ_s as well as the extraction of interfacial correlation lengths ξ_x and ξ_z through analyses of off-specular x-ray diffraction data.

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