

# Mössbauer study of the influence of thermal treatment on giant magnetoresistance and interface structure in Fe/Cr multilayers

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The dependence of the giant magnetoresistance effect (GMR) on the interface structure in Fe/Cr multilayers was studied by magnetoresistivity and Mössbauer spectroscopy. The Fe/Cr multilayers consisting of Fe(6 nm)+[Cr(1.1 nm)/Fe(3 nm)]<sub>60</sub>+Cr(1.1 nm) were deposited by dc magnetron sputtering. Samples were annealed for 1 h at temperatures ranging from 200 to 500 °C. The interface structure was characterized by conversion electron Mössbauer spectroscopy (CEMS). Various different Fe sites: the bulk, step and perfect interface positions were identified. The evolution of the fraction of Fe atoms in different environments vs annealing temperature revealed that annealing at 300 °C induces bulk and in-plane diffusion of atoms. Higher annealing temperature causes substantial roughening of the interface related to a strong bulk diffusion of atoms. Microstructural changes observed in the CEMS spectra correspond well to the GMR measurements which reveal an enhancement of magnetoresistivity in Fe/Cr multilayers annealed at about 300 °C and its decrease due to deterioration of the interface at higher annealing temperatures. © 1999 American Institute of Physics. [S0021-8979(99)19608-7]

Since the discovery of the giant magnetoresistance (GMR) effect in Fe/Cr multilayers (MIs),<sup>1</sup> many experimental and theoretical studies have been performed to elucidate the origin of spin-dependent transport properties.<sup>1-6</sup> In particular, the influence of the interface roughness on the GMR amplitude was recognized. However, the correlation between GMR, magnetization and interface structure in Fe/Cr MIs<sup>4-8</sup> is still not well understood. Petroff *et al.*<sup>4</sup> have found that the Fe/Cr superlattices with low and weakly temperature-dependent resistivity have very small GMR that was related to sharp interfaces. They observed that annealing increased the residual resistivity as well as interface roughness and the scattering, i.e., roughening of the interfaces enhanced the GMR effect. The results of Fullerton *et al.*<sup>7</sup> have also shown that the GMR is enhanced by interface roughness. In contrast to these results the largest GMR effect (220% at 1.5 K) has been found in Fe/Cr multilayers with sharp interfaces and low residual resistivity.<sup>6</sup> The interface structure was usually investigated by x-ray diffraction (see, e.g., Refs. 5 and 7) which is suitable for analyzing long-range atomic order but which gives ambiguous information for the lateral scale of interfacial structure making the determination of the lateral length of roughness difficult. Rensing *et al.*<sup>8</sup> have performed a conversion electron Mössbauer spectroscopy (CEMS) study which demonstrated that the structural effect of annealing the Fe/Cr MIs is the promotion of Cr interdiffusion into the bulk of the Fe layers rather than short scale intermixing at the interfaces.

In this article, we present a systematic study of the correlation between the GMR, resistivity and interface structure, characterized by CEMS, for annealed Fe/Cr MIs obtained by the sputtering technique.

The Fe/Cr multilayers were deposited on oxidized Si substrates, attached to a computer controlled rotating platform, using dc magnetron sputtering.<sup>9</sup> The sputtering process raises a deposition temperature by about 50 °C from ambient. The deposition rate was monitored by quartz-crystal oscillators. A 6 nm thick iron layer was deposited first as a buffer layer, and a final 1.1 nm thick chromium layer was used as a capping layer. The structure of our samples is: SiO<sub>x</sub>+Fe(6 nm)+[Cr(1.1 nm)/Fe(3 nm)]<sub>60</sub>+Cr(1.1 nm). The multilayers were patterned into a conventional geometry for resistivity measurements (1 mm×7 mm) using photolithographic and lift-off techniques. The total film thickness was measured with a profilometer.

The as-deposited Fe/Cr multilayers were annealed for 1 h at temperatures ranging from 200 to 500 °C in an argon atmosphere under normal pressure. The structure of the as-deposited and annealed films was studied by CEMS which probes the surface region about 120 nm deep. The CEMS measurements were performed at room temperature by using a He-6%CH<sub>4</sub> gas flow electron counter. The Mössbauer spectral parameters (hyperfine fields, linewidths, isomer shifts, quadrupole splittings) were calculated by fitting Lorentzian line shapes to the experimental data by using the least-squares method. The isomer shifts are related to the  $\alpha$ -Fe standard.

The transport properties have been studied using a standard dc four terminal method. The current ( $i=5$  mA) was

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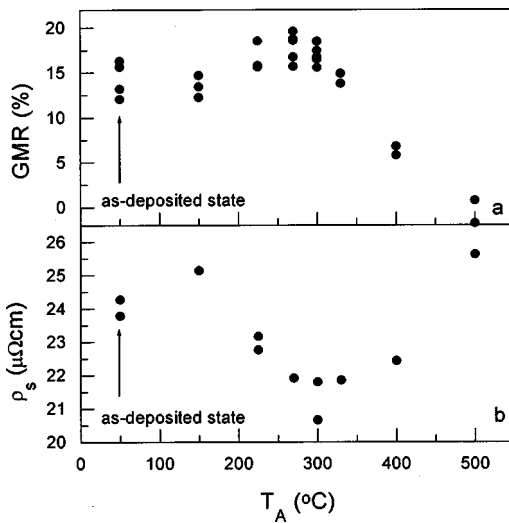


FIG. 1. (a) GMR and (b) residual resistivity  $\rho_s$ , measured at saturation field measured at 4.2 K vs annealing temperature, collected for four sample sets with the same nominal thicknesses of the Fe and Cr layers.

always in the plane of the sample and the magnetic field (up to 5 T) was parallel to the current. The magnetoresistivity and the resistivity were measured at 4.2 K before and after sequential annealing.

A standard vibrating sample magnetometer (VSM) was used to measure magnetisation loops at 4.2 K and the existence of an antiferromagnetic alignments of the adjacent Fe layers.

In Fig. 1 the GMR amplitude (for four sample sets with nominally the same sublayer thicknesses), defined as

$$\text{GMR} = \frac{\rho(H=0) - \rho_s(H=H_s)}{\rho_s(H=H_s)},$$

and resistivity  $\rho_s$ , measured in the plane of the samples at saturation field  $H_s$  at 4.2 K, are shown vs annealing temperatures  $T_A$ . As can be seen, the magnetoresistivity, after the initial increase observed for the samples annealed at 270 °C, is gradually reduced by the thermal treatment, although for samples annealed at 400 °C a considerable magnetoresistivity is still observed. Annealing at 500 °C destroyed the GMR effect; one sample showed only an anisotropic magnetoresistance effect of  $-1.7\%$  [Fig. 1(a)]. The residual resistivity  $\rho_s$ , determined by crystal imperfections, decreases for  $T_A < 300$  °C [Fig. 1(b)]. Therefore, if the interface scattering rather than the bulk one dominates in our multilayers, the minimum  $\rho_s(T_A)$  should be observed at  $T_A$  for which the interface roughness is smallest. Such a minimum in  $\rho_s(T_A)$  has been observed for  $T_A \approx 300$  °C. For  $T_A > 400$  °C, interdiffusion at the interfaces started to occur, resulting in a gradual increase of  $\rho_s$ . Atomic force microscopy (AFM) measurements of the films have shown a very smooth surface with a root mean square roughness  $h_{\text{rms}} \approx 0.7$  nm and the grain size  $D \approx 50$  nm. These values remained constant up to  $T_A \leq 400$  °C. For the MIs annealed at 500 °C, a considerable grain growth ( $D \approx 100$  nm) accompanied by the increase of roughness to about 3 nm was observed.

These results led us to the conclusion that the changes of the magnetoresistivity due to annealing can not be simply

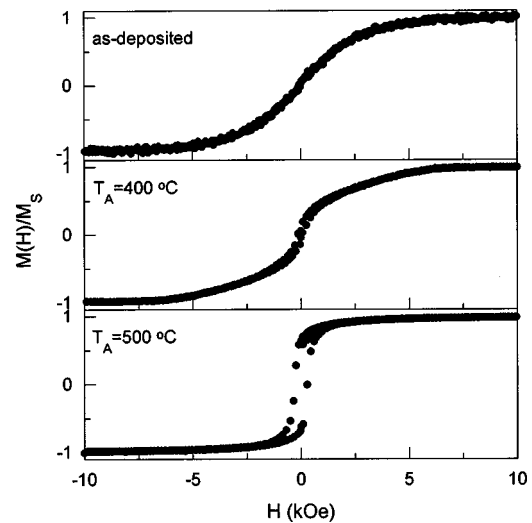


FIG. 2. Comparison of the magnetization loops measured at 4.2 K for Fe/Cr sample at different stages of annealing.

explained in terms of roughening of the interfaces by compositional mixing as a result of interdiffusion. If interdiffusion occurs in the multilayer structures, the residual resistivity of the films should markedly increase due to enhanced electron-impurity scattering. Therefore the reduction of the residual resistivity in the annealed samples can be attributed to the annealing effect in which some of the structural defects, such as vacancies, dislocations and grain boundaries, have been annihilated.

Figure 2 shows the examples of the evolution of the  $M(H)/M_s$  curves upon annealing for as-deposited and annealed at 400 and 500 °C samples. The remanent magnetization  $M_R$  for  $T_A < 400$  °C was negligible. At  $T_A = 400$  °C a small  $M_R$  appeared; however, the shape of the  $M(H)$  loop remained typical for antiferromagnetically coupled MIs. Annealing at 500 °C induced a significant increase of the coercivity and remanence. Since for  $T_A < 300$  °C  $M_R = 0$ , local ferromagnetic and/or biquadratic coupling (due to presence of pinholes) leading to a canted arrangement of the magnetization vectors in the adjacent Fe layers can be excluded. Therefore the increase of GMR amplitude observed for  $T_A \leq 300$  °C can be due to the annihilation of structural defects and/or reduction of the interface roughness leading to the increase of the mean free path of the conduction electrons. Such a conclusion is confirmed by the reduction of the residual resistivity, as noted previously for  $T_A \leq 300$  °C [Fig. 1(b)].

The interface structure of the Fe/Cr MIs was studied by the Mössbauer spectroscopy. The CEMS spectra for the as-deposited and annealed samples are shown in Fig. 3. The spectra, interpreted in terms of the Landes *et al.* model,<sup>10</sup> were fitted with four individual magnetic components, H1, H2, H3, and H4, associated with different iron positions. The parameters of the Mössbauer spectral components H1–H4, such as the hyperfine fields,  $H_{\text{hf}}$ , isomer shifts, IS, quadrupole splittings, QS, linewidths,  $\Gamma$ , and the line intensity ratios, D23, were similar in the CEMS spectra of all our samples and were the following: component H1: $H_{\text{hf}} = 32.9$  T,

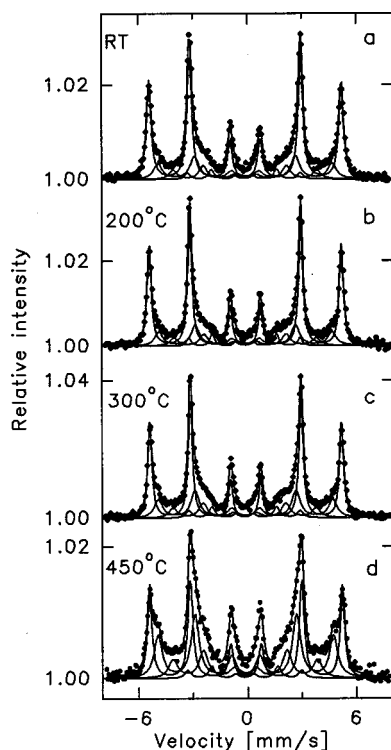


FIG. 3. (a) CEMS spectra recorded for the Fe/Cr MLs in the as-deposited state and after annealing at (b)  $T_A=200^\circ\text{C}$ , (c)  $300^\circ\text{C}$ , and (d)  $450^\circ\text{C}$ , respectively.

$IS=0.00$  mm/s,  $\Gamma\approx 0.3$  mm/s, corresponds to the iron atoms in bulk sites in Fe layer; component H2:  $H_{\text{hf}}\approx 30$  T,  $IS\approx 0.00$  mm/s,  $\Gamma\approx 0.5$  mm/s, and component H3:  $H_{\text{hf}}\approx 24$  T,  $IS\approx -0.05$  mm/s,  $\Gamma\approx 0.5$  mm/s, corresponds to Fe atoms in various step sites at the interface; component H4:  $H_{\text{hf}}\approx 19$  T,  $IS\approx -0.05$  mm/s,  $\Gamma\approx 0.4$  mm/s, corresponds to the Fe atoms in perfect interface positions. For all spectral components, the quadrupole parameter is negligible. The line intensity ratio D23 (the intensity of line 2 with respect to line 3) is close to 4, which suggests that the spins are aligned preferentially in the plane of the film. In the earlier studies of Fe/Cr multilayers<sup>8,9</sup> in addition to magnetic components a paramagnetic quadrupole doublet, corresponding to isolated Fe atoms in Cr layers, was observed. However, in our present study such a paramagnetic component was not detected (Fig. 3).

Assuming the Mössbauer recoilless fractions were the same for various Fe sites, the spectral area of each component is equal to the atomic fraction of Fe occupying a given site. A summary of the atomic fractions of Fe in a given environment, calculated from CEMS spectra in Fig. 3, is shown in Fig. 4. As can be seen, the spectral component H1 related to the bulk Fe sites strongly dominates in the as-deposited sample. The fraction of Fe atoms at a perfectly flat interface (H4) is only about 5.5%. Low temperature anneal-

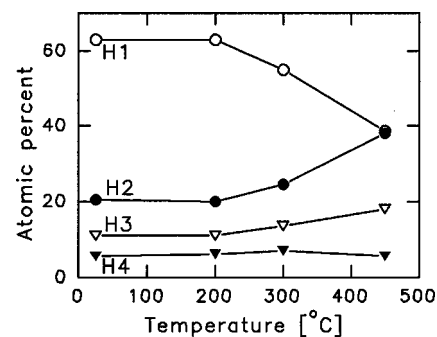


FIG. 4. Atomic fractions of Fe in a given site: bulk (H1), step (H2 and H3), and perfect interface (H4), vs annealing temperature, calculated from CEMS spectra.

ing ( $T_A=200^\circ\text{C}$ ) does not affect markedly the interface structure. Annealing at  $300^\circ\text{C}$  causes a significant increase of the Fe step sites and a small, but clear increase of the perfect interface sites, as shown by the increase of the H2, H3, and H4 components at the expense of the H1 one [Fig. 3(c) and Fig. 4]. High temperature annealing ( $T_A=450^\circ\text{C}$ ) induces strong roughening of the interface. The fraction of Fe atoms at various step sites increases substantially as evidenced by a strong increase of H2 and H3 components [Fig. 3(d), Fig. 4]. Simultaneously, the H4 component decreases which shows that the perfectly flat interface deteriorates.

Such changes in the microstructure of the Fe/Cr interface suggest that two diffusion processes occur during annealing: one related to the diffusion of atoms in the plane of the interface leading to its smoothing, and the other related to the volume atomic diffusion which causes roughening of the interface. At annealing temperatures of about  $300^\circ\text{C}$  both processes occur. At higher annealing temperatures ( $T_A\approx 450^\circ\text{C}$ ), a bulk diffusion strongly dominates and leads to a significant roughening of the interface.

<sup>1</sup>M. N. Baibich, J. M. Broto, Á. Fert, F. Nguyen Van Dau, F. Petroff, P. Eitenne, G. Creuzet, A. Friederich, and J. Chazelas, *Phys. Rev. Lett.* **61**, 2472 (1988).

<sup>2</sup>R. E. Camley and J. Barnas, *Phys. Rev. Lett.* **63**, 664 (1989); see also J. Barnas, A. Fuss, R. E. Camley, P. Grünberg, and W. Zinn, *Phys. Rev. B* **42**, 8110 (1990).

<sup>3</sup>P. M. Levy and S. Zhang, *Phys. Rev. Lett.* **65**, 1643 (1990).

<sup>4</sup>F. Petroff, A. Barthelemy, A. Hamzic, A. Fert, P. Etienne, S. Lequien, and G. Creuzet, *J. Magn. Magn. Mater.* **93**, 95 (1991).

<sup>5</sup>J. M. Colino, I. K. Schuller, V. Korenivski, and K. V. Rao, *Phys. Rev. B* **54**, 13030 (1996).

<sup>6</sup>R. Chad, C. D. Potter, P. Belien, G. Verbanck, V. V. Moshchalkov, and Y. Bruynseraede, *Appl. Phys. Lett.* **64**, 3500 (1994).

<sup>7</sup>E. Fullerton, D. M. Kelly, J. Guimpel, I. Schuller, and Y. Bruynseraede, *Phys. Rev. Lett.* **68**, 859 (1992).

<sup>8</sup>N. M. Rensing, B. M. Clemens, and D. L. Williamson, *J. Appl. Phys.* **79**, 7757 (1996).

<sup>9</sup>T. Luciński, D. Elefant, G. Reiss, and L. van Loyen, *Phys. Status Solidi A* **160**, 133 (1997).

<sup>10</sup>J. Landes, Ch. Sauer, R. A. Brand, W. Zinn, S. Mantl, and Zs. Kajcsos, *J. Magn. Magn. Mater.* **86**, 71 (1990).