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Sputtering pressure effect on the giant magnetoresistance of Fe/Cr superlattices

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Abstract

We have studied the magnetic, structural and transport properties of sputtered Fe/Cr(1 1 0) superlattices as a function of the Ar pressure during the deposition. A systematic increase in the roughness is induced, as shown by low-angle X-ray diffraction measurements. As the Ar pressure becomes higher, it is also found an increase in the electrical resistivity, related with internal stresses, and a reduction in the antiferromagnetic coupling between the layers. However, the giant magnetoresistance ($\Delta\rho$), normalized by the antiferromagnetically coupled fraction of the sample, increases for the highest Ar pressures, implying that the spin-dependent scattering mechanisms are enhanced by roughening of the interfaces. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

Since the discovery of giant magnetoresistance (GMR) in magnetic/normal superlattices [1] much interest has been devoted to the understanding of this phenomenon, because of the possible applications in artificially tailored magnetic materials. Already in the first work, Baibich et al. [1] pointed at the two most important ingredients in giant magnetoresistance: the existence of antiferromagnetic coupling between the magnetic layers and the spin-dependent scattering of the conduction electrons, as it has been widely analyzed both from the theoretical [2–6] and experimental [7–10] points of view. Of particular interest in the Fe/Cr system is the relation between GMR and superlattice structure; theoretical models [4,6] have suggested the importance of spin-dependent scattering at the magnetic/nonmagnetic interfaces, and there is a growing experimental evidence [8,11–14] of the major role of interface scattering in the GMR of Fe/Cr superlattices. However, the influence of roughness in the magnetotransport of these multilayers is still an open question and there are conflicting experimental reports on the subject

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[8,14,15], since structural properties strongly depend on growth method and growth parameters [16]. Therefore, it is desirable to perform controlled experiments in which the deposition conditions are varied in a systematic way and correlated with the magnetic, transport and structural properties of the multilayers. In this work, we have studied the effect of the structural changes induced by increasing the Ar pressure during the deposition on the GMR of sputtered Fe/Cr(1 1 0) superlattices. The results indicate a progressive roughening of the interfaces that enhances the spin-dependent scattering mechanisms, once that the changes in antiferromagnetic coupling and scattering by bulk imperfections are taken into account.

2. Experiments

Fe/Cr(1 1 0) superlattices have been grown by DC sputtering on Si(1 0 0) substrates at ambient temperature, as has already been reported [8]. The sample structure has been characterized by low-angle and high-angle X-ray diffraction using a Rigaku rotating anode diffractometer with Cu K_α radiation. The in-plane magnetization was measured by a SQUID magnetometer at 10 K. Four lead magnetotransport measurements were performed in a helium cryostat at 10 K and in magnetic fields up to 5 T, parallel to the plane of the films and perpendicular to the current.

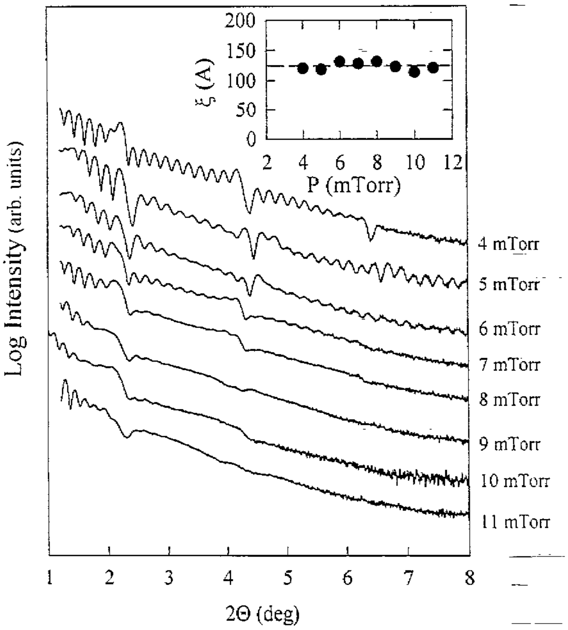

Fig. 1. Low-angle X-ray θ–2θ scans of a series of [Fe(30 Å)/Cr(12 Å)]$_{10}$ superlattices, grown at different Ar pressure. The spectra are offset for clarity. Inset shows the pressure dependence of the crystalline coherence length calculated from the width of the high-angle (1 1 0) peak. Dashed line indicates the average value of 122 Å.

Fig. 1 presents the X-ray low-angle θ–2θ scans for a series of [Fe(30 Å)/Cr(12 Å)]$_{10}$ superlattices, where the subindex indicates the number of bilayers. The deposition conditions are the same in all the samples, except the Ar pressure that has been varied in a systematic way in the range 4–11 mTorr. For the samples grown at low sputtering pressure clear superlattice peaks are present up to the second order (and a shoulder at the third order), together with finite-size effect oscillations up to $2\theta \approx 7$–8° due to the total thickness of the multilayers. As the sputtering pressure is increased these features become broadened and less well defined and, finally, for the highest pressures there is only a shoulder at the first-order superlattice peak position and low-angle finite-size peaks can only be seen up to $2\theta \approx 2^\circ$. These changes in the X-ray spectra indicate an increasing roughness at the interfaces and also the loss of coherence between the top and bottom film surfaces [17] as a function of the sputtering pressure during deposition. On the other hand, high angle X-ray diffraction shows that these samples grow with an overall (1 1 0) orientation and that the crystallinity of the films remains constant in the whole Ar pressure range investigated. This fact can be appreciated in the inset of Fig. 1 where the structural coherence length ξ, calculated from the width of the high-angle (1 1 0) reflection using Sherrer's equation, is plotted versus the Ar pressure. The coherence length perpendicular to the film direction is $\xi = 122 \pm 7$ Å, approximately independent of the pressure. This is in contrast with previous studies, in which the interface roughness was modified either by high-temperature annealing [18] or by ion irradiation [13] giving rise to an enhanced bulk crystallinity. In the following, the magnetic and transport properties of these multilayers will be analyzed in detail in order to get further insight into the influence of the interface roughness on GMR, taking into account the observed trends in their structural properties.

3. Results and discussion

Fig. 2a shows the field dependence of the magnetoresistance ($\Delta\rho(H) = \rho(H) - \rho_S$, where ρ_S is the resistivity at saturation) for two [Fe(30 Å)/Cr(12 Å)]$_{10}$ multilayers grown at different Ar pressures. The Cr thickness (t_{Cr}) has been selected at 12 Å, because it corresponds to the position of the first peak in the magnetoresistance and antiferromagnetic coupling [8] and in this way the highest value of GMR can be obtained. Another reason to choose this t_{Cr} value is in order to minimize the effect of possible changes in the GMR versus t_{Cr} curves, that may be caused by the local thickness variations present in the rougher samples. This would produce a broadening of the antiferromagnetic coupling peaks with different growth conditions, as reported by Rensing et al. [14], but will have little effect at the chosen $t_{Cr} = 12$ Å due to the reduced slope of the GMR versus t_{Cr} curve at the maximum.

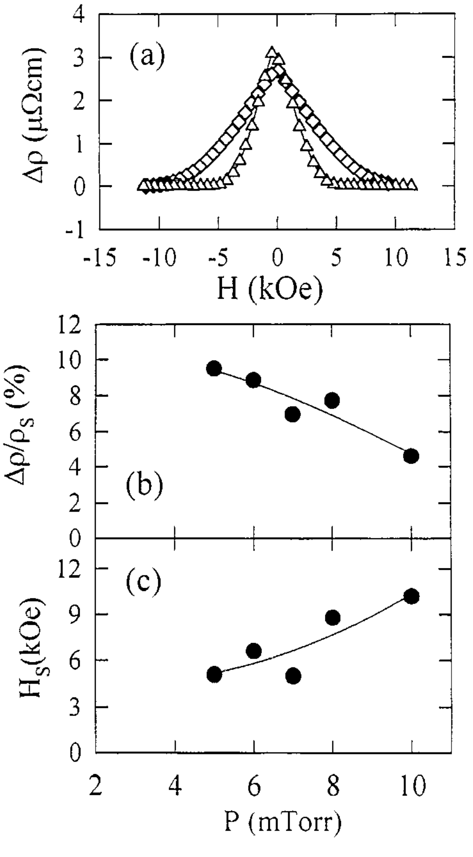

Fig. 2. (a) Magnetoresistance versus field curves for two [Fe(30 Å)/Cr(12 Å)]$_{10}$ superlattices grown at 10 mTorr (◇) and 5 mTorr (△); ($\Delta\rho = \rho - \rho_S$, where ρ_S is the saturation resistivity). (b) Magnetoresistance ratio $\Delta\rho/\rho_S$ and (c) saturation field H_S as a function of the sputtering pressure for a series of multilayers with the same Fe and Cr thicknesses as in (a). Line is a guide to the eye.

Some general trends can be observed in this Fig. 2a: first, the total change in resistivity of the multilayers as a function of magnetic field ($\Delta\rho = \rho_{max} - \rho_S$, where ρ_{max} is the maximum value of the resistivity) depends weakly on the sputtering pressure and, second, the magnetoresistance versus field curves become broader for increasing roughness in the samples. A more quantitative analysis is presented in Fig. 2b and Fig. 2c, where the pressure dependence of the magnetoresistance ratio ($\Delta\rho/\rho_S$) and of the saturation field (H_S) are shown. A reduction in the magnetoresistance ratio is found for the samples grown at the highest Ar pressure, whereas the saturation field increases. This second fact could be an indication of an enhanced antiferromagnetic coupling between the Fe layers, since higher fields are needed to achieve complete saturation of the samples; however, an enhanced coupling should improve the GMR, and this is opposite to the observed decrease in $\Delta\rho/\rho_S$.

To further clarify this question and obtain a better picture of the roughness influence on spin-dependent scattering and GMR, it is interesting to study the behavior of the saturation resistivity ρ_S and, in this way, separate the magnetoresistive contribution from the influence of the sample crystalline quality in the scattering processes. As it can be seen in Fig. 3a, ρ_S varies from 33 to 60 μΩ cm when the deposition Ar pressure is raised from 5 to 10 mTorr. Note that the slope of the ρ versus T curves is essentially the same in all the pressure range studied, $d\rho/dT \approx 0.055 \pm 0.005$ μΩ cm/K, so the observed changes correspond mainly to the temperature-independent part of the resistivity,

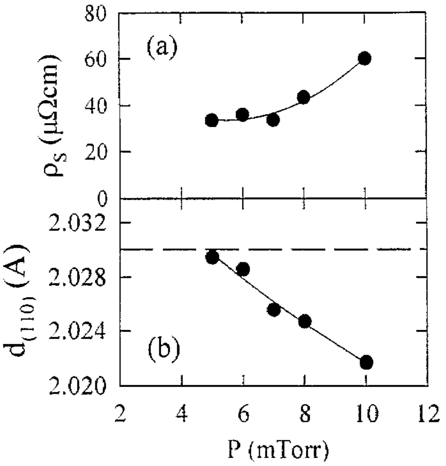

Fig. 3. Pressure dependence of (a) the saturation resistivity and (b) the perpendicular lattice spacing $d_{(1\,1\,0)}$ for a series of $[\mathrm{Fe}(30\,\text{Å})/\mathrm{Cr}(12\,\text{Å})]_{10}$ superlattices. The dashed line in (b) indicates the ideal lattice spacing calculated from the bulk values of Fe and Cr. Solid line is a guide to the eye.

attributed to scattering by crystal imperfections. Therefore, this remarkable enhancement in the saturation resistivity should be related either to an increase in interface scattering by the rougher interfaces or to a higher degree of scattering by bulk disorder. A possible source of bulk scattering due to crystalline imperfections could be the presence of internal stresses in the samples, induced by the growth conditions. Fig. 3b shows the pressure dependence of the perpendicular lattice spacings $d_{(1\,1\,0)}$ of these Fe/Cr superlattices, obtained from the position of the high-angle X-ray peak. It can be clearly seen that for a multilayer grown at 5 mTorr this value is very similar to the ideal $d_{(1\,1\,0)} = 2.030$ Å (calculated for a superlattice with $t_{\mathrm{Fe}} = 30$ Å and $t_{\mathrm{Cr}} = 12$ Å, and assuming the bulk lattice spacings of Fe and Cr as 2.027 and 2.039 Å, respectively), but that it becomes smaller when the pressure is increased. This evolution of the lattice spacings is indicative of the existence of internal stresses and could be correlated with the resistivity curve shown in Fig. 3a. It is also worth noticing that the resistivity of single Fe films, grown in the same conditions as the multilayers, varies from 24 μΩ cm at 6 mTorr to 43 μΩ cm at 10 mTorr which is an almost twofold increase very similar to the multilayers. Therefore, the behavior of the saturation resistivity in these superlattices seems to be dominated by the increase in the spin-independent bulk scattering mechanisms, mainly related with these internal stresses. So, to characterize the GMR the total change in resistivity $\Delta\rho$ may be a better choice than $\Delta\rho/\rho_{\mathrm{S}}$.

Another fact that could be affecting the magnitude of GMR in these Fe/Cr superlattices could be the degree of antiferromagnetic coupling as the interfaces become rougher. Fig. 4a presents the hysteresis loops of two superlattices grown at 4 mTorr (hollow symbols) and 9 mTorr (filled symbols). The shape of the loop is clearly modified by the sputtering pressure during growth. In Fig. 4b the pressure dependence of the remanent magnetization M_{R}, normalized by the saturation magnetization M_{S}, is plotted for two series of Fe/Cr superlattices with $t_{\mathrm{Cr}} = 12$ and 15 Å. This quantity gives a first estimate of the fraction of the sample which is not antiferromagnetically aligned in zero

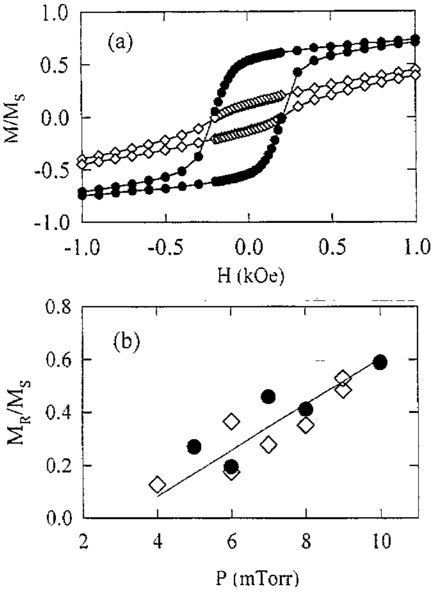

Fig. 4. (a) Hysteresis loops for two $[\mathrm{Fe}(30\,\text{Å})/\mathrm{Cr}(12\,\text{Å})]_{10}$ superlattices grown at 4 mTorr (◇) and 9 mTorr (●), (b) Remanent magnetization normalized by the saturation magnetization, $M_{\mathrm{R}}/M_{\mathrm{S}}$, for a series of: (●) $[\mathrm{Fe}(30\,\text{Å})/\mathrm{Cr}(12\,\text{Å})]_{10}$ and (◇) $[\mathrm{Fe}(30\,\text{Å})/\mathrm{Cr}(15\,\text{Å})]_{10}$ superlattices as a function of the sputtering pressure. The line is a guide to the eye.

field. It clearly becomes higher as the Ar pressure is raised for both series of samples. This remanent magnetization may arise from ferromagnetically coupled regions, suggesting an increasing number of shorts or pinholes between the ferromagnetic layers. Also, parts of the sample coupled through biquadratic exchange can result in a non-zero remanence [19]. These non-collinearly aligned regions would be favored by the interlayer thickness fluctuations resulting from the increased roughness [20,21]. It should be stressed that, in general, this loss of antiferromagnetic coupling is not a direct consequence of the roughening of the interfaces. For example, it is interesting to note that similar effects of degradation in the antiferromagnetic coupling for high Ar pressure have been found in Fe/Cr(1 0 0) superlattices grown on MgO, but in which the quality of the interfaces remained essentially the same in the whole pressure range 4–11 mTorr [16]; also, in other studies of ion irradiated Fe/Cr(1 1 0) samples, a monotonous increase in M_R was observed while the samples roughness changed in a non-monotonic way [13].

This loss of antiferromagnetic coupling must be effectively reducing the observed GMR. In order to characterize properly the effect of the enhanced roughness on the spin-dependent scattering mechanisms, the changes in resistivity should be normalized by the fraction of the sample that remains antiferromagnetically coupled at each pressure. The graph of this normalized magnetoresistance can be seen in Fig. 5, where a clear correlation is found for two different Cr thickness; the higher the Ar pressure, the higher the magnetoresistance or, in other words, the increase in the roughness at the Fe/Cr interfaces due to the growth conditions is inducing an enhancement of the spin-dependent scattering.

This effect of roughness on GMR is in agreement with the leading role of interface scattering in the magnetotransport properties of the Fe/Cr system [11–13]. However, the influence of roughness on the spin-dependent scattering mechanism is still controversial and there are reports of both enhancement [8,13,18] and degradation [14,15] on GMR upon increasing interface roughness and interdiffusion. The results presented in Fig. 5, in comparison with the behavior shown in Fig. 2, stresses the need of careful examination of the whole magnetic and transport properties of the multilayers; for example, some reports of reduction in GMR with increasing roughness do not take into account the evolution of the antiferromagnetic coupling [14]. It is worth noting that this observed enhancement of the spin-dependent scattering mechanisms for a systematic increase in the roughness is in good agreement with previous experimental work on this subject, where roughness was modified by different growth conditions [8] and after-deposition processes [13,18] in sputtered Fe/Cr superlattices. These observed changes in the spin-dependent scattering at the interfaces shown in Fig. 5 could be related with an increasing intermixing of Fe and Cr, as the interdiffused atoms could act as asymmetric spin-dependent scattering centers [22].

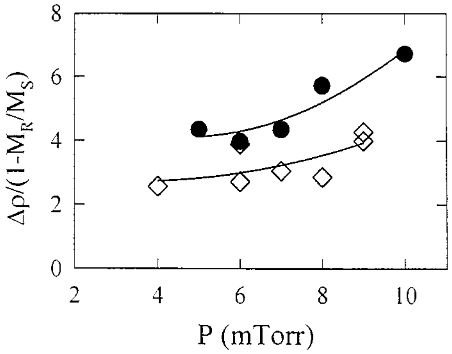

Fig. 5. Giant magnetoresistance normalized by the antiferromagnetically coupled fraction of the sample, $\Delta\rho/(1 - M_R/M_S)$ versus the sputtering pressure for a series of: (●) [Fe(30 Å)/Cr(12 Å)]$_{10}$ and (◇) [Fe(30 Å)/Cr(15 Å)]$_{10}$ superlattices. Solid lines are a guide to the eye.

Theoretical calculations of the effect of interface quality on GMR have shown that any increase in the asymmetry of spin-dependent scattering by the roughness present in the sample leads to an increase on GMR [6]. In this way, it has been proposed that an increase in the strength of correlated roughness would enhance GMR and, also, an enhancement in the diffuse interface scattering could improve GMR [2,23], such as would be caused by an increase in interdiffusion of Fe and Cr atoms.

4. Conclusions

In summary, the systematic study of the effect of the sputtering Ar pressure on the structural properties and GMR of Fe/Cr(1 1 0) superlattices has revealed the relation between interface roughness and spin-dependent scattering in this system. The changes in the Ar pressure during the deposition result in a progressive roughening of the interfaces, while the crystalline coherence remains constant. This increase in disorder at high deposition pressure induces an enhancement of the saturation resistivity, related with internal stresses, and a reduction in the antiferromagnetic coupling. The magnetoresistance, normalized by the antiferromagnetically coupled fraction of the sample, is enhanced at high Ar pressure, indicating the increase in the spin-dependent scattering at the rougher interfaces.

Acknowledgements

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