Giant magnetoresistance in Fe/Cr superlattices with very thin Fe layers

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Carefully tailored Fe/Cr epitaxial superlattices with extremely thin Fe layers have been grown on MgO(100) by molecular beam epitaxy. The low-angle x-ray spectra reveal the presence of sharp interfaces down to an Fe layer thickness of a few monolayers. An $[Fe(4.5\ Å)/Cr(12\ Å)]_{50}$ superlattice shows a 220% magnetoresistance at 1.5 K, and a saturation field of 110 kOe. A further decrease of the Fe layer thickness produces a drastic decrease in the magnetoresistance.

The discovery of the giant magnetoresistance (GMR) in Fe/Cr superlattices[1] opened a new field of possible applications of artificially tailored materials.[2,3] The GMR is closely related to the antiferromagnetic (AF) exchange coupling of Fe layers through the nonmagnetic spacer layer.[4–6] The parallel alignment of the magnetization of the Fe layers forced by an externally applied magnetic field changes the scattering probabilities of spin-up and spin-down electrons and produces a decrease of the resistivity.[1,7–9]

Besides atomic potential scattering, which increases only the overall resistivity, two locations of the magnetic electron scattering have to be considered: the scattering inside the individual layers (in the following referred as bulk scattering) and scattering at the interface between the Fe and Cr layers (interface scattering). These two contributions influence the MR effect in a different way. An enhancement of the bulk scattering produces only a small increase of the magnetoresistance amplitude.[7] However, experimental studies[11,12] and theoretical models[8–10] emphasize the importance of the magnetic scattering processes at the interfaces. The mechanisms involved are however still subject to discussion since the scattering processes may lead to an increase[11] or a decrease[12] of the MR amplitude. On the other hand, the higher MR values reported for Fe/Cr multilayers[13–15] were all found in epitaxial superlattices showing a high degree of interface perfection. The most promising substrate among the ones which were used in these studies is MgO(100),[15] which shows no intermixing with the Fe/Cr superlattice and consequently provides a small bulk defect density.

In order to prepare Fe/Cr superlattices with a high MR the following steps are important: (i) decrease the bulk resistivity in order to increase the influence of the interface scattering which governs the MR;[8,9] (ii) increase the number of bilayers in order to reduce the influence of the outer surfaces of the superlattice and thus enhance the MR;[9] (iii) reduce the thickness of the Fe layers (t_{Fe}), since the bulk of the Fe layers do not contribute to the MR.[7] The first point can be met by growing epitaxial superlattices from ultrapure target materials. The second is just a matter of experimental skill and patience. The third point, a substantial reduction of t_{Fe}, requires sharp interfaces. Finally, the thickness of the Cr layers (t_{Cr}) must be constant and equal to the thickness value ($t_{Cr}=12$ Å) which ensures a strong AF coupling.

In this letter we show that it is possible to grow epitaxial Fe/Cr superlattices with an Fe layer thickness down to three monolayers without losing the high degree of interface perfection. By carefully adjusting the preparation conditions, using a MBE system, and by increasing the number of bilayers, it is possible to obtain GMR values of 220% as well as high saturation fields of $H_s=110$ kOe.

The Fe/Cr superlattices were prepared in a Riber MBE deposition system (2×10^{-11} mbar base pressure) equipped with two *e*-beam evaporators. The rate is stabilized within 1% by a homemade feedback control system using two Balzers quadrupole mass spectrometers (QMS). Integration of the QMS signal is used to control the shutters of the individual evaporation sources. The Fe and Cr layers (starting materials of 99.996% purity) are deposited with a rate of 1 Å/s onto a 50-Å-thick Cr seed layer which covers a single crystalline MgO(100) substrate held at 50 °C. In this report the notation $[Fe(k)/Cr(l)]_n$ is used where k and l are respectively the thickness of the Fe and the Cr layers in angstroms and n is the number of bilayers.

In situ reflective high energy electron diffraction (RHEED) measurements are used to monitor the quality of the superlattices during growth. *Ex situ* x-ray diffraction studies at both low angle (LA) and high angle (HA) have been utilized to determine the structural and layering quality. The magnetoresistance is measured at 1.5 and 300 K by the "van der Pauw" four-point probe method using a temperature controlled cryostat equipped with a 15-T superconducting magnet. The magnitude of the MR is defined as the ratio $\Delta\rho/\rho_s=(\rho_0-\rho_s)/\rho_s$ with ρ_0 the resistivity at $H=0$ kOe, and ρ_s the resistivity at the saturation field H_s. The direction of the applied magnetic field is in the film plane.

Figure 1(a) shows the LA XRD spectra of two Fe/Cr superlattices. The well pronounced structure up to 12° in 2θ shows that even the Fe(4.5)/Cr(12) superlattice has a well defined layering structure with sharp interfaces. Indeed we note that in superlattices with identical interface quality but varying bilayer thickness, not the number of superlattice peaks, but the angle at which the highest-order superlattice peak appears, is important to assess the quality of the superlattice. This is illustrated in Fig. 1(b), where LA XRD spectra have been simulated (SUPREX program[16]) for Fe/Cr superlattices with identical interface quality (0.05 atomic layer variation of the layer thickness) and different t_{Fe}. The fact that superlattice structure is more pronounced in the measured spectra [Fig. 1(a)] than in the simulation [Fig. 1(b)] may be

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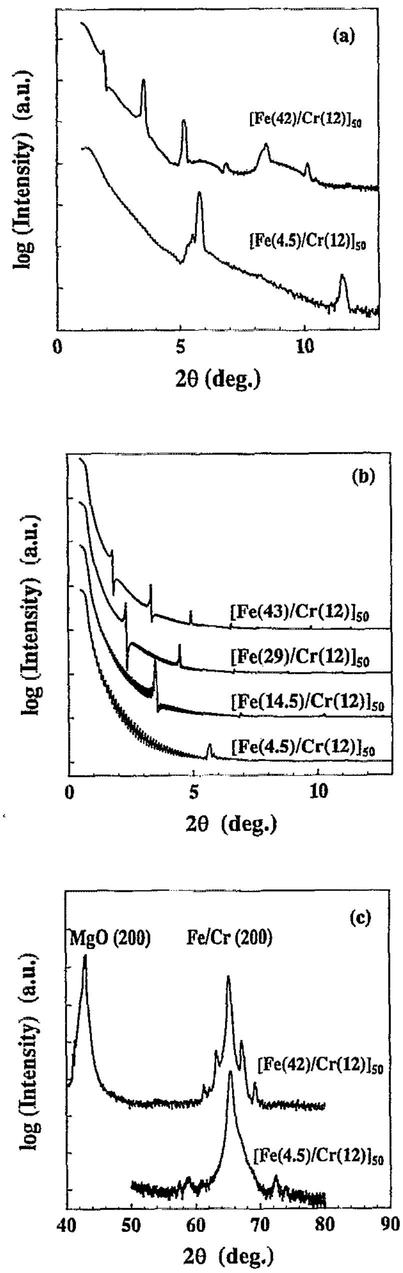

FIG. 1. (a) Experimental and (b) simulated low-angle spectra of different Fe/Cr superlattices. The set of simulated spectra (b) is calculated for different Fe layer thicknesses while all other parameters have been kept constant. The high-angle x-ray spectra (c) show the Fe/Cr(200) peak at 64.7° in 2θ and no indications of any minority phases. The spectra are offset for clarity.

due to some strain in the superlattice caused by the epitaxial registry producing a larger electron contrast.

The HA XRD spectra [Fig. 1(c)] shows the crystallographic (100) orientation of these epitaxial layers as well as the clearly distinguishable satellite peaks around Fe/Cr(200). These satellites are characteristic for sharp interfaces.

Besides good layering quality of the superlattices, it is also important to minimize the electron scattering in the bulk of the layers. In order to determine the defect density in the bulk we performed electrical transport measurements in epitaxial (100) oriented single layers of Fe and Cr with different thicknesses. Applying the Fuchs–Sondheimer theory,[17,18] the extrapolated bulk values of the resistivity at 4.2 K are respectively $\rho(\mathrm{Fe}) \approx 0.2\ \mu\Omega$ cm and $\rho(\mathrm{Cr}) \approx 0.35\ \mu\Omega$ cm. These values are much lower than the total resistivity of the superlattices, $\rho_s \approx 15\ \mu\Omega$ cm, indicating that the interface scattering is dominating the transport properties.

The magnetoresistance curves of a [Fe(4.5)/Cr(12)]$_{50}$ superlattice measured at 1.5 and 300 K, respectively, are shown in Fig. 2(a). To our knowledge, the $\Delta\rho/\rho_s = 220\%$ value is the highest MR measured in a magnetic superlattice.[13–15] Even at 300 K the MR is still 42%. The resistivities are respectively $\rho_0 = 50.4\ \mu\Omega$ cm and $\rho_s = 15.8\ \mu\Omega$ cm at 1.5 K and $\rho_0 = 62.3\ \mu\Omega$ cm and $\rho_s = 43.7\ \mu\Omega$ cm at 300 K. Very remarkable is the fact that this high MR is observed in a sample where the individual Fe layers consist of only three monolayers, from which at least two are in contact with Cr. This leads to a change in the magnetic properties of the Fe atoms as measured by conversion electron Mössbauer spectroscopy.[19] Accordingly, each Fe layer of this superlattice contains only one monolayer of Fe which has a complete Fe nearest-neighbor shell.

Surprisingly large is the value of the saturation field $H_s = 110$ kOe which is much higher than the saturation fields usually observed in superlattices ($H_s \approx 10$ kOe)[1] or in a Fe/Cr/Fe trilayer system ($H_s \approx 0.5$ kOe).[20] We note that the

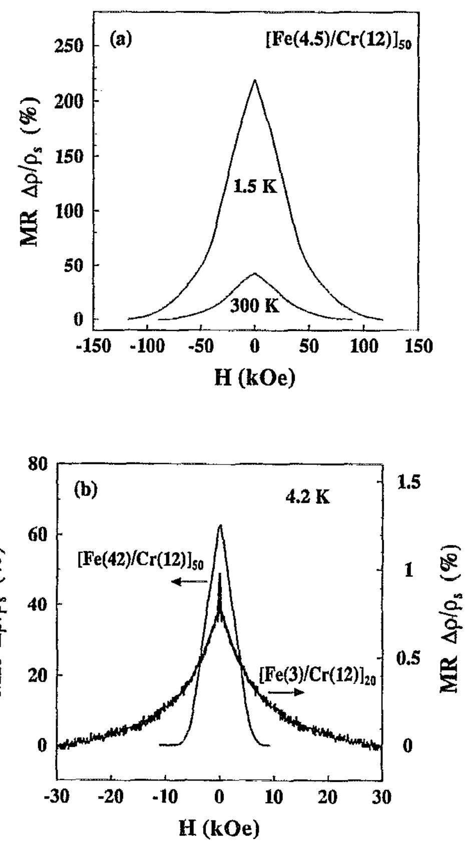

FIG. 2. The magnetoresistance $\Delta\rho/\rho_s$ of different Fe/Cr superlattices as a function of the magnetic field applied in the film plane. The [Fe(4.5)/Cr(12)]$_{50}$ superlattice (a) shows a record value of $\Delta\rho/\rho_s = 220\%$ at 1.5 K and 42% at 300 K. Increasing or decreasing the Fe layer thickness (b) produces a decrease of the MR and H_s amplitudes.

saturation field is related to the exchange coupling strength (J), the thickness of the Fe layers (t_{Fe}) and the saturation magnetization (M_s) according to the relation $H_s \sim J/(M_s t_{Fe})$.[6] Assuming a constant J, an increase of the Fe layer thickness by a factor ten should produce a decrease of H_s by the same factor, which is indeed observed for the $[Fe(42)/Cr(12)]_{50}$ superlattice [Fig. 2(b)]. Moreover, H_s increases with the number of bilayers.[21] The difference between the saturation fields of the $[fe(42)/Cr(12)]_{50}$ superlattice (H_s=8 kOe) and the Fe/Cr/Fe trilayers with the same Fe layer thickness (H_s=0.5 kOe)[20] is much higher than the influence of the number of bilayers on H_s, as reported by Parkin *et al.*[21] It might be that the different crystallographic orientation (single crystalline versus polycrystalline) plays a role in the determination of H_s.

The reduction of the MR upon increasing the Fe layer thickness is in qualitative agreement with its dependence on t_{Fe}, as calculated for spin-dependent interface scattering.[7] A decrease of the Fe layer thickness below three monolayers causes a drastic decrease of the MR amplitude, as shown in Fig. 2(b) for a $[Fe(3)/Cr(12)]_{20}$ superlattice. It is probable that the magnetic ordering of the superlattice is lost which leads to the absence of the AF coupling. The small MR can be due to other interaction mechanisms, such as electron localization.[22]

Excellent Fe/Cr superlattices have been grown with Fe layer thicknesses down to three monolayers. Record magnetoresistance and saturation field values have been measured. Further studies are needed to explain the influence of the superlattice periodicity on the magnetic coupling strength of these layers.

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