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Analysis of the transport properties of epitaxial Fe and Cr films

R Schad†§, P Beliën‡‖, G Verbanck‡, V V Moshchalkov‡ and Y Bruynseraede‡

† Research Institute for Materials, KU Nijmegen, NL-6525 ED Nijmegen, The Netherlands
‡ Laboratorium voor Vaste-Stoffysika en Magnetisme, KU Leuven, B-3001 Leuven, Belgium

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Abstract. Molecular beam epitaxy of Fe and Cr on single crystalline MgO(001) substrates produces films with very small defect concentrations. Accordingly, their low temperature transport properties are characterized by extremely low bulk resistivities. The temperature dependence of the resistivity of the Cr films shows anomalies due to magnetic ordering effects around the Néel temperature T_N. We find values of $T_N = 300$ K which is close to the bulk value. The anomalous Hall effect of the Fe films shows typical ferromagnetic behaviour. The spontaneous Hall coefficient is proportional to the square of the resistivity indicative of side jump scattering. Fe/Cr superlattices grown under similar conditions are particularly suitable to study the influence of interface scattering on the giant magnetoresistance amplitude.

1. Introduction

The electrical transport properties of thin metallic films are strongly influenced by electron surface scattering. This surface contribution becomes predominant with decreasing film thickness or decreasing defect density inside the film. The film thickness dependence of the resistivity was first formulated phenomenologically by Fuchs [1, 2] who introduced a specularity parameter p which is the fraction of electrons elastically scattered at the surfaces. This theory was further developed and refined taking into account other contributions [3–5] or using a quantum mechanical approach [6]. In most cases the theory of Fuchs is applied [7–9] which, although providing only a phenomenological description of the surface scattering, has the advantage of simplicity and allows the straightforward extraction of the transport parameters.

Recently, renewed interest in the influence of surface scattering on the transport properties of metallic films has arisen from the importance of interface scattering in superlattices. An important example is the giant magnetoresistance (GMR) effect of magnetic superlattices [10, 11] which is governed by the spin dependent electron scattering [12]. However, the dependence of the GMR amplitude on the interface quality is not fully understood. Although it was theoretically shown that a certain amount of interface roughness is necessary to obtain a high GMR amplitude contradictory experimental results have been reported [13–16].

§ Corresponding author: Dr Rainer Schad, Research Institute for Materials, Katholieke Universiteit Nijmegen, Toernooiveld 1, NL 6525 ED Nijmigen, The Netherlands. Tel.: +31 (0)24 3653094. Fax: +31 (0)24 3652190. E-mail address: schad@sci.kun.nl.
‖ Present address: Philips Optical Storage, Kempische Steenweg 293, B-3500 Hasselt, Belgium.

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A usual experimental complication is that a variation of the preparation parameters (sputtering gas pressure, substrate temperature during deposition, deposition rate, substrate structure and roughness etc) not only influences the interface quality but also changes the bulk properties. In the case where bulk scattering is important the separation of bulk and interface contributions to the electron scattering will be difficult [17]. In such cases the relation between GMR amplitude and interface structure will be dubious. Therefore, it is important to use optimized deposition techniques and substrates to grow Fe and Cr films with negligible bulk scattering. Epitaxial Fe films grown on GaAs or textured films on Corning glass show rather high resistivities [8, 18] making them unsuitable for such studies. However, epitaxial Fe and Cr films on MgO(001) are characterized by a very small bulk defect densities [19].

In this paper we present a detailed analysis of the electrical transport properties of high purity epitaxial Fe and Cr films in dependence of thickness, temperature and magnetic field. This reveals valuable information about the competition between surface and bulk scattering, magnetic ordering effects and specific surface scattering mechanisms.

2. Experiment

Fe and Cr films were prepared in a Riber MBE deposition system (2×10^{-11} mbar base pressure) using two e-beam guns. The evaporation rate of 0.1 nm s^{-1} was stabilized within 1% by a home made feedback control system using Balzers quadrupole mass spectrometers (QMSs). The starting material (99.996% purity) was deposited on single crystalline MgO(001) substrates (5×5 mm^2) held at 50 °C. Identical growth conditions were insured by deposition of films with different thickness in a single deposition run using a computer controlled shutter near the sample holder. The film thicknesses were 20 to 100 nm for Cr and 2 to 110 nm for Fe. Sample oxidation was prevented by a 10 nm SrF_2 protection layer. Resistance and Hall effect were measured by the van der Pauw method [20] in a temperature controlled cryostat equipped with a 15 T superconducting magnet. Reflective high energy electron diffraction and *ex situ* x-ray diffraction (XRD) spectra showed that the films grow epitaxially on the MgO(001) substrates [19, 21].

3. Experimental results and discussion

3.1. Thickness dependence of the resistivity

The model of Fuchs [1, 2] describing the thickness dependence of the electrical resistivity (ρ) enables us to discriminate bulk and surface contributions to the electron scattering. The resistivity of a thin metallic film is described by:

$$\rho(t) = \rho_\infty + \frac{3}{8}(1-p)\frac{\rho_\infty l_\infty}{t} \tag{1}$$

with film thickness t, bulk resistivity ρ_∞, bulk electron mean free path l_∞ and specularity parameter p, which is the fraction of electrons specularly reflected at the film surfaces. The different parameters of (1) are best evaluated from a plot of $\rho(t)t$ versus t as shown in figure 1 for three different measuring temperatures (300, 77, 4.2 K). The ρ_∞ values obtained are shown in figure 2. The l_∞ values calculated assuming $p = 0$ (which gives a lower limit for l_∞) are of the order of 14 nm at room temperature (RT) and increase to 630 nm (Cr) and >1100 nm (Fe) at 4.2 K. The bulk resistivities at RT (9.7 $\mu\Omega$ cm for Fe and 12.9 $\mu\Omega$ cm for Cr) are close to reported bulk properties whereas the low temperature values indicate the

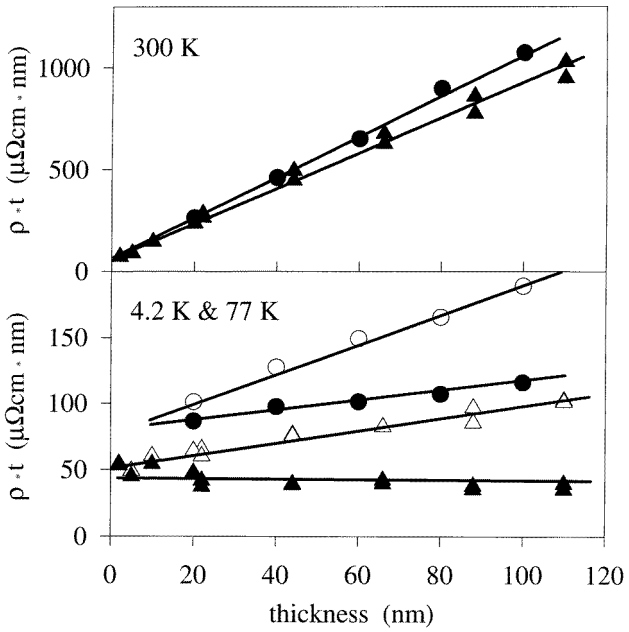

Figure 1. Thickness dependence of the resistivity times the thickness ρt for Fe (triangles) and Cr (circles) films, measured at respectively 300 K (upper graph), 77 K (lower graph, empty data points) and 4.2 K (lower graph, filled data points). The straight lines are best fits according to (1).

predominance of surface scattering even for rather thick films. The residual resistance ratios $\rho(\mathrm{RT})/\rho(4.2\ \mathrm{K})$ for the bulk are respectively 29 (Cr) and >90 (Fe). The major decrease of ρ_∞ is observed upon cooling from RT to 77 K but ρ_∞ still drops by about another factor of three when reducing the temperature to 4.2 K (figure 2).

Fe/Cr superlattices which have typical individual layer thicknesses around 1 nm show low temperature resistivities of about 20 $\mu\Omega$ cm [21] (when grown under identical conditions as the single layers). This value is of the order of the resistivities of single layers with 1 nm thickness. Thus the transport properties of such superlattices are characterized by strong interface scattering. At RT the ratio between bulk mean free path (about 14 nm) and individual layer thickness in Fe/Cr superlattices is still large. Therefore, one might argue that also at RT interface scattering dominates the transport properties and the bulk contribution is negligible. Then the RT amplitude of the GMR should be comparable to the low temperature value, which is clearly not the case [21]. Accordingly, one should be careful when comparing the electron mean free path with the layer thickness [17, 22]. The electron states might still be extended over several layers requiring a much longer mean free path to avoid bulk contributions. The long low temperature mean free paths in the bulk of the Fe and Cr layers certainly form the basis for the size of the GMR effect of epitaxial Fe/Cr superlattices grown on MgO(001).

3.2. Temperature dependence of the resistivity

The analysis of the temperature dependence of the resistivity reveals details about the transport properties and underlying scattering mechanisms. The resistivity of normal metals

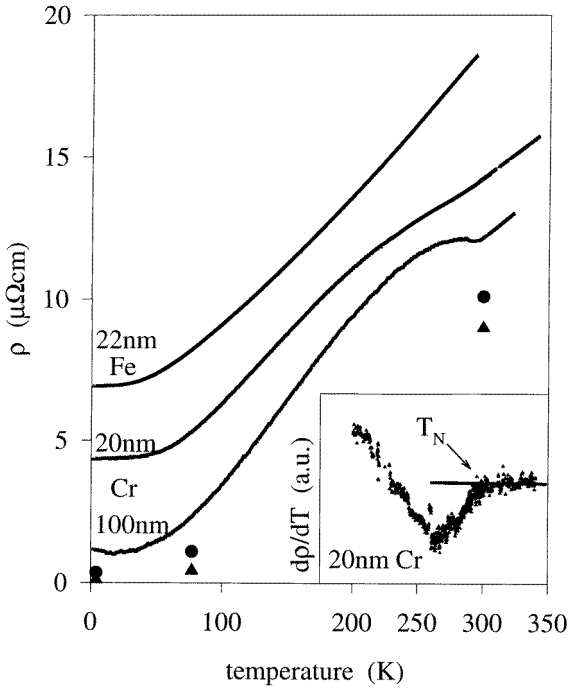

Figure 2. Temperature dependence of the resistivity of Cr films (20 and 100 nm), a Fe film (22 nm) and the bulk values of Fe (triangles) and Cr (circles) derived from figure 1. The resistivity data of the 22 nm Fe film are shifted upwards by 5 $\mu\Omega$ cm for clarity. The inset shows the derivative of the resistivity for the 20 nm thick Cr film. The point where $\partial\rho/\partial T$ starts to deviate from the constant high temperature value (horizontal line) indicates the Néel temperature T_N.

has two characteristic temperature regimes: low temperature saturation at the residual resistivity value which is determined by the concentration of static scattering centres such as impurities, grain boundaries and, in particular for thin films, surfaces. At high temperature ρ increases linearly with temperature. The slope in this linear regime is called the temperature coefficient of the resistivity which is an element specific quantity. However, a linear temperature dependence of the resistivity is not found for ferro- and antiferromagnetic materials [23, 24]. In this case shape and slope of $\rho(T)$ are influenced by the magnetic properties which allows us to extract information about magnetic ordering processes.

The resistivity of Fe films as a function of the temperature shows a continuous bending (figure 2) which is in agreement with reported results [23]. The temperature range around the Curie temperature T_C (which can be expected to be close to the bulk T_C value) could not be reached without causing irreversible changes of the film structure. The temperature dependence of the resistivity of Cr films is characterized by a nonlinear behaviour between 200 and 300 K (figure 2). This nonlinearity is caused by changes in the magnetic order of Cr at the Néel temperature T_N [24]. The value of T_N is defined by the temperature at which the resistivity starts to deviate from the linear behaviour above T_N [25]. This can be observed directly in $\rho(T)$ (100 nm Cr in figure 2) or estimated from the derivative $\partial\rho/\partial T$ as shown in the inset of figure 2. We find a value of $T_N = 300$ K for Cr films of 20 and 100 nm thicknesses which is close to the bulk T_N of 311 K [24]. The difference may be

due to epitaxial stress [24, 26] caused by the 3.5% lattice parameter misfit between Cr and the MgO substrate.

3.3. Hall effect

The Hall effect of non-magnetic metals is a linear function of the magnetic field and the size of the effect is related to the charge carrier density. The magnetization of ferromagnets causes strong deviations from linearity (figure 3) such that the Hall resistivity R_H is described by the ordinary Hall coefficient R_o and the spontaneous Hall coefficient R_s via

$$R_H = R_0 B + 4\pi R_s M \tag{2}$$

with the induction $B = H + 4\pi M(1 - N)$ and the magnetization M [18, 27]. The demagnetization factor N is equal to 1 due to the geometry of the magnetic field applied perpendicular to the film plane. R_o can be derived from the high field slope of the Hall resistivity whereas R_s is found by extrapolation of this high field slope towards $H = 0$; the saturation magnetization M_s can be extracted from the field at which the initial slope saturates [18] (figure 3). While the ordinary Hall effect arises from the Lorentz force, the spontaneous Hall effect is caused by spin–orbit interaction between conduction electrons and impurities [28–31].

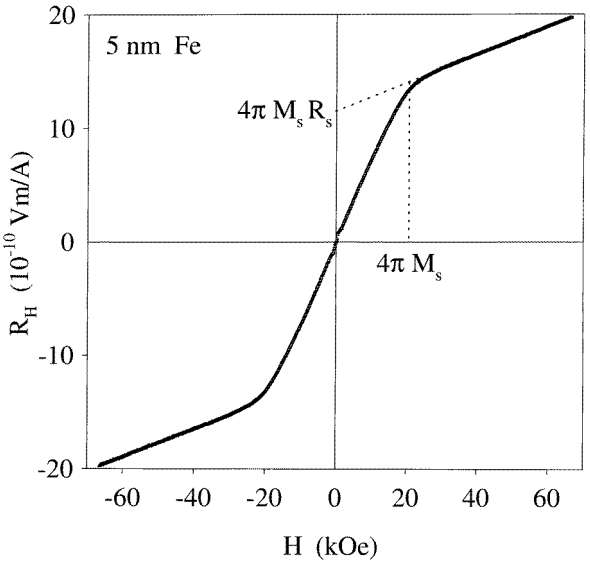

Figure 3. Magnetic field dependence of the Hall resistivity R_H of a 5 nm thick Fe film measured at 4.2 K. The dashed lines indicate respectively the point where the magnetization saturated and the slope of the ordinary Hall effect at high field. The extrapolation of the latter towards $H = 0$ gives the value of R_s.

The low temperature ordinary Hall effect R_o of the films shows a weak dependence on film thickness (figure 4). The value of R_o is around $R_o = 10^{-10}$ m^3 C^{-1} and seems to decrease for the thickest Fe films, i.e. the lowest resistivities. This size of R_o is characteristic for Fe films and also some resistivity dependence is known from literature [27]. The spontaneous Hall coefficient depends strongly on film resistivity. A linear or

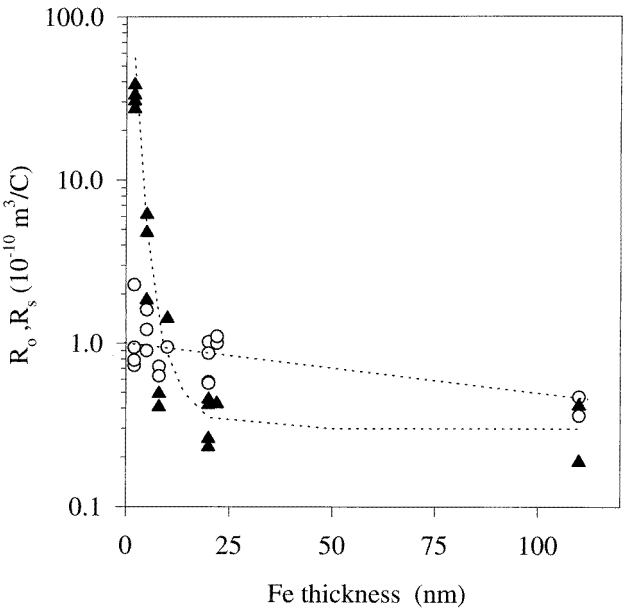

Figure 4. Normal (R_o, open circles) and spontaneous (R_s, filled triangles) Hall coefficient of Fe films measured at 4.2 K as a function of film thickness. The lines are guides to the eye.

square resistivity dependence of R_s is representative for respectively skew scattering or side jump scattering mechanisms of the electrons [27–31]. The resistivity may be altered by variation of sample temperature or thickness. Although the temperature can be varied easily this leads to an entangled combination of various scattering mechanisms (scattering at impurities, phonons and magnons) with different contributions to R_s [27]. Accordingly, the interpretation of the experimental results would be difficult. The layer thickness variation used here enables the adjustment of resistivity over two orders of magnitude with the surfaces acting as the exclusive source for electron scattering. R_s first decreases drastically with increasing Fe layer thickness and saturates for the thickest layers (figure 4). As a function of film resistivity R_s does not show a simple behaviour. The high resistivity part ($\rho > 1\ \mu\Omega$ cm) is well described by $R_s \sim \rho^n$ with $n = 1.8 \pm 0.2$ (full line in figure 5) indicative of side jump scattering. However, at lower resistivities the data deviate from this straight line and seem to saturate. A combination of a linear and a square ρ dependence also does not describe the measured data. Phenomenologically, the resistivity dependence can be fitted introducing a constant term in the form

$$R_s = 0.23 \times 10^{-10}\ \mathrm{m^3\ C^{-1}} + 5.7 \times 10^4\ (\Omega\ \mathrm{m\ T})^{-1}\rho^2$$

as shown by the dashed line in figure 5. However, such a constant term lacks theoretical explanation and would also require further experimental verification. The layer thicknesses and resistivities of Fe/Cr superlattices are typically in the regime where R_s obeys the simple power law behaviour. The comparison of the resistivity dependence of R_s with theory [32] should reveal insight into details of the scattering mechanisms. Provided that the resistivity is varied via the layer thickness rather than via the temperature the obtained information will be truly interface sensitive.

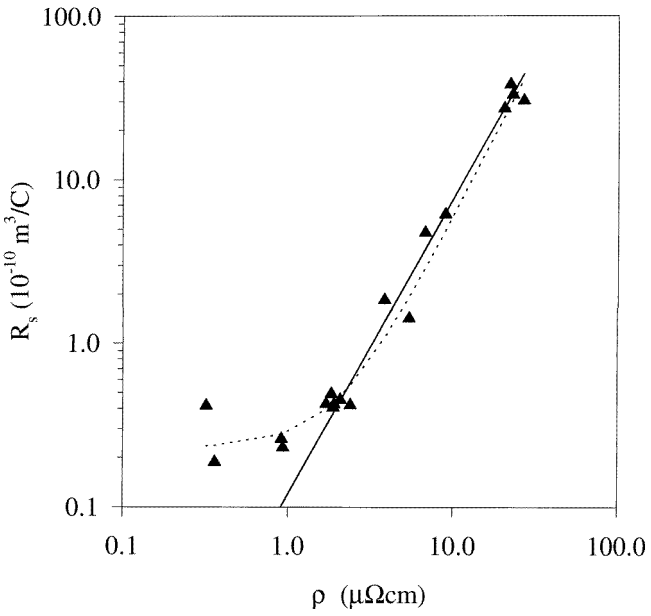

Figure 5. Spontaneous Hall coefficient R_s of Fe films measured at 4.2 K as a function of film resistivity ρ. The straight full line represents $R_s \sim \rho^{1.8}$. The dashed line is a fit consisting of the sum of a constant and a ρ^2 term.

4. Conclusions

Epitaxial Fe and Cr films grown on MgO(001) by MBE have extremely low intralayer defect densities represented by their low bulk residual resistivities. The temperature dependence of the resistivity of Cr films shows anomalies due to magnetic ordering effects close to T_N. We find a value of $T_N = 300$ K (close to bulk T_N of Cr) for Cr thicknesses of 20 nm and 100 nm. The Hall effect of Fe films shows typical ferromagnetic behaviour. The spontaneous Hall effect is dominated by side jump scattering (resistivity dependence close to $R_s \sim \rho^2$) with possibly a small resistivity independent contribution.

Such Fe and Cr layers are suitable for the preparation of Fe/Cr superlattices with negligible intralayer scattering. The resulting dominance of the interface scattering allows the study of the interplay between interface scattering and giant magnetoresistance without interfering bulk contributions [33]. The anomalies in the temperature dependence of the resistivity are related to the antiferromagnetic ordering in the Cr layers, the knowledge of which is essential for the understanding of the exchange coupling [34, 35]. The study of the resistivity dependence of the anomalous Hall effect sheds light upon the electron scattering processes. Such experiments are surface sensitive provided that other scattering mechanisms like phonon or magnon scattering are avoided, i.e. when the temperature is kept low and the resistivity is modified via the layer thickness. Applying this concept to epitaxial Fe/Cr superlattices will reveal details about the interface scattering.

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