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Electric transport properties of epitaxial Fe and Cr films with very low intralayer scattering

R. Schad*, P. Beliën[1], G. Verbanck, C.D. Potter[2], K. Temst, V.V. Moshchalkov, Y. Bruynseraede

Laboratorium voor Vaste-Stoffysika en Magnetisme, K.U. Leuven, Celestijnenlaan 200 D, B-3001 Leuven, Belgium

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Abstract

The low-temperature transport properties of epitaxial Fe and Cr films grown on MgO(1 0 0) substrates by molecular beam epitaxy are characterised by extremely low intralayer resistivities, indicating a very small concentration of defects. This makes such films particularly suitable for studies of the influence of the interface scattering on the transport properties of Fe/Cr superlattices. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

It is well known that the electrical transport properties of thin metallic films are strongly influenced by scattering of electrons at the surfaces. This was first described phenomenologically by Fuchs [1,2], who introduced a specularity parameter p which is the fraction of elastically reflected electrons scattered at the surface. This theory was further developed and refined, taking into account thickness variations over the film [3] and scattering at grain boundaries [4]. Other models include a dependence of p on the angle of incidence of the conduction electrons [5] or provide a quantum mechanical description in the presence of surface roughness on the atomic scale [6]. It turned out that the thickness dependence of the resistivity of polycrystalline films is dominated by grain-boundary scattering [4,7] rather than surface scattering. This is due to the fact that in these systems the average grain size often scales with the film thickness, causing a thickness dependence of the

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* Corresponding author. Present address: Research Institute for Materials, Katholieke Universiteit Nijmegen, Toernooiveld 1, NL 6525 ED Nijmegen, Netherlands Tel.: + 31 24 365 3094; fax: + 31 24 365 2190; e-mail: schad@sci.kun.nl.

[1] Present address: Philips Research Labs, Prof. Holstlaan 4, NL 5656 AA Eindhoven, Netherlands.

[2] Present address: Argonne National Lab, 9700 S. Cass Ave, Argonne, IL 60439, USA.

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resistivity similar to the case of pure surface scattering. In order to make sure that the transport properties of such polycrystalline films are governed by real surface scattering, it is necessary to investigate their topography providing information about the granularity and its variation with film thickness.

The surface topography of epitaxial films, however, is a consequence of the growth kinetics. The internal film structure has defects like dislocations rather than grain boundaries so that the theory of Ref. [4] does not apply. Accordingly, the electron mean-free path in epitaxial thin films can exceed the average grain diameter [8]. Additionally, epitaxial stress due to the lattice misfit between substrate and film often produces the formation of misfit dislocations as the film thickness increases [9]. This would contribute to the thickness dependence of the resistivity in a way opposite to surface scattering, thus not miming a Fuchs-like behaviour. Thus, the thickness dependence of the resistivity of epitaxial thin films is commonly well described by surface scattering and a constant bulk contribution even down to extremely thin films [10].

In the case that grain-boundary scattering can be neglected, three different models exist, describing the influence of surfaces on the transport properties. The theory of Fuchs [1,2] is most often used, although it provides only a phenomenological description of the surface scattering. This theory has the advantage of simplicity and it allows straightforward extraction of the transport parameters. The application of the model of Soffer [5] leads sometimes to unrealistic values of the parameters [5,11] which makes it less credible. The model of Tesanovic et al. [6] is based on a more realistic description of the transport process and was successfully applied in several cases [6,9,12]. However, its drawback is that the bulk electron mean-free path cannot be separated from the surface roughness, so one of those has to be determined independently.

Recently, a renewed interest in the influence of surface effects on the properties of metallic films has arisen due to the importance of interfaces in superlattices. An excellent example is the giant magnetoresistance (GMR) observed in Fe/Cr superlattices [13] which is governed by spin-dependent scattering of the electrons at the interfaces [14]. However, it is not fully clear yet how the GMR effect depends on the interface quality. Although it was shown theoretically that a certain amount of interface roughness is necessary to obtain high GMR amplitudes, both an increase [15–17] and a decrease [18] of the GMR amplitude was observed with increasing interface quality. In these studies, the interface quality is altered by external preparation parameters like sputtering gas pressure, substrate temperature during deposition, deposition rate, substrate structure and roughness, etc. Unfortunately, this will influence the intralayer properties as well. When the intralayer scattering dominates the total resistivity of the superlattice it will be very difficult to relate the observed changes in the GMR amplitude to changes of the interface properties.

Therefore optimised deposition techniques and substrates have to be used to grow Fe and Cr films with low resistivities and only surface scattering. For example, epitaxial Fe films grown on GaAs and textured films on Corning glass are unsuitable since they show rather high resistivities [11,19]. It is necessary to use an insulating substrate which does not intermix with Fe and Cr and supports epitaxial growth of these materials.

In this paper we analyse the electrical transport properties of epitaxial Fe and Cr films with extremely low intralayer scattering, showing low-temperature bulk resistivities of $\rho_\infty < 0.1$ μΩ cm for Fe and $\rho_\infty = 0.35$ μΩ cm for Cr.

2. Experimental

The Fe and Cr films were prepared in a Riber MBE deposition system (2×10^{-11} mbar base pressure) using two electron-beam guns. The evaporation rate of 0.1 nm/s was stabilised within 1% by a homemade feedback control system using Balzers quadrupole mass spectrometers (QMS). Integration of the QMS signal allowed automatic control of the shutters for the individual evaporation sources. The 99.996% pure starting material was deposited on single-crystalline MgO(1 0 0) substrates held at 50°C. In order to ensure identical growth conditions, films with different thicknesses were deposited during the same evaporation cycle using a computer-controlled, stepper-motor-driven shutter near the sample holder. We will report on the properties of single films with thicknesses ranging from 20 to 100 nm for Cr and 2 to 110 nm for Fe. Oxidation of the samples was prevented by a 10 nm SrF_2 cap layer deposited using a Knudsen cell.

Ex situ X-ray diffraction (XRD) spectra were obtained in the θ–2θ mode using a Rigaku rotating anode system with a Cu target operating at about 4 kW and a wavelength of the Cu K_α radiation of 1.542 Å. Finite-size peaks observed at low angle were used to calibrate the film thickness. High-angle spectra were used to determine the crystallographic orientation.

The resistance was measured on $5 \times 5\ \mathrm{mm}^2$ samples by the Van der Pauw method [20] in a temperature-controlled cryostat equipped with a 15 T superconducting magnet.

3. Results and discussion

3.1. Structure

Fig. 1 shows a high-angle XRD spectrum of a single Cr film grown on MgO(1 0 0). Besides the two substrate peaks, only a sharp Cr(2 0 0) peak is observed, indicating excellent epitaxial growth. This was also verified using reflective high-energy electron diffraction and off-axis XRD. The XRD spectra of the epitaxial Fe films on MgO(1 0 0) look similar.

The polar and azimuthal rocking-curve widths are around 1° for both materials. The polar rocking-curve width is determined by two contributions: first, a polar mosaic spread of the film orientation and, second, a limited lateral coherence length. The narrow rocking curves indicate the absence of large-angle grain boundaries.

The surface roughness was examined ex situ using atomic force microscopy (AFM). Fig. 2 shows the surface topography of, respectively, 5 nm (a) and 100 nm (b) thick Cr films. The rms roughness values for both surfaces is about 0.5 nm. Accordingly, the resistivity data can be described without taking into account corrections due to large thickness fluctuations [3]. The typical lateral diameter of protrusions is similar for both thicknesses (about 10 nm). This rules out a granular structure with grain diameter proportional to film thickness as assumed in Ref. [4]. The surface topography of Fe layers is similar.

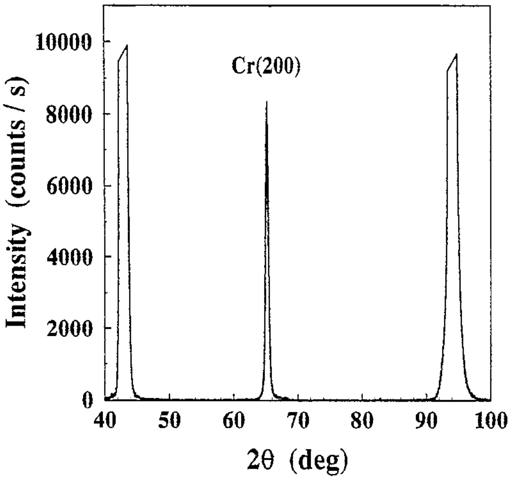

Fig. 1. Typical high-angle XRD spectrum of an epitaxial Cr film with (1 0 0) orientation grown on MgO(1 0 0). Besides the substrate peaks at $2\theta = 43°$ and 94° (which are cut off) only the Cr(2 0 0) peak at $2\theta = 65°$ is observed. The XRD spectra of Fe films look similar.

3.2. Thickness dependence of the resistivity

For the description of the thickness dependence of the resistivity, the theories from Refs. [3,4] are unsuitable since the films are epitaxial and have a small surface roughness. Accordingly, we describe the experimental data using the theories of Refs. [1,2,6]. The Fuchs–Sondheimer model [1,2] describes the thickness dependence of the electrical resistivity $\rho(t)$ as

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

with t the film thickness, ρ_∞ the bulk resistivity, l_∞ the bulk electron mean-free path, and p the specularity parameter. On the other hand, $\rho(t)$ can also be calculated using the model of Tesanovic et al. [6]:

$$\rho(t) = n_c \rho_\infty \left[\sum_{n=1}^{n_c} (1 + n^2 l_\infty / l_{\max})^{-1} \right]^{-1}, \tag{2}$$

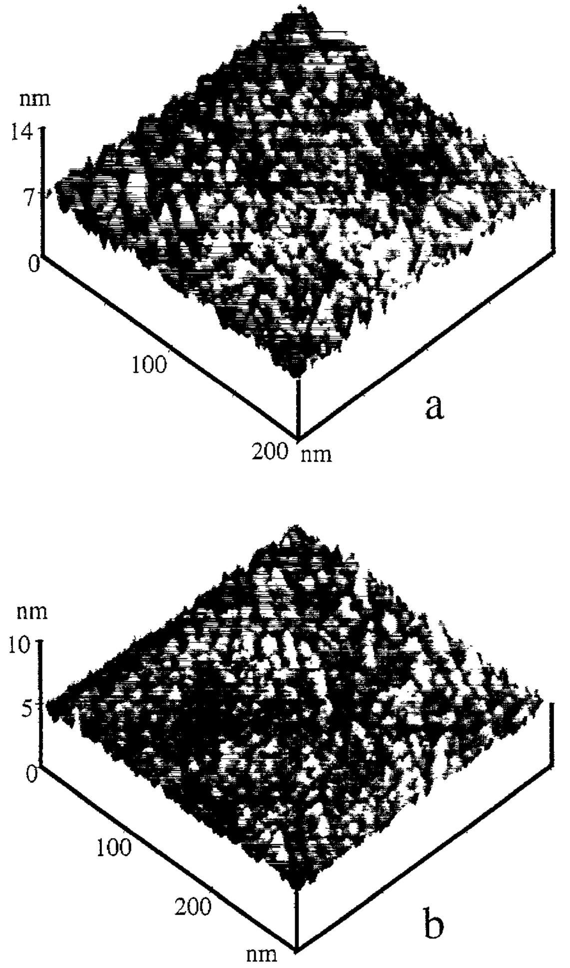

Fig. 2. Surface topography measured by AFM of epitaxial Cr films with a thickness of 5 nm (a) and 100 nm (b).

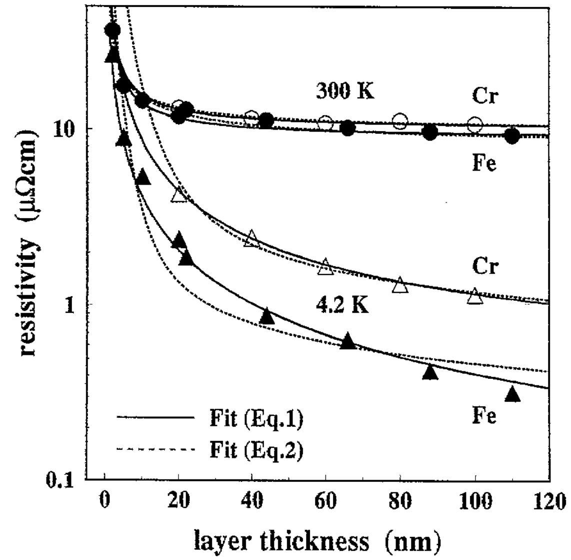

Fig. 3. Thickness dependence of the resistivity of Fe (filled symbols) and Cr (empty symbols) films, measured at, respectively, 300 K (circles) and 4.2 K (triangles). The full and dashed lines are best fits according to, respectively, Eqs. (1) and (2).

Table 1

Bulk transport parameters of epitaxial Fe and Cr films obtained by fitting the thickness dependence of the resistivity (Fig. 3) using Eqs. (1) and (2)

	T (K)	ρ_∞ (μΩ cm)		l_∞ (nm)	
		Eq. (1)	Eq. (2)	Eq. (1)	Eq. (2)
Fe	300	9.0	8.3	15	50
	4.2	< 0.1	0.025	> 1100	10^5
Cr	300	10.1	10.0	13	30
	4.2	0.35	0.03	630	5×10^5

with $n_c = k_F t/p$ the number of subbands in the k space located at the Fermi level, $l_{max} = 6\pi n_c^2 t/(k_F h)^2$, k_F the Fermi momentum and h the microscopic roughness amplitude.

Fig. 3 shows the resistivity of the epitaxial Fe and Cr films measured at 300 and 4.2 K as a function of the film thickness. Also displayed are best fits using Eq. (1) or Eq. (2). The values of l_∞ and ρ_∞ obtained from these fits are summarised in Table 1. Since the separation of the parameters p and l_∞ in Eq. (1) is problematic, we assumed $p = 0$ and obtain, therefore, a lower limit of the mean-free path. In Eq. (2) we assumed that $h =$ 0.3 nm, corresponding to twice the atomic step height, which is a realistic value [9].

The room-temperature resistivities are well described by both theories producing ρ_∞ values close to the bulk resistivity. At low temperature, however, the Fuchs–Sondheimer model provides the better fit (Fig. 3) and lower (higher) values for l_∞ (ρ_∞) (Table 1) than the theory of Tesanovic et al. The value of $\rho_\infty = 0.1$ μΩ cm for the Fe films at 4.2 K is an upper limit since the resistivity does not tend to saturate, therefore ρ_∞ may be even smaller and, accordingly, l_∞ longer than 1.1 μm. The values obtained by the fit with Eq. (2) are rather high but still plausible and of the order of values typical for high-purity bulk samples [21].

These values have to be compared with those reported in the literature for thin-film samples. A similar study was performed on polycrystalline samples but only at ambient temperature [11]. But assuming the validity of Matthiesen's rule, the low-temperature bulk resistivities have to be of the order of several μΩ cm, which is about one order of magnitude higher than ρ_∞ of our films. Epitaxial Fe films ($t_{Fe} = 20$ nm) grown on GaAs showed $\rho = 7$ μΩ cm at 4.2 K but with stronger thickness dependence [19] than described by Eq. (1) or Eq. (2). Accordingly, the authors attributed this high resistivity to either impurity scattering due to interdiffusion of substrate material into the film or to defects introduced by the lattice mismatch between film and substrate [19]. Since the lattice mismatch between Fe and GaAs (1.4% misfit) is smaller than for Fe on MgO (3.7% misfit), these high resistivities are likely to be due to As interdiffusion from the substrate.

Surprisingly, much higher resistivities of about 200 μΩ cm for $t_{Fe} = 2$ nm were also reported for epitaxial Fe films grown on MgO(1 0 0) [22]. This exceeds the corresponding value of our Fe films by one order of magnitude. Furthermore, compared to other epitaxial, non-intermixing systems such as Ag on Si(1 1 1) ($l_\infty = 100$ nm [8]), our Fe and Cr films show very low resistivities indicating a small defect density.

Finally, we may compare the single-film transport properties with the ones of Fe/Cr superlattices grown under similar conditions on MgO(1 0 0) [23]. The low-temperature resistivity of about 15 μΩ cm corresponds approximately to the resistivity of a 2 nm thick single Fe layer. Thus, the transport properties of such superlattices are characterised by dominant interface scattering and negligible intralayer scattering.

4. Conclusions

Epitaxial Fe and Cr films grown on MgO(1 0 0) by MBE show extremely low defect densities, resulting in very small values of the bulk resistivity. The large intralayer electron mean-free path of 1.1 μm in the epitaxial Fe layers may also require ballistic transport experiments in metal films to be performed.

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References

[1] K. Fuchs, Proc. Cambridge Philos. Soc. 34 (1938) 100.
[2] E.H. Sondheimer, Adv. Phys. 1 (1952) 1.
[3] Y. Namba, Jpn. J. Appl. Phys. 9 (1970) 1326.
[4] A.F. Mayadas, M. Shatzkes, Phys. Rev. B 1 (1970) 1382.
[5] S.B. Soffer, J. Appl. Phys. 38 (1967) 1710.
[6] Z. Tesanovic, M.V. Jaric, S. Maekawa, Phys. Rev. Lett. 57 (1986) 2760.
[7] J.R. Sambles, Thin Solid Films 106 (1983) 321.
[8] R. Schad, Leitfähigkeit ultradünner epitaktischer Silberfilme auf Silizium(111), VDI-Verlag, Düsseldorf, 1991.
[9] J.M. Phillips, J.L. Batstone, J.C. Hensel, M. Cerullo, Appl. Phys. Lett. 51 (1987) 1895.
[10] M. Jalochowski, E. Bauer, H. Knoppe, G. Lilienkamp, Phys. Rev. B 45 (1992) 13607.
[11] M. Jacob, G. Reiss, H. Brückl, H. Hoffmann, Phys. Rev. B 46 (1992) 11208.
[12] U. Jacob, J. Vamcea, H. Hoffmann, Phys. Rev. B 41 (1990) 11852.
[13] M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, J. Chazelas, Phys. Rev. Lett. 61 (1988) 2472.
[14] R.Q. Hood, L.M. Falicov, D.R. Penn, Phys. Rev. B 49 (1994) 368.
[15] P. Beliën, R. Schad, C.D. Potter, G. Verbanck, V.V. Moshchalkov, Y. Bruynseraede, Phys. Rev. B 50 (1994) 9957.

[16] N.M. Rensing, A.P. Payne, B.M. Clemens, J. Magn. Magn. Mater. 121 (1993) 436.
[17] S. Joo, Y. Obi, K. Takanashi, H. Fujimori, J. Magn. Magn. Mater. 104–107 (1992) 1753.
[18] E.E. Fullerton, D.M. Kelly, J. Guimpel, I.K. Schuller, Y. Bruynseraede, Phys. Rev. Lett. 68 (1992) 859.
[19] M. Rubinstein, F.J. Rachford, W.W. Fuller, G.A. Prinz, Phys. Rev. B 37 (1988) 8689.
[20] L. van der Pauw, Philips Res. Rep. 13 (1958) 1.
[21] G.J.C.L. Bruls, J. Bass, A.P. van Gelder, H. van Kempen, P. Wyder, Phys. Rev. B 32 (1985) 1927.
[22] C. Liu, Y. Park, S.D. Bader, J. Magn. Magn. Mater. 111 (1992) L225.
[23] R. Schad, C.D. Potter, P. Beliën, G. Verbanck, V.V. Moshchalkov, Y. Bruynseraede, Appl. Phys. Lett. 64 (1994) 3500.