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Spin-valve behaviour of anti-ferromagnetic boundaries in ultrathin magnetite films

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Abstract

Magneto-resistance (MR) measurements on epitaxial Fe_3O_4 films grown on polished MgO have been performed. The measurements presented here are interpreted by a model that describes the MR behaviour as spin-polarised transport across anti-ferromagnetic (AF) interfaces. The Fe_3O_4 films consist of structural domains, separated by anti-phase boundaries where an AF coupling is present. These AF interfaces enhance the resistance of the films. Upon application of a magnetic field the AF-spins rotate towards each other and the resistance decreases. The AF interfaces are thus behaving as spin-valves. In agreement with the model, the observed magneto-resistance is negative and shows linear and quadratic field dependence up to the anisotropy field for fields applied parallel and perpendicular to the film plane respectively. Above the anisotropy field, the slopes of the two MR curves are expected to be equal, which is observed at 60 K. Above the Verwey transition, the shape of the normalised MR curves is independent of temperature. Below the Verwey transition the MR curve becomes more linear with decreasing temperature. A large difference between parallel and perpendicular MR is observed at the Verwey transition.

Keywords: Spin-valve; Magneto-resistance; Spin-polarised transport; Magnetite

1. Introduction

Magnetite, Fe_3O_4, is an interesting material since it is both conducting and magnetic with a high Curie temperature of 858 K. Moreover, Fe_3O_4 is a half metallic ferrimagnet with a gap at the Fermi level for the majority electrons, but not for the minority of electrons [1]. Epitaxial Fe_3O_4 films grown on MgO substrates show magneto-resistance (MR) even at room temperature, whereas single crystals do not [2]. The cause of this difference in MR behaviour is due to the fact that epitaxial Fe_3O_4 films consist of structural domains separated by anti-phase boundaries (APBs) [3–5]. The magnetic coupling over a large fraction of these boundaries is anti-ferromagnetic (AF). These AF interfaces block the spin-polarised electron transport between neighbouring domains. Upon application of a magnetic field, the AF spins are tilted and the electron transport across the boundaries increases. In this way, the AF-APBs behave as spin-valves.

In contrast with conventional spin-valves (magnetic multilayers [6]), the spin-valves reported here are in the film plane and separate two ferromagnetic regions by an atomically sharp anti-ferromagnetic boundary. The anti-phase boundaries result as growth defects as a consequence of the fact that the lattice constant of Fe_3O_4 ($a = 8.397$ Å) is twice as large as the lattice constant of MgO ($a = 4.212$ Å). Furthermore, the Fe_3O_4 film has a lower symmetry (Fd3□m) than the underlying substrate (Fm3□m). When different islands meet, they can be shifted or rotated with respect to each other [3,4]. The resulting defect is an anti-phase boundary with a shift vector of the type $1/4\ a\langle 110\rangle$. The structural domains coalesce with a well-defined boundary between them and are strongly coupled magnetically.

We have investigated the spin-valve behaviour of 6, 12 and 30 nm-thick epitaxial Fe_3O_4 films grown on polished MgO by studying the magneto-resistance

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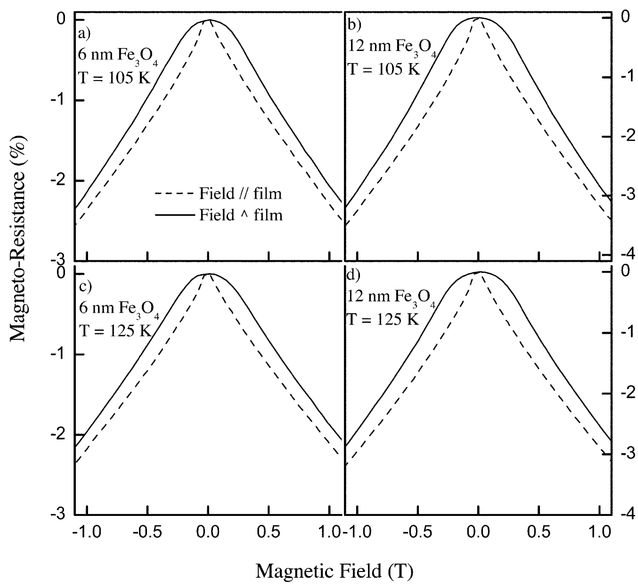

Fig. 1. Low field magneto-resistance of Fe_3O_4 films, with the field applied perpendicular (solid line) and parallel (dashed line) to the film plane for 6 and 12 nm films measured at 105 K (a and b), 125 K (c and d), respectively.

behaviour in magnetic fields up to 5 T. The films have been grown on polished MgO in contrast to previous experiments [7], because their resistance is much lower than for films grown on cleaved MgO.

2. Experimental

The epitaxial Fe_3O_4 films were grown on polished magnesium oxide (MgO) substrates using Molecular Beam Epitaxy (MBE) in an ultra high vacuum system with a background pressure of 10^{-10} mbar. The films were grown in an oxygen pressure of 10^{-6} mbar with an iron flux of 1.25 Å/min at a substrate temperature of 525 K. For the MR measurements, four contacts were made by depositing 20 Å of Ti and 40 Å of Au. Resistance measurements were performed in a Quantum Design PPMS system using magnetic fields up to 5 T, with the magnetic field applied both parallel and perpendicular to the film plane. The MR measurements were done in the constant voltage mode and the current was measured along the [100] direction of the films.

3. Results and discussion

Fig. 1 shows the low field (up to 1 T) MR measurements for 6 and 12 nm Fe_3O_4 at 105 and 125 K, respectively. For parallel applied fields, the MR is larger than for perpendicular applied fields and the MR depends linearly on the applied magnetic field. For perpendicular applied fields, the MR behaviour is distinctly different. Up to 0.5 T the field dependence is quadratic.

The different field dependencies have been explained by a model for spin-polarised transport across an atomically sharp anti-ferromagnetic interface between two ferromagnetic chains [7]. The model calculates the change in orientation of the AF coupled spins upon application of a magnetic field. The MR can be calculated because the conductivity is proportional to $t_0^2\cos^2\varphi_{AF}$, where t_0 is the hopping integral [10] and φ_{AF} is the angle between the AF-spins at the boundary. The MR has a linear field dependence for magnetic fields applied parallel to the film plane:

$$MR = -C_1H \tag{1}$$

where C_1 is independent of the magnetic field.

For magnetic fields applied perpendicular to the film plane, out-of-plane anisotropy energy has to be taken into account and two regimes can be distinguished. Up to the anisotropy field H_{an}, the field dependence is quadratic and above the anisotropy field it is linear:

$$H < H_{an} \quad MR = \frac{C_1}{2H_{an}}H^2 \tag{2a}$$

$$H > H_{an} \quad MR = -C_1H + D. \tag{2b}$$

For perpendicularly applied fields larger than the anisotropy field, the slope of the MR curve is equal to the slope of the MR curve in case of parallel applied fields.

The low field MR behaviour for 6 and 12 nm thick films at both 105 and 125 K, is in agreement with this model, since the field dependence is linear for parallel fields and quadratic for perpendicular fields up to the anisotropy field, which is 0.53 T for a 115 nm thick epitaxial Fe_3O_4 film grown on MgO [8].

The high field MR-curves (i.e. above the anisotropy field) is expected to have the same slope for parallel and perpendicular applied fields (equations 1 and 2b). Fig. 2 shows the high field MR at 105 and 125 K for 6 and 12 nm films, respectively. The value of the MR increases with decreasing temperature and increasing film thickness. Similar behaviour has been found for Fe_3O_4 films grown on cleaved MgO substrates [7]. The MR curves start to coincide for parallel and perpendicular applied fields at 2 T. However, coalescence of the two curves is not expected. Even though ultrathin films (<5 nm) are known to be superparamagnetic [3] and the out-of-plane anisotropy becomes comparable to the (in-plane) magneto-crystalline anisotropy [9], these effects are of minor importance for the thicknesses of the measured films.

At lower temperatures the MR curves appear distinctly different. This is shown in Fig. 3, where the MR is plotted at 60 K for 12 and 30 nm-thick Fe_3O_4 films.

At 60 K, the perpendicular magneto-resistance remains smaller than the parallel one with a similar slope from 0.5 T in agreement with our model. There is

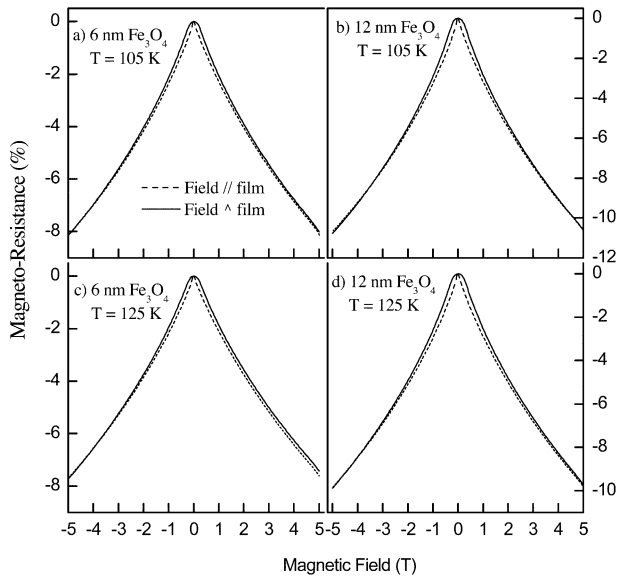

Fig. 2. High field magneto-resistance of Fe_3O_4 films, with the field applied perpendicular (solid line) and parallel (dashed line) to the film plane for 6 and 12 nm films measured at 105 K (a and b), and measured at 125 K (c and d), respectively.

small hysteresis in the MR curves for fields applied perpendicular to the film plane, but no hysteresis is observed for fields applied parallel.

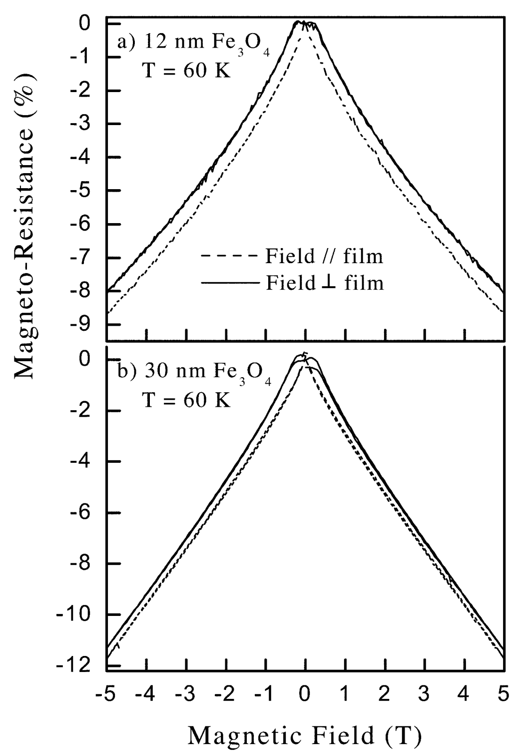

Fig. 3. Magneto-resistance at 60 K for (a) 12 and (b) 30 nm Fe_3O_4 film and the magnetic field applied perpendicular (solid line) and parallel (dashed line) to the film, respectively.

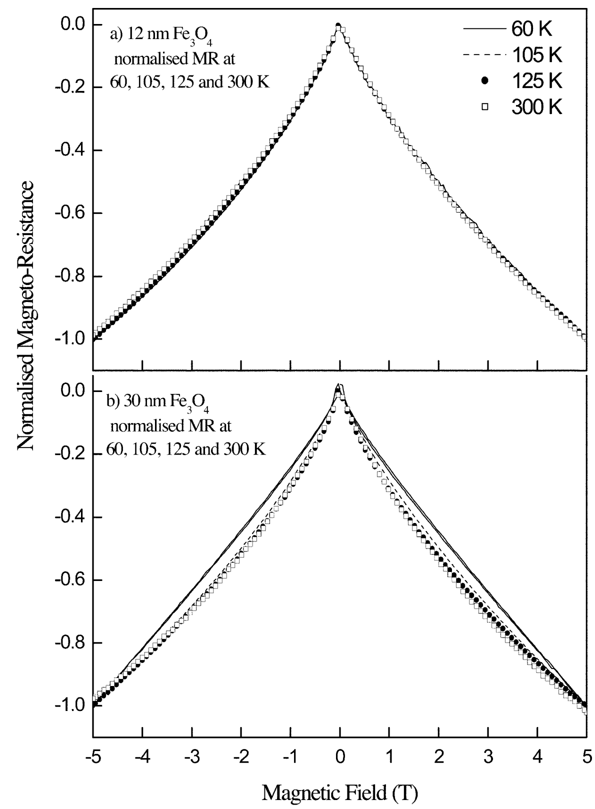

Fig. 4. Normalised magneto-resistances for (a) 12 and (b) 30 nm Fe_3O_4 films, both at 60, 105, 125 and 300 K, and the magnetic field applied parallel to the film.

Even though the absolute magnitude of the MR curves changes with temperature, the shape of the MR curves is independent of temperature for the 12 nm thick film as shown in Fig. 4a where the magneto-resistance has been normalised by taking the value at 5 T to be -1. This is not the case for the 30-nm thick film as can be seen in Fig. 4b. Below the Verwey transition, the MR behaviour becomes more linear with decreasing temperature. Even though the decrease of the high field MR starts to level off (for both thicknesses), the curves do not saturate even at 5 T. Similar behaviour has been observed in the magnetisation which does not saturate at 5 T [4]. This has also been attributed to the strong AF coupling of the spins at the boundary.

From resistance vs. temperature measurements (not shown here), the Verwey transition (T_V, which occurs around 120 K for bulk Fe_3O_4) of the films has been determined. The 12 nm-thick film does not show a Verwey transition in the temperature range 50–300 K, and the 30-nm thick film has a Verwey transition region between 95 and 107 K. This is consistent with measurements performed on Fe_3O_4 films grown by PLD [5]. The fact that the 30-nm-thick film is below T_V at 60 K might explain the difference in the shape of the MR

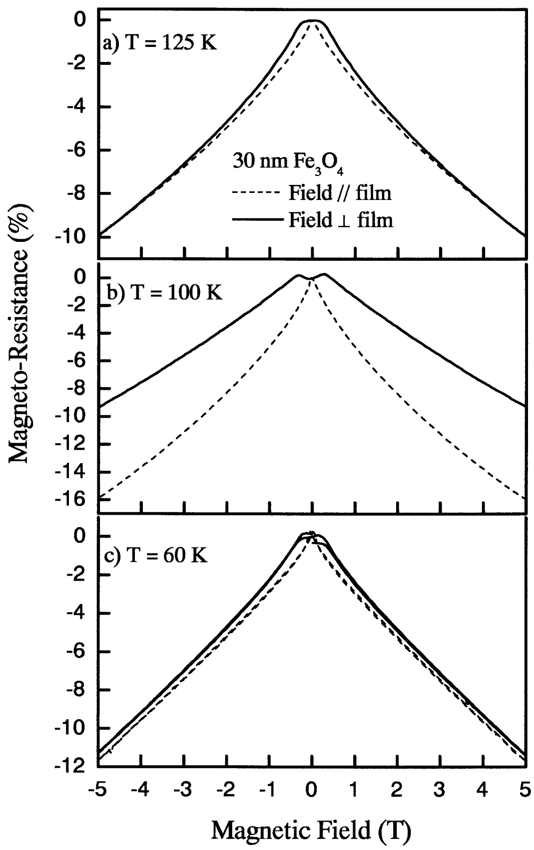

Fig. 5. Magneto-resistance for the 30 nm thick Fe_3O_4 film at (a) 125, (b) 100 and (c) 60 K, with the magnetic field applied perpendicular (solid line) and parallel (dashed line) to the film.

curves, even though the model holds both below and above T_V. The total conductivity is in both cases proportional to $t_0^2\cos^2\varphi$. In Fe_3O_4, both band and hopping conductivity have to be considered [10]. At lower temperatures where short range ordering is significant, hopping conductivity is small compared to the band conductivity. At higher temperatures, hopping conductivity becomes more important due to a gradual decrease of short range ordering [10].

Both single crystals [11] and epitaxial Fe_3O_4 films [12] exhibit a peak in the MR at the Verwey transition. The 30-nm-thick film also shows a peak in the MR behaviour at 100 K (MR $= -16\%$), but only for fields applied parallel to the film plane. There is a large difference between parallel and perpendicular applied fields at this temperature. This is shown in Fig. 5, where the MR is plotted for the 30-nm-thick film at 125, 100 and 60 K.

The large difference in parallel and perpendicular MR at 100 K is remarkable. In our model, only uniaxial out-of-plane anisotropy has been taken into account, because the value of the out-of-plane anisotropy constant K_u (0.13 MJ/m^3) is much larger than the in plane magneto crystalline anisotropy constant K_1 (9.4 kJ/m^3). However, K_1 changes sign below T_V and at T_V, it crosses zero; whereas the out-of-plane anisotropy is only partly reduced [8].

4. Conclusions

Sharp anti-ferromagnetic boundaries behaving as spin-valves have been grown successfully in ultrathin (6, 12 and 30 nm) epitaxial magnetite films. The observed magneto-resistance of the films is negative and can be explained by a model for spin-polarised transport across anti-ferromagnetic boundaries between two ferromagnetic chains. Below the anisotropy field, the MR behaviour is distinctly different for parallel and perpendicular applied fields, with quadratic field dependence for the latter case and linear dependence for parallel fields, in agreement with the proposed model. For applied fields larger than the anisotropy field (0.5 T), the MR curves are expected to have a similar slope for parallel and perpendicular applied fields. At 60 K, similar slopes for both curves are observed above 0.5 T. At 105 and 125 K, the two curves coincide for applied fields larger than 2 T. Above the Verwey transition the shape of the MR curves is independent of temperature. Below the Verwey transition, the shape becomes more linear with decreasing temperature. At the Verwey transition, a large difference in MR has been observed for fields applied parallel (-16%) and perpendicular (-9.5%) to the film plane.

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