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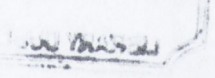
Mesoscopic Transport in Ferromagnetic Oxides

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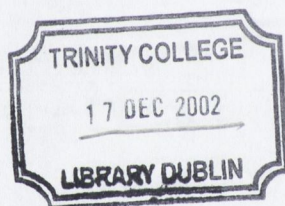
Jan Johannes Versluijs

A thesis submitted for the degree of
Doctor of Philosophy
in the University of Dublin

Department of Physics
Trinity College Dublin



October 2002



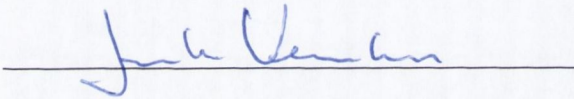
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DECLARATION

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Summary

The study presented in this thesis is concerned with mesoscopic transport in half-metallic ferromagnetic oxides. Two techniques have been developed to gain insight into the extrinsic transport properties of these oxides.

The first, scanning tunneling potentiometry (STP), was used to correlate topographic features of a thin film sample with the flow of current through the sample. A Scanning Tunneling Microscope (STM) was modified successfully to obtain simultaneously topographic and potentiometric images of a polycrystalline $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ thin film. Large voltage drops were observed to coincide with grain boundaries, whereas in the grains themselves essentially no change in voltage was found. From the voltage drop associated with the grain boundaries, an estimate of the resistivity of these grain boundaries can be obtained. It is found that it ranges from $3 \times 10^{-7} \Omega \text{ cm}^2$ to $3 \times 10^{-5} \Omega \text{ cm}^2$, with an average value of $6 \times 10^{-6} \Omega \text{ cm}^2$, which is comparable with other reports in the literature.

The second method described in this thesis is the study of magneto-transport properties in point contacts. Two tiny single crystals of either magnetite (Fe_3O_4) or $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ are mounted in a device where their separation can be controlled using a piezo-electric actuator. The two crystallites are brought into contact, and the resistance of the contact between them is monitored while they are being pulled apart again. Simple vibration insulation ensures that the contacts are stable for up to a minute or more, which is long enough to obtain current-voltage characteristics and measure the magnetoresistance and its hysteresis.

$(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ shows clear conductance quantization when the contacts are broken rapidly, but when broken slowly the contact can be stabilized at any value of conductance. Fe_3O_4 only shows a weak peak near $1G_0$ ($G_0 = \frac{2e^2}{h} \simeq (12.9 \text{ k}\Omega)^{-1}$), which is suppressed in

a field. Strong non-linear current-voltage characteristics are observed, which can be fitted to an $I = GV + cV^3$ law. Room temperature magnetoresistance (MR) values of up to 85 % for Fe_3O_4 and up to 45 % for $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ have been found for contacts with a low zero-bias conductance ($G \ll G_0$), in fields of 7 mT. For increasing conductance the MR falls off quickly.

The huge room temperature MR values in small fields are larger than those reported in any other ferromagnetic materials, such as ballistic Ni nanocontacts or tunnel junctions. Here MR is defined as $\frac{G(H)-G(0)}{G(H)}$. We rule out magnetostriction as the main explanation for these large effects. The magnetic hysteresis of around 1 mT shows that it is related to the magnetization process of the crystallites. Two possible explanations are considered; tunneling and spin pressure on a narrow domain wall pinned at the contact. Seeing the contact as a magnetic tunnel junction can explain the dependence of the MR on contact conductance, assuming that the smallest contacts, with a low conductance value, contain fewer impurities in the insulating barrier. The bias dependence of the MR could be then be due to magnon creation in the electrodes.

The other explanation, is based on the idea that a domain may be pinned at the contact. In contrast to the bulk, the width of the domain wall will then be small, of the order of the size of the constriction. For the current flowing through the contact, this narrow domain wall acts as an additional resistance. Application of the field removes the domain wall, leading to the large observed MR. Interaction between electrons traversing the contact and the wall lead to force, or pressure, exerted by the electrons on the wall. This spin pressure on the wall causes it to be pushed away from the constriction, out into the electrode, and thereby widening it, leading to bias-dependent resistance of the contact. Possible application of this

effect in a 'peanut' device with two unequal constrictions is discussed.

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Chapter 1

Introduction

In recent years, there have been a huge number of studies concerned with using magnetic materials for possible applications, such as non-volatile memory devices or sensors. This had led to a search for new materials, with higher Curie temperatures and better intrinsic magnetic properties. Half-metals and magnetic semiconductors especially are at the center of the attention. But also well-known materials, such as magnetite and CrO_2 , have recaptured interest for use in novel devices. The requirements for switching devices of practical use are a high Curie temperature ($> 200\text{ }^\circ\text{C}$), low power consumption, and large enough output for small fields.

Most ferromagnetic manganese oxides exhibit a large change in electrical resistivity when subjected to a magnetic field. This effect is most pronounced in the vicinity of the Curie temperature, where the Colossal Magnetoresistance is associated with a metal insulator transition.. The potential of Colossal MagnetoResistance (CMR) for practical applications, however, is limited because of the large field (of the order of 1 T) needed for the modification of the intrinsic magnetic order required to observe the effect.

Recently, studies on ferromagnetic tunnel junctions and on polycrystalline ferromagnets made progress by showing a low field magnetoresistance. This low field MR has been found in bulk, polycrystalline thin films, and in films with artificial grain boundaries, and was in all cases attributed to the presence of grain boundaries or interfaces between crystallites, although the mechanism responsible is not fully understood. One aim of this work is to contribute to the understanding of transport through grain boundaries.

The other line of investigation in this work is concerned with the understanding of electronic transport in nanometric structures. This is important, both from a basic point of view and from a technological perspective, considering the miniaturisation of device dimensions in communication and information technology. The study of metallic nanowires and nanocontacts has received a great deal of attention in the last few years. Here we extend this to nanocontacts in half-metallic oxides, and in particular to magnetite, Fe_3O_4 , and $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ (LSMO). Magnetite is the oldest known magnetic material; it is a ferrimagnet, with a high Curie temperature (860 K). LSMO has the highest known Curie temperature of the manganites (380 K). Both have a spin polarization of $\approx 100\%$, which makes it the most suitable candidate for potential applications.

This work is therefore mainly concerned with the investigation of microscopic or mesoscopic scale transport properties of half-metallic oxides, and of LSMO and magnetite in particular. Several experimental methods using point probes have been used to study transport through grain boundaries, crystallite interfaces and nanocontacts. Section 1.1 outlines the emerging field of spin electronics, and introduces a few important concepts. Section 1.2 gives a basic explanation of the different transport mechanisms in the nanometric half-metallic oxides structures studied here. Section 1.3 introduces magnetite and LSMO to the reader. This chapter concludes with some remarks on the Scanning Tunneling Potentiometry (STP) used here to study transport through grain boundaries in LSMO. STP can be used to simultaneously measure surface topography and spatial variations of the electrical potential.

1.1 Spin Electronics

Ever since the invention in 1947 of the transistor by Bardeen, Brattain and Shockley

at Bell, conventional electronics has proceeded to change modern daily life, with its use in everyday equipment like computers, radio, television and the telephone. Electronics, with its transistors and other devices, is based on manipulating and controlling a current flow by applying an electric field, which acts as a force on the charge of the electrons. Only a few devices, like relays, solenoids and Hall probes make use of a magnetic field. The fact that electrons have a magnetic moment (they are either spin-up or spin-down) as well as a charge has until recently led to very little practical application: "Conventional electronics has hitherto ignored the spin of the electron" [1].

Spin electronics, also known as spintronics or magnetoelectronics, is a new branch of physics where the electron spin (as opposed to, or in addition to the electron charge) is the active element manipulated in transport processes. This new field [2][3] combines two well established areas of research: magnetism and electronics. Devices based on spin electronics can potentially complement or replace various conventional electronic devices with an improved performance, or even find application in quantum computing and communication [4].

Efforts in spin electronics are directed at a wide range of issues. From a fundamental point of view, concepts such as spin-dependent transport, spin diffusion length, spin injection, spin accumulation and spin-induced magnetic switching are receiving a great deal of attention, while also the feasibility of novel device structures is studied. At the same time new materials are investigated, with special emphasis on half-metallic materials and magnetic semiconductors. All these topics are briefly introduced below.

In the 1930's Mott modelled the electric conduction in a ferromagnetic material by postulating that the current is carried via two distinct conduction channels, with the carri-

ers consisting of the spin-up electrons and the spin down electrons, respectively, and with different transport properties. For this to be a valid model, the conduction in the channels has to be independent, ie. that there is no mixing of the two channels. This means that the characteristic spin flip time has to be sufficiently long compared with other scattering processes.

1.1.1 Spin polarization

It is useful to now introduce here the concept of spin polarization. For spin electronics to work one needs a source of spins to manipulate. Ferromagnetic materials are obvious candidates, with their spin dependent band structure, with the majority subband containing electrons with spin parallel to the magnetization direction, and the minority subband where the spins are antiparallel. The spin polarization P is a measure of the ratio between spin-up and spin-down electrons 'available' at the Fermi level. Several definitions are possible, depending on which physical processes are being considered. Perhaps the most natural definition is:

$$P_n = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}}. \quad (1.1)$$

where P_n is now defined as the normalized difference between spin-up and spin-down density of states ($n_{\uparrow}$ and $n_{\downarrow}$ respectively) at the Fermi level. This definition may be appropriate when the spin polarization is determined by photo emission experiments, which directly extract electrons from near the Fermi level (See Figure 1.1). When transport properties are of interest, for example in magnetic tunnel junctions or Andreev reflection experiments (section 1.1.2), one should define the spin polarization in terms of the current densities J . In

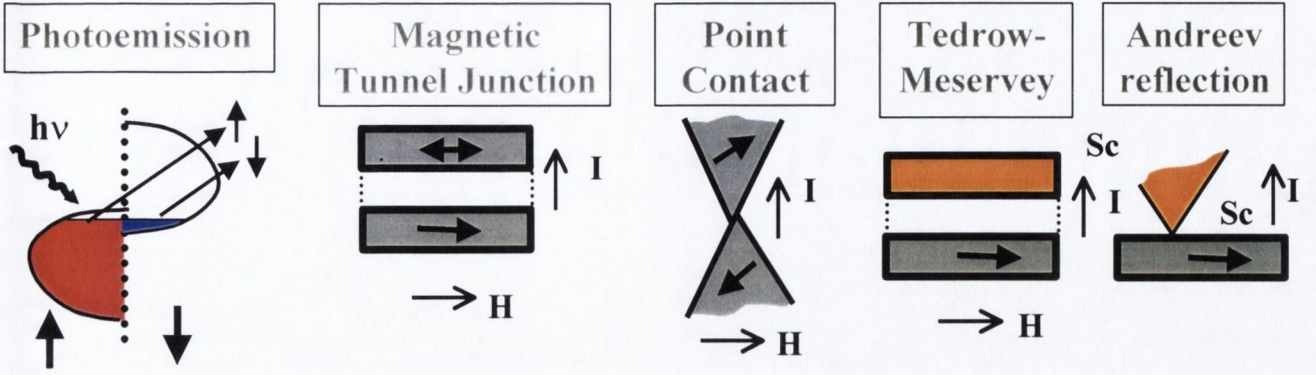


Figure 1.1. Several ways to determine the spin polarization

the case of ballistic transport, the spin polarization is defined as:

$$P_{nv} = \frac{J_{\uparrow} - J_{\downarrow}}{J_{\uparrow} + J_{\downarrow}} = \frac{\langle nv \rangle_{\uparrow} - \langle nv \rangle_{\downarrow}}{\langle nv \rangle_{\uparrow} + \langle nv \rangle_{\downarrow}}, \quad (1.2)$$

with v the wave velocity. For diffusive transport, or tunneling through a large barrier this becomes [5]:

$$P_{nv^2} = \frac{\langle nv^2 \rangle_{\uparrow} - \langle nv^2 \rangle_{\downarrow}}{\langle nv^2 \rangle_{\uparrow} + \langle nv^2 \rangle_{\downarrow}} \quad (1.3)$$

Materials with a 100 % spin polarization are known as half-metals, since only electrons with one spin orientation are available for transport at the Fermi level.

1.1.2 Andreev Reflection

An important technique to measure spin polarization is based on Andreev reflection. Andreev reflection is the process that converts normal current into supercurrent at the interface between a normal metal and a superconductor. In a normal metal the current is carried by electrons, while in superconductors charge transport is governed by Cooper pairs. The top part of Figure 1.2 shows a simplified energy diagram of a normal metal (polarisation $P = 0\%$) and a superconductor. An electron in the up-band of the normal

metal is impinging on the interface. Since there are no single particle states available within the superconducting gap Δ , for it to enter into the superconductor, and become part of the supercurrent, it must be a member of a Cooper pair. This means that the other electron of the pair, with spin down, leaves a hole behind at the interface. This hole, with momentum and spin opposite to that of the spin-down electron, will be Andreev reflected back into the metal, and will be part of a parallel conduction channel. This leads, for voltages less than Δ/e , to a doubling of the conductance $G = dI/dV$ compared to the normal state case.

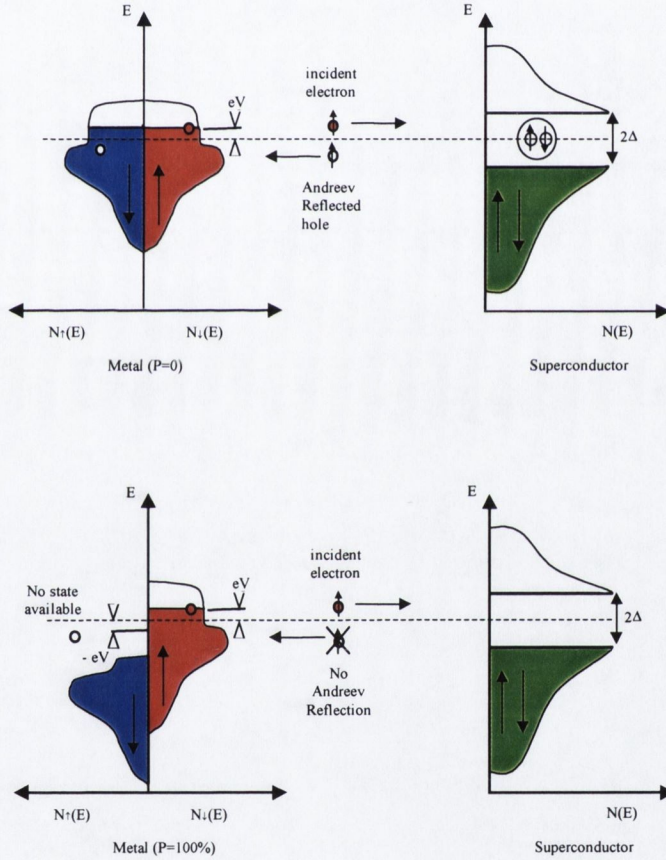


Figure 1.2. Andreev reflection at a normal metal / superconductor interface (top) and at a half metal / superconductor interface (bottom).

Now consider the bottom part of Figure 1.2, the case of a fully spin polarized-metal, i.e. $P = 100\%$ and a superconductor. Again, an electron from the up band approaches the

normal metal / superconductor interface. This time, a Cooper pair cannot be formed, since the density of states near the Fermi level in the down band is zero: there are no spin-down electrons available. So, Andreev reflection is not possible, and the conversion from normal current into supercurrent is blocked. This means that in the case of a fully spin polarized metal the conductance $G = 0$ for voltages smaller than Δ/e . So, for $P = 0$ the conductance is doubled compared to the normal state conductance, because of Andreev reflection at the interface, while for $P = 100\%$ Andreev reflection is blocked, leading to a suppression of the conductance. For intermediate values of the polarization, a modified version of the Blonder-Tinkham-Klapwijk (BTK) theory [6] is needed to determine the value of P from the measured dI/dV curve. The BTK theory takes interfacial scattering into account, through a parameter Z , with $Z = 0$ for ballistic transport, and $Z \rightarrow \infty$ for tunnel junctions. For $T \rightarrow 0$, $V \rightarrow 0$ and $Z \rightarrow 0$, the theory yields:

$$\frac{1}{G_n} \frac{dI}{dV} = 2(1 - P_c), \quad (1.4)$$

where G_n is the conductance in the normal state case ($V > \Delta$), and P_c is the contact polarization:

$$P_c = \frac{N_\uparrow(E_F) v_{F\uparrow} - N_\downarrow(E_F) v_{F\downarrow}}{N_\uparrow(E_F) v_{F\uparrow} + N_\downarrow(E_F) v_{F\downarrow}}, \quad (1.5)$$

with N_σ the spin dependent density of states. In this case P_c can be obtained directly from the dI/dV at $V = 0$, while for finite temperatures and interface scattering a numerical fitting is needed.

Material	NiFe	Co	Fe	Ni	NiMnSb	LSMO	CrO ₂
$P_c(\%)$	37	42	45	46.5	58	78	90

Table 1. Spin polarization for different materials, after Soulen *et al.* [7]

This procedure was first used by Soulen *et al.* [7] to obtain the spin polarization for a

variety of materials, see Table 1. Here a superconducting tip is driven into the material under investigation (or vice versa: a tip of the material is driven into a superconducting film), and the current-voltage characteristics measured at liquid helium temperatures. The values of P_c thus obtained for the 3d metals are a little higher than those reported by Tedrow and Meservey [8], who measured the (tunneling) polarisation using ferromagnetic-insulator-superconductor junctions. However, this technique is sensitive to the nature of states in the first few monolayers of the ferromagnetic electrode [9], while the method described above probes the polarization of the bulk current. Both LSMO and CrO_2 show a high spin polarization, close to the value expected for a half metallic ferromagnet. The complete spin polarization of CrO_2 was subsequently confirmed in Andreev [10] and tunnel junction experiments [11].

1.1.3 Spin Accumulation and Diffusion

Assume a current flowing from a ferromagnetic material into a paramagnet. The electrons coming from the ferromagnet are spin polarized, say, there are more spin-up electrons than spin-down. So, there is a net influx into the paramagnet of spin-up electrons over spin-down ones. This surplus is causing a certain magnetic moment per volume in the paramagnetic material near the interface. At the same time, spin flip scattering will tend to flip the up-spins into down-spins. These two competing processes will lead to a dynamic equilibrium, with a spin accumulation near the interface, which will drop off over a certain length scale known as the spin diffusion length (See Figure 1.3). The spin diffusion length is strongly dependent on the amount of impurities in a material since these introduce more spin-flip scattering. Typical spin diffusion lengths vary from microns in very pure metals to 10 nm in silver doped with 1 % Au impurities.

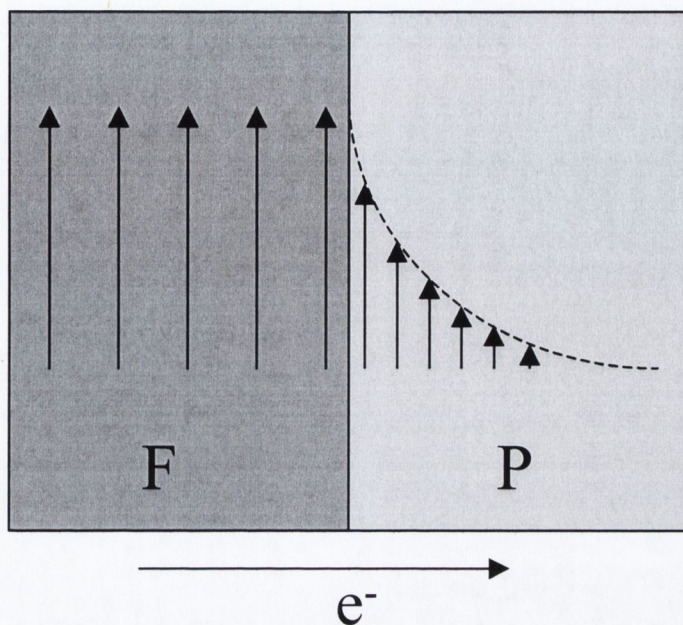


Figure 1.3. Spin accumulation occurs when a spin-polarized current from a ferromagnet (F) enters a paramagnet (P).

1.2 Basic Concepts of Transport

The nature of transport in small, or mesoscopic, structures is dependent on the mutual relationship between the various length scales involved. The most obvious one is the size of the system, L . The second one is the mean free path. Charge carriers, or more simply electrons, will travel, on average, a certain length before being scattered. This scattering can be caused by dislocations in the crystal, impurities or sample boundaries, in which case it will be elastic scattering: no energy is exchanged. Other scattering mechanisms are phonon scattering and electron-electron interactions which are inelastic. The typical length an electron will travel before it is scattered is called the mean free path λ_{mfp} . One can define an elastic mean free path, l_e , which is the length travelled between two elastic scattering events, and likewise an inelastic mfp, l_i . Finally, we can associate a length scale with the wave character of the electrons: the Fermi wave length λ_F , which can be related to

the Fermi velocity $v_F = \frac{h}{m\lambda_F}$.

We can now classify the two distinct transport regimes in mesoscopic structures as follows. When $\lambda_F \ll L < l_e, l_i$, the electrons are said to behave ballistically, that is, they travel through the structure without being scattered. The other limit, $\lambda_F < l_e, l_i \ll L$, is the diffusive regime, where the electron undergo many scattering events while traversing the structure.

1.2.1 Ballistic Transport and Quantum Conductance

The resistance of a point contact has been calculated by Sharvin [12]. It is found that for a small point contact, i.e. with a radius r smaller than the mean free path λ_{mfp} , the resistance R is given by

$$\frac{1}{R} = \frac{2e^2}{h} \frac{k_F^2 A}{4\pi}, \quad (1.6)$$

with k_F the Fermi wave vector and $A = \pi r^2$ the area of the point contact. For a contact with $r = 10$ nm this would give a resistance of 1Ω .

Quantization of the conductance through a small constriction was first seen by Van Wees *et al.* [13] in a two dimensional electron gas (2DEG) when the width of this constriction is varied. This quantization has subsequently been observed in metallic nanocontacts, using a wide range of experimental techniques. Widespread use is made of a STM, where the tip is driven into the sample, and then pulled back. This causes a nanowire to be formed, a behaviour similar to pulling some chewing gum apart. Both the electrical and mechanical properties of these nanowires can then be measured [14][15][16][17]. Mechanically-controlled break junctions also offer accurate control over the formation of nanowires [18]. Experimentally simpler techniques such as observing macroscopic wires vibrating in and out of contact

[19], and electromagnetic relays [20] also show clear evidence for quantized conductance.

The quantum of conductance found is $G_0 = \frac{2e^2}{h} \approx (12.9 \text{ k}\Omega)^{-1}$ where e is the charge of an electron, and h the Planck constant. The conductance quantization is caused by the quantization of electron momentum in directions perpendicular to that of current flow. Consider two electron gas reservoirs connected by a narrow constriction with a diameter a , see Fig 1.4. We assume that the electrons move ballistically between the two reservoirs,

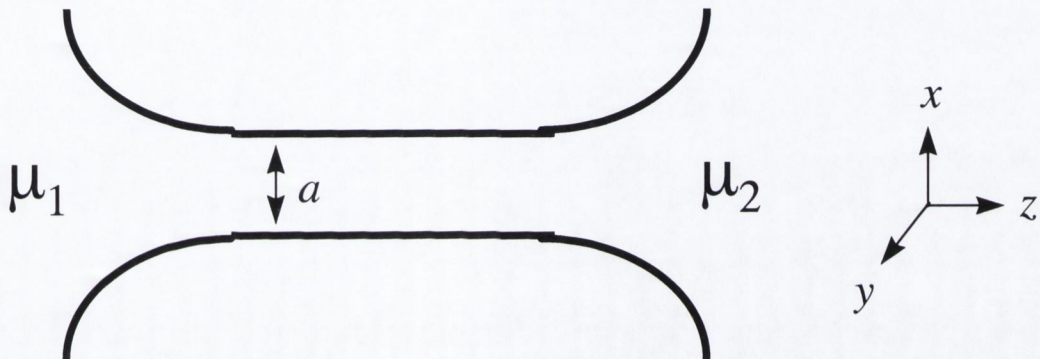


Figure 1.4. Two reservoirs with chemical potential μ_1 and μ_2 connected by a constriction of size a .

and are only scattered by the boundaries of the constriction. This means that the electrons behave like a one-dimensional electron gas: in the z -direction the electron momentum is continuous, while in the x - and y -directions, due to the small size a of the constriction, the momentum is quantized. The energy of the electron can then be written:

$$E_n(k) = (n_x^2 + n_y^2) \frac{\pi^2 \hbar^2}{2ma^2} + \frac{\hbar^2 k_z^2}{2m}, \quad (1.7)$$

with n_x and n_y the indices for the allowed transverse modes, $\hbar$ the reduced Planck constant, m the electron mass, and k_z the component of the wavevector in the z -direction. The first term on the right hand side of equation (1.7) is the quantized term, while the other term represents the continuous contribution. Applying a small potential difference between the two reservoirs, $V_{bias} = (\mu_1 - \mu_2)/e$, leads to a difference in chemical potential $(\mu_1 - \mu_2)$.

The number of electrons N then contributing to the current is:

$$N = \frac{1}{2} g_n(E_F) (\mu_1 - \mu_2), \quad (1.8)$$

with $g_n(E_F)$ the density of state at the Fermi level for mode n . This leads to an expression for the charge current I_n due to mode n :

$$I_n = ev_{Fn} N = \frac{ev_{Fn}}{2} g_n(E_F) (\mu_1 - \mu_2), \quad (1.9)$$

with v_{Fn} the group velocity of mode n at the Fermi level. The density of states at the Fermi level for a 1D electron gas is given by:

$$g_n(E_F) = \left(2\pi \frac{dE_n(k)}{dk} \right)^{-1} = \frac{1}{\hbar v_{Fn}}. \quad (1.10)$$

Finally, substituting equation (1.10) into (1.9) gives a conductance G_0 per mode of:

$$G_0 = \frac{I_n}{V_{bias}} = \frac{2e^2}{h}, \quad (1.11)$$

or for the total conductance G :

$$G = NG_0 = \frac{2e^2}{h} N, \quad (1.12)$$

with N the number of available modes below the Fermi level.

When changing the size of the constriction, the number of allowed modes available for transport changes too, and this leads to a stepwise change in the observed conductance. When the size of the constriction is small enough (of the order of Ångströms), the energy level spacing becomes larger than the thermal energy, and conductance quantization can be observed at room temperature.

In the above derivation it is assumed that each electron emitted from the, say, left reservoir, will reach the right reservoir. This implies a complete transmission for each electron

wave in each mode. In general, this condition need not hold, and this leads to the Landauer formula, a generalization of equation (1.12)

$$G = \frac{e^2}{h} \left(\sum_1^{N_\uparrow} T_{i\uparrow} + \sum_1^{N_\downarrow} T_{i\downarrow} \right), \quad (1.13)$$

where the conductance is the sum over all available modes of the spin-dependent transmission probabilities T_i . For systems with spin degeneracy, and completely open or closed modes (T_i either 0 or 1), the Landauer formula reduces to equation (1.12).

1.2.2 Diffusive transport and hopping

When an electron is scattered many times while traversing a system, it is said to behave diffusively. Drude's expression for the conductivity σ of such a system,

$$\sigma = \frac{\vec{j}}{\vec{E}} = \frac{e^2 n_e \tau}{m}, \quad (1.14)$$

is based on the assumption that the electrons can be viewed as an ideal gas. Here electrons are seen as particles that can move freely in between occasional scattering events, where they lose all information about energy, direction and velocity after each event. The electrons are moving randomly, with the mean velocity zero. Application of an electric field will accelerate the electrons, leading to a mean velocity, also known as the drift velocity.

Hopping is a form of transport where charge carriers, be they holes, electrons or small polarons, are localized and hop from one atomic site to another. The charge carriers will have to overcome an energy barrier to move from one site to the next. This activation energy leads to an temperature dependence of the conductivity of the form $\sigma \sim \exp(-b/T^\alpha)$, where the factor α ranges from $\frac{1}{4}$ to $\frac{1}{2}$. This hopping can also be seen as thermally-activated internal tunneling [21].

1.2.3 Magnetoresistance

The change in resistance of a material when applying a field is known as the magnetoresistance (MR). It is often expressed as

$$MR(H) = \frac{R(0) - R(H)}{R(0)} \times 100\%, \quad (1.15)$$

with $R(H)$ and $R(0)$ the resistance with and without applied field H . The effects of a magnetic field can influence the transport properties of materials via several physical mechanisms. The most well-known are normal, anisotropic and giant magnetoresistance.

Normal, or Lorentz magnetoresistance (NMR) is related to the Hall effect. Charge carriers in a field are deflected from the direction of the current because of the Lorentz force $\vec{F} = e \vec{v} \times \vec{B}$, and this reduces their effective mean free path causing a change in resistivity of the material. To lowest order, the NMR has a B^2 dependence, with the largest effect found in low resistivity materials, where the mean free path is long and the electrons travel a significant part of a cyclotron orbit before scattering (limit $\omega_c \tau > 1$, where $\omega_c = eB/m$).

Anisotropic magnetoresistance (AMR) finds its origins in the spin-orbit coupling, which causes the spin-up and spin-down states to be mixed. It can be determined by measuring the resistivity change as function of the angle between current direction and magnetization. The AMR values for manganites and Fe_3O_4 at room temperature are low ($\sim -0.2\%$, with a peak of $\sim -2\%$ at the Curie temperature for LCMO [22]; -0.2% for Fe_3O_4 [23]).

In 1988, Baibich *et al.* [24] found an MR of 50 % in Fe/Cr multilayers, at 4.2 K. This effect was dubbed giant magnetoresistance (GMR). It is strongly dependent on the thickness of the layers, due to the oscillatory exchange coupling between the ferromagnetic layers. In simple terms, GMR can be explained in the two-current model by spin dependent scattering

of the electrons as they travel through the multilayer.

1.3 Materials

1.3.1 Manganese oxides

Mixed valence manganites, $R_{1-x}^{3+}A_x^{2+}Mn_{1-x}^{3+}Mn_x^{4+}O_3^{2-}$, where R is a rare earth and A is a transitional metal (see Figure 1.5.), have generated a great deal of interest recently, due

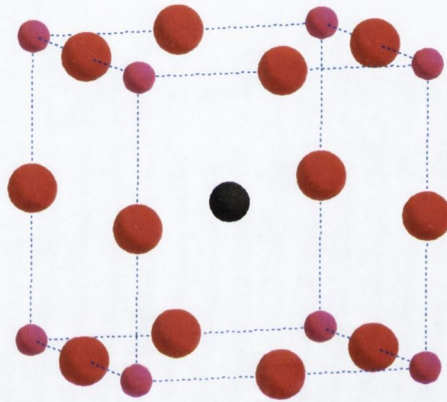


Figure 1.5. The ideal perovskite crystal structure, ABO_3 . Here A is a rare earth (La^{3+}) or transition metal (Sr^{2+}) (black), and B is Mn^{3+} or Mn^{4+} , (pink). The oxygen is shown in red.

to their peculiar magnetic and transport properties [25]. End members, like $La^{3+}Mn^{3+}O_3^{2-}$ and $Ca^{2+}Mn^{4+}O_3^{2-}$ are insulating and anti-ferromagnetic, while compounds with values of $x \approx 0.3$ are ferromagnetic below the Curie temperature and undergo an insulator to metal transition. The Colossal Magnetoresistance (CMR) effect is closely related to this transition [26]. Zener [27] proposed the double exchange mechanism to explain the ferromagnetism of these mixed valence manganites. Double exchange is the name given to the process where an electron is simultaneously transferred from an Mn^{3+} to an oxygen atom and from the

oxygen to the neighbouring Mn^{4+} . Due to the strong intra-atomic Hund's coupling the electrons can only hop if the spins of the manganese ions are parallel. The configurations $\text{Mn}^{3+}-\text{O}^{2-}-\text{Mn}^{4+}$ and $\text{Mn}^{4+}-\text{O}^{2-}-\text{Mn}^{3+}$ are degenerate; the lowest energy of the system at low temperature corresponds to a parallel alignment. The transfer of electrons becomes more difficult if the spins of the manganese ions are not parallel. This leads to a direct relation between conductivity and ferromagnetism. A core of localised spin polarised electrons form the t_{2g} band, while the conduction band (the e_g band) contains less than one electron per Mn ion. A more thorough treatment [28] shows that in some cases (like CrO_2 and LSMO) the conduction band is split into a majority and minority bands until the conduction electrons remain in only one of the spin states. These materials are the half metallic ferromagnets. See Fig 1.6. There is some dispute about whether the lower edge of the spin-down band in LSMO lies just above [29], or below [30] the Fermi level E_F . However, even if it lies below E_F , the electrons in the band will still be localized, and the current is then carried by the spin-up electrons. This is a type IIIA half-metal [31].

However, the double exchange mechanism alone is not sufficient to explain the magnitude of the change in resistivity around T_C or the large resistivity for temperatures $T > T_C$ [32]. Recent work has focussed on mechanisms that localize the conduction electrons, such as the Jahn-Teller effect, charge fluctuations due to cationic disorder, or magnetic impurities.

The usefulness of the CMR effect for practical applications, however, is limited due to the large magnetic field required to observe the effect. However, polycrystalline samples show a low field magnetoresistance which is absent in single crystals [33]. This low field MR is therefore an extrinsic effect; it has been reported in bulk, polycrystalline thin films [34], and in films with artificial grain boundaries [35] [36][37], and is attributed to the presence of

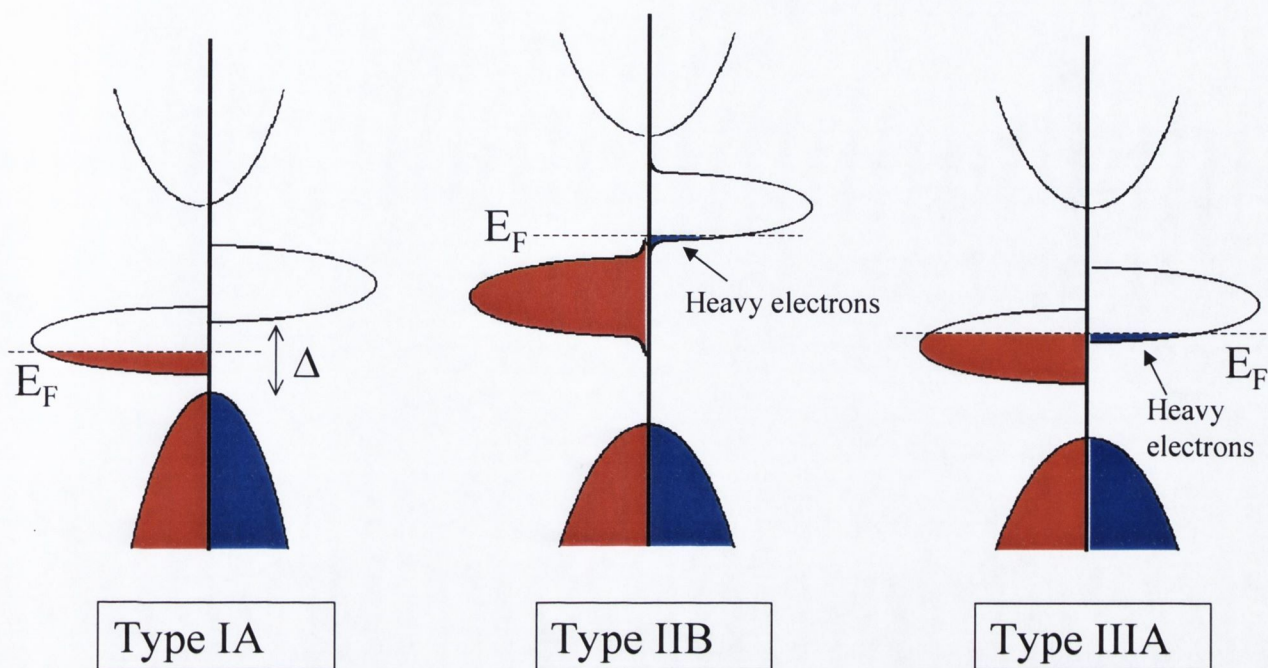


Figure 1.6. Schematic bandstructure of different types of half-metallic ferromagnets. For Type IA, only spin-down electron are present at the Fermi level, the density of states for spin-up electron is zero. CrO_2 is an example of this type half-metal. Magnetite is an example of a Type IIB half-metallic ferromagnetic, where the carriers at the Fermi level are in a band sufficiently narrow for them to be localized. LSMO is of type IIIA, with localized spin-up carriers and delocalized spin-down carriers. Classification after Coey *et al.* [31]

grain boundaries or crystallite interfaces. Compared with single crystals and epitaxial films the polycrystalline samples have a higher resistivity, presumably caused by the presence of grain boundaries. Two groups [35][36] devised an elegant experiment to separate the intrinsic CMR effect from the extrinsic low field effect. Whereas the CMR effect decreases with lower temperatures, the low field MR becomes larger for decreasing temperatures.

1.3.2 Magnetite

Magnetite, Fe_3O_4 , is the oldest magnetic material known to man. In the form of naturally magnetized lodestone it was used as a primitive compass by the Chinese as far back as 100AD, and it has been the subject of intensive study due to its interesting magnetic and transport properties [43][44]. Magnetite crystalizes in the cubic inverse spinel structure,

as shown in Figure 1.7, with the tetrahedral sites occupied by the ferric Fe^{3+} ions, and the octahedral sites shared by ferrous Fe^{2+} and Fe^{3+} ions. It can be seen as a face centered cubic lattice of oxygen anions with the iron cations situated on the interstices. The lattice parameter is $0.8398 \text{ \AA}$ [44]. From an ionic point of view, magnetite can be written as $\text{Fe}_A^{3+}[\text{Fe}^{3+}\text{Fe}^{2+}]_B\text{O}_4^{2-}$, where A and B denote the tetrahedral and octahedral sites, respectively. The presence of chains of octahedral sites along all $\langle 110 \rangle$ directions, leads to a low

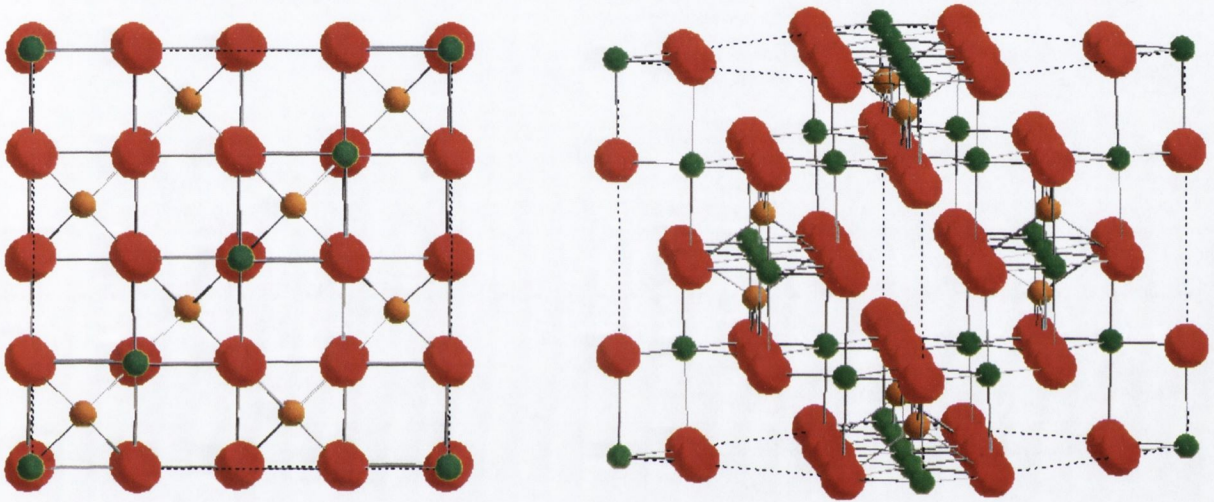


Figure 1.7. The magnetite (Fe_3O_4) crystal structure. The oxygen anions (red) form a face centered cubic lattice, with the ions cations situated on the interstices. The Fe ions sit at either octahedral sites (green) or tetrahedral sites (brown). The chains of octahedral sites in the $\langle 110 \rangle$ directions are clearly visible.

resistivity of around $5 \text{ m}\Omega\text{cm}$ at room temperature, due to rapid carrier hopping between Fe^{2+} and Fe^{3+} ions at these B -sites. Carriers are small polarons in a minority spin $3d^{\downarrow}(t_{2g})$ band [45]. All the electrons are localized, but only spin-down carriers are present at E_F . This is a Type IIB half-metal [31]. (See Figure 1.6). The electronic configuration of the Fe^{3+} ions is $3d^5$, whereas that of the Fe^{2+} ions is $3d^6$. This means that the Fe^{2+} ions have one extra electron in an anti-bonding t_{2g} orbital, and it is this electron that can hop from site to site. The electronic structure of the Fe ions is shown in Figure 1.8. This, however, is only possible

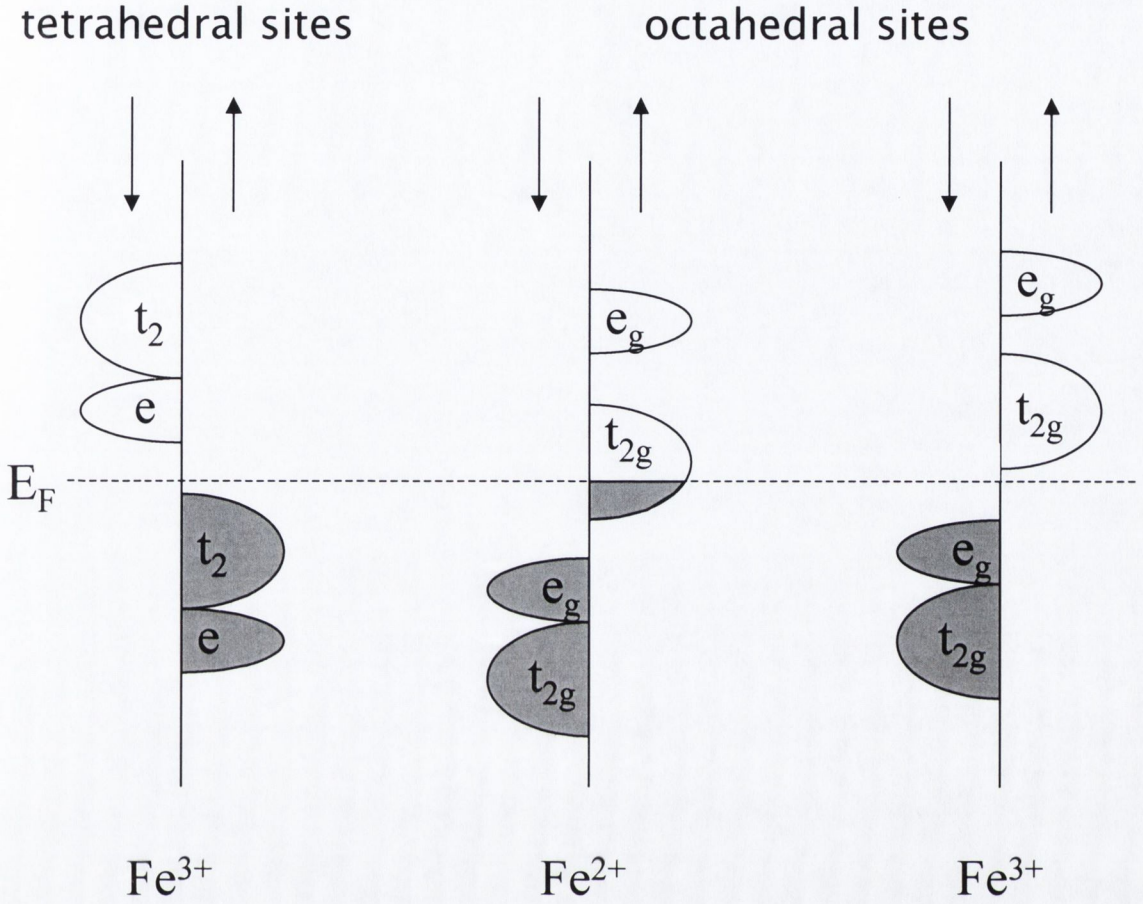


Figure 1.8. A schematic representation of the electronic structure of the Fe ions in magnetite.

when the two neighbouring *B*-sites are aligned ferromagnetically which is indeed the case for bulk magnetite, due to the dominant antiferromagnetic *A-B* superexchange coupling. At a certain temperature, called the Verwey temperature, $T_V = 120$ K, the resistivity increases by about two orders of magnitude. The details of this transition are not exactly understood [46], but it is generally agreed that at T_V the hopping freezes out, leading to an ordered array of Fe^{2+} and Fe^{3+} ions with static charges. Below the Verwey temperature the resistivity roughly follows a $T^{-\frac{1}{4}}$ law, evidence for variable range hopping [47].

Magnetite is a ferrimagnet with a Curie temperature of 850 °C. Both the Fe^{2+} and Fe^{3+} ions are in the high-spin state, i.e. $S = 2$ and $S = \frac{5}{2}$, respectively. One half of the

Fe^{3+} ions occupy tetrahedral A -sites, the other half occupy half of the octahedral B -sites. Since these two sublattices are coupled antiferromagnetically via superexchange, their $5 \mu_B$ moments cancel out, and the total net moment of $\sim 4 \mu_B$ per formula unit Fe_3O_4 comes from the Fe^{2+} ion on the octahedral B -site.

1.4 Scanning Tunneling Potentiometry

Ever since the first Scanning Tunneling Microscope (STM) was built by Binnig *et al.* [50], the number of applications has increased tremendously. It has been used to study the structural and electronic properties of conductive or semiconductive surfaces in air, liquid and UHV environments, with atomic or near atomic resolutions. STM's have also been used for lithography, and even for manipulating single atoms. Scanning Tunneling Potentiometry (STP) is an experimental technique that expands a STM to simultaneously image the surface topography and map the potential distribution of the material under study. STP was first developed by Murali and Pohl [51] to study electrical transport in discontinuous gold films. Semiconductors, heterojunctions [52], metals [53] and high- T_C superconductors [54] have also been investigated, to probe variations in the potential near film discontinuities and grain boundaries, and localized defects. Convolution between the tip shape and the surface topography can lead to artifacts in the potential distribution [55], but careful experiments have unambiguously shown steps in the potential near grain boundaries and crystallite interfaces.

In this work we use STP to show that the resistivity of polycrystalline LSMO films comes mainly from the grain boundaries, and that steps in the local potential coincide with these grain boundaries. Section 3.2 deals with the experimental set-up, the electronics and the data acquisition software that was developed. In Section 4.1 we present our results on

polycrystalline and epitaxial LSMO thin films, which will be discussed in section 5.1.

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Chapter 2

Literature Review

In recent years there has been a growing interest in transport on a mesoscopic scale. On the one hand this interest is driven by the need to understand the physics of devices with decreasing dimensions, due to the miniaturisation in the information and communication technology industry. On the other hand, the advances in lithography and patterning techniques, as well as the rise of scanning probe microscopy, has made this study of transport properties in mesoscopic samples possible. In this chapter we review the literature on transport through grain boundaries (Section 2.1), and artificially made nanowires and nanocontacts (Section 2.2). We will conclude with a brief overview of how domain walls in small devices differ from those in bulk, the role domain walls play in transport on a mesoscopic scale, and how they interact with charge carriers (Section 2.3).

2.1 Grain Boundaries

A precise understanding of the nature of grain boundaries and their transport properties is still lacking, but several models have been proposed [1]-[6]. From one point of view, the grain boundary is considered to be an insulating layer, of order a nanometer wide, and transport takes place through spin dependent tunneling. The opposing view is that a low resistance grain boundary acts as interface between two ferromagnetic grains, and it is the scattering at this interface that leads to GMR type magnetoresistance [7]. Transport measurements of grain boundaries show non-linear IV curves that agree with the Glazman-Matveev model [8], suggesting resonant tunneling through grain boundaries via localized defect states. In this model, the grain boundary is modelled as an insulator, with one or more localized defects

inside the barrier. When the Fermi level of the electrode coincides with the energies of these localized states, the conductivity is resonantly enhanced. Lattice mismatch between two neighbouring grains will lead to dislocations and stress that can result in a disordered region a few nanometers wide. It has been suggested that this region will have a reduced T_C , so that it may be paramagnetic [6].

2.2 Nanowires and Nanocontacts

Quantization of electrical conductance has first been observed in gated structures in two-dimensional electron gas systems [9][10], where it is found that the conductance varies in a step-like manner, with conductance values of integer multiples of $G_0 = 2e^2/h$. In the Landauer formalism the conductance G of a contact is expressed as

$$G = \frac{e^2}{h} \left(\sum_1^{N_\uparrow} T_{i\uparrow} + \sum_1^{N_\downarrow} T_{i\downarrow} \right), \quad (2.1)$$

with $T_{i\uparrow,\downarrow}$ the transmission for channel i with spin up or down, as discussed in Section 1.2.1.

The total conductance is determined by summing over all channels. For ballistic contacts with spin degeneracy (having T_i either 0 or 1), this reduces to $G = N \frac{2e^2}{h} = NG_0$.

Since the first observation of quantized conductance in 2-DEGs, the effect has also been observed in simple metallic nanowires such as Au, Na, Pt, Ni, Bi, [11][12][13][14][15] and also in manganites [16]. For these measurements, a wide range of experimental techniques have been used, such as the scanning tunnelling microscope [15], mechanical break junctions [14], and even simple table-top set-ups using macroscopic wires. Using an STM inside a transmission electron microscope, Ohnishi *et al.* [18] were able to study the relation between conductance and structure of gold nanocontacts. The influence of structure on the conductance of the contact has also been recognized by Van Ruitenbeek and coworkers [19][20], who show that

the steps in conductance are related to rearrangements of the contact.

For monovalent metals, such as Au, the conductance is directly related to the number of atoms in the neck of the contact. For materials with a higher chemical valence, for example Al, the relation is not so straightforward, as shown by Scheer *et al.* [21]. Current-voltage characteristics of superconducting Al nanocontacts were measured and fitted to show that even for the smallest obtainable contacts, with $G < G_0$, three channels (each with $T_i < 1$) contribute to the transport. For Au nanocontacts they find that the smallest contacts contain only one channel. Similar results have been found in Nb and Pb contacts [22]. Other work, by García-Mochales *et al.* [23] has shown that the shift of the peaks in conductance histograms, away from integer values of G_0 , can be explained by the existence of internal disorder in the nanocontacts.

From Landauer's formula (2.1) it could be concluded that for magnetic materials, with the spin degeneracy lifted, for example by applying a field, the conductance could vary with step sizes of $\frac{1}{2}G_0 = e^2/h$. A few papers have been published, both theoretical and experimental, on this topic [24][25][26]. Ono *et al.* find quantization of conductance in units of G_0 for Ni nanowires in zero field, but observe quantization in units of $\frac{1}{2}G_0$ when the magnetization of the sample is almost completely saturated in a field of 6.7 mT. This suggests that domain walls play a role, which is indeed confirmed by theoretical work [25][26]. Finally, it is worth mentioning that the peaks at $0.5 G_0$ and $1.5 G_0$ in conductance histograms have even been found in non-magnetic Au contacts under electrochemical potential control [27].

It is a remarkable experimental fact, which seems not to be fully understood, that in many nanowires and nanocontacts the conductance does seem to be quantized at approximately integral multiples of G_0 .

2.3 Domain Walls

In ferromagnetic materials the exchange interactions, due to Pauli's exclusion principle, tend to align the magnetic moments parallel to each other. In an infinite bulk sample, the ground state would therefore be a homogeneously magnetized single domain. In finite samples however, one single domain would lead to magnetic charges at the surface, and with it shape demagnetizing fields. This increases the magnetostatic energy of the system. In order to minimize the total energy of the system, multiple domains will form, with alternating directions of magnetization [28]. In between the domains, the magnetization vector has to rotate from one direction to another and the resulting magnetic structure is called a domain wall (Figures 2.1 and 2.2). Obviously, the magnetization in a domain wall is not uniform,

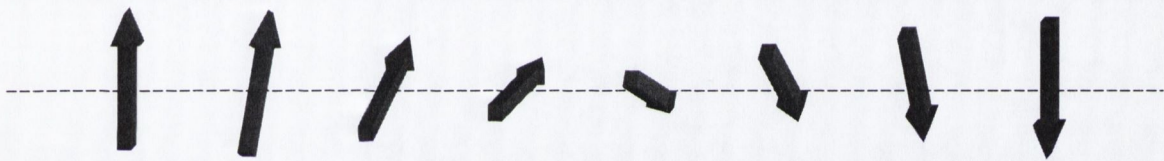


Figure 2.1. A Bloch wall, where the magnetization rotates out of plane from one direction to another.

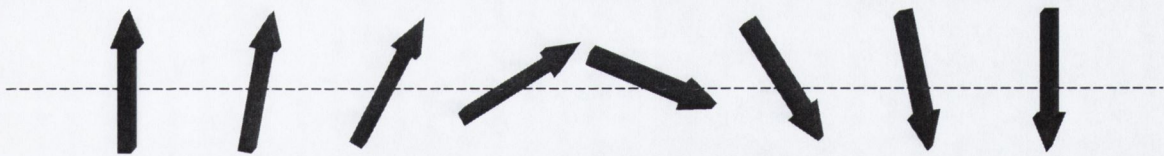


Figure 2.2. A Néel wall, where the magnetization rotates in plane from one direction to another.

leading to an increase in exchange, anisotropy and dipole energies. The process of domain formation is therefore a balance between the energy cost of forming the domain walls on the one hand, and the decrease in the magnetostatic energy on the other hand. Section 2.3.1 investigates how the geometric constraints of a sample influence the formation, configuration

and location of domain walls. Section 2.3.2 reviews theoretical and experimental studies on the resistivity of a domain wall, while Section 2.3.3 finishes this chapter with a discussion of the interactions of a domain wall with a spin and/or charge current.

2.3.1 Domain Walls in nanowires and contacts

Bloch [29], followed by Landau and Lifshitz [30] was the first to study theoretically the structure of a domain wall in the bulk. The width of the wall is determined by how quickly the moments rotate from one direction to the other. The exchange energy, which favors a parallel alignment of the moments, tends to broaden the wall, with only a very gradual rotation of the magnetization vector. The anisotropy favors an abrupt change and hence a narrow wall. The total energy E of the wall, is given by:

$$E = E_{ex} + E_a = \int_{-\infty}^{\infty} \left[A \left(\frac{\partial \theta}{\partial z} \right)^2 + f_a(\theta) \right] dV, \quad (2.2)$$

with A the exchange stiffness, f_a the anisotropy energy density and θ the rotation of the magnetization between two adjacent moments. A stable wall configuration requires a minimization of the total energy with respect to the rotation of magnetization: $\frac{\partial E}{\partial \theta} = 0$. This is equivalent with the Euler equation for a domain wall:

$$\frac{\partial f_a}{\partial \theta} - 2A \frac{\partial^2 \theta}{\partial z^2} = 0. \quad (2.3)$$

For the uniaxial anisotropy case, with $f_a = K_u \sin^2 \theta$, where K_u is the uniaxial anisotropy constant, the wall profile can be calculated analytically:

$$\theta(z) = \arctan \left[\sinh \left(\frac{\pi z}{\delta} \right) \right] + \frac{\pi}{2}, \quad (2.4)$$

where $\delta_w = \pi(A/K_u)^{\frac{1}{2}}$ is the domain wall width. Typical values for δ_w in bulk are given in table 1. An extensive discussion of domain walls is given in Ref. [31] and references therein. In

Material	Ni	Fe	Co	Permalloy	Rare earth magnet
δ_w (nm)	72	30	15	1000	5

Table 1. Values for the domain wall width for a variety of materials, in nm.

the above calculations, the geometry of the sample is assumed to be of no importance. With the increasing miniaturization of devices and bit size in the magnetic recording industry, the question is if this is still valid. Bruno [32] addressed this issue, and calculated the structure and properties of a geometrically constrained domain wall in a constriction separating two wider regions. He found analytical expressions for the domain wall width in three different models of constriction:

Model	Constriction	δ_w
I	$S = \begin{cases} S_0 & \text{for } x \leq d \\ S_1 > S_0 & \text{for } x \geq d \end{cases}$	$\frac{8d}{\pi^2}$
II	$S = S_0(1 + \frac{x^2}{d^2})$	$\frac{8d}{\pi}$
III	$S = S_0 \cosh(x/d)$	$2d$

In these constrictions the domain wall width has become independent of material parameters such as the exchange stiffness and anisotropy constant, and depends only on the geometry. One can see that δ_w is now of the order of the size of the constriction d . These results are confirmed by Monte Carlo simulations of "isthmus" and "hourglass" geometries (similar to Model I and II, respectively) [33]. These simulations also show that the wall goes from a Néel-like configuration for narrow constrictions, via a cross-over region to Bloch-type for larger constrictions. In the bulk, the domain wall width δ_w is much larger than the Fermi wavelength λ_F , whereas for nanometric constrictions, the wall can become very narrow. This has important implications for the resistivity of the domain wall (See Section 2.3.2). The wall energy γ_w , which is $2\sqrt{AK}$ for a bulk wall, is now $\frac{\pi^2 AS_0}{2d}$ for the "isthmus" geometry - independent of the anisotropy constant K .

Ounadjela *et al.* have given a review of work on micromagnetic configurations in thin

films and submicron magnetic dots, rings and wires [34]. For the work described in this thesis, the domain structure at the surface and in particular in point contacts is of most importance.

Since the formation of a domain wall costs energy, walls tends to localize themselves in constrictions to minimize their surface area, and hence their total energy. A number of studies on this topic are reported in the literature. Hirohata *et al.* [35][36] patterned mesoscopic "bow-tie" structures in Permalloy thin films, and investigated the domain structure with MFM and Scanning Kerr effect microscopy in the demagnetized and remanent state. In the demagnetized (as grown) state, no domain walls were observed in the junction, whereas in the remanent state (after applying a 1 T field), walls were found in the central bow-tie structure, suggesting that the junctions trap the domain wall during the magnetization reversal process. Micromagnetic calculations by the same authors confirm these observations. Van Gorkom *et al.* calculated the domain structure in Co and Permalloy point contacts [37][38] and found that a Néel wall is formed in the constriction. More extensive investigation of magnetic configurations in nanocontacts are found in Ref. [39]. The authors considered two semi-infinite oppositely magnetized ferromagnetic bars, connected through a nanocontact. Depending on the form factor of the contact, a blurred Néel- or Bloch-type transition region over the length of the contact is found for a weak anisotropy. In the case of strong anisotropy, a head-to-head Néel wall is formed. Domain wall trap elements have been proposed for use in MRAM applications, due to their lower switching fields (~ 1 mT) and similar switching times (~ 1 ns), compared with simple rectangular elements [40]. Depending on the shape and size of the point contacts, thermally-excited fluctuations between a Néel-type and a Bloch-type domain walls with different chirality can occur [41].

Unconnected bow-tie structures show a complicated domain structure at the sharp end, which has also been observed by Schrefl *et al.* [42] in NiFe and Co thin film elements.

2.3.2 Domain Wall Resistivity

In recent years, there has been a growing interest in the question of how the presence of domain walls changes the resistance, compared to the single domain case. The matter is still not clearly resolved, and there is no agreement on the magnitude or even the sign of the effect. On the experimental side, numerous papers have appeared in the literature claiming positive as well as negative contributions to the resistance, while on the theoretical side theories have been proposed supporting both cases. Matters are further complicated due to the difficulty of separating domain wall effects from ordinary, or Lorentz, and other anisotropic magnetoresistive effects, related for example to the presence of closure domains.

Experimentally, a reduction of the resistance due to the presence of a domain wall has been reported in narrow Ni wires [43][44], in Fe and Co wires [45]-[49], and Co zigzag structures [51], while an increase of the resistance has been reported in Ni, Co, FePd, SrRuO₃ and manganite thin films [52]-[58], Co wires [59][60], Ni nanocontacts [61][62], Co and LSMO bridges [63][64], and NiFe crossed wires [65].

Taylor *et al.* [69] observed large magnetoresistance effects in Fe whiskers as far back as 1968. The resistance of the single crystals was found to drop sharply when a field was applied, and the sample went from a multidomain to a single domain state. The results were interpreted to be (partially) due to domain wall scattering. Experimental interest in domain wall resistance returned in 1996 following work by Viret, Gregg *et al.* [52][53]. Here, a large positive domain wall contribution to the resistance was reported for Ni and Co thin films. The magnetoresistance of a Co thin film with a stripe domain pattern was measured by ap-

plying the current perpendicular to the stripes, and the field perpendicular to the plane of the film. It was claimed that in this way the AMR contribution could be suppressed because current and magnetization remain perpendicular when a field is applied. This was contested by Rüdiger *et al.* [46], who, on the basis of MFM imaging and micromagnetic calculations, concluded that a saturating field will both erase DWs and reorient the magnetization with respect to the current and the crystal. Thus the assumption that the low field MR observed by Gregg *et al.* [53] is only due to domain wall resistance is not valid. The group of Rüdiger and Kent *et al.* have in a range of papers [45]-[50] carefully studied the magnetoresistance of Fe, Co and FePt microstructures. By measuring the magnetoresistance in different configurations and as a function of temperature, and in conjunction with MFM imaging and micromagnetic calculations, the various contributions to the magnetoresistance could be deduced and quantified. Figure 2.3 shows the MR as a function of field of a $2\text{ }\mu\text{m}$ wide and 100 nm thick Fe wire in transverse and longitudinal measurement geometries. A clear resistivity anisotropy is observed which in high fields changes sign with temperature. At 270 K (Fig. 2.3a), the resistivity at high fields in the longitudinal case is higher than in the transverse case, while at 1.5 K (Fig. 2.3b), it is the other way around. The anisotropy is due to a combination of Lorentz magnetoresistance and anisotropic magnetoresistance (AMR) which are of opposite sign. Lorentz magnetoresistance depends on the relative orientation of current and field, whereas AMR depends on the orientation of current and magnetization. At high temperatures the anisotropy comes mainly from AMR, lowering the temperature increases the importance of the Lorentz MR. At a certain temperature, T_{comp} , the two contributions cancel each other out, and the low field MR due to resistivity anisotropy vanishes. For the Fe wire studied here, the authors find $T_{comp} = 65.5\text{ K}$. MR measurements at this tempera-

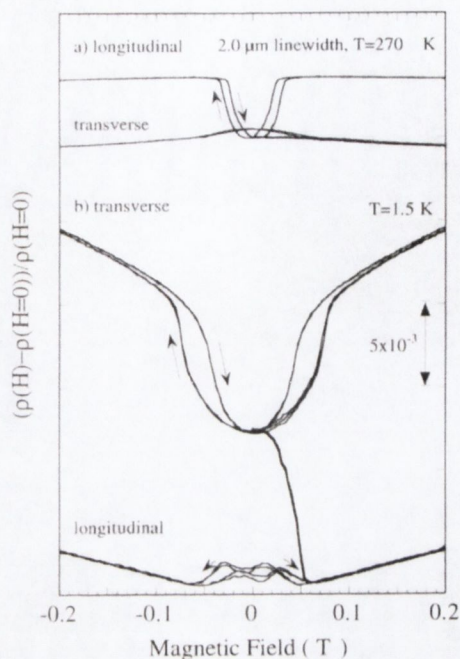


Figure 2.3. Magnetoresistance of a 2 μm wide and 100 nm thick Fe wire at a) 270 K and b) 1.5 K. In the longitudinal geometry the applied field is in-plane and parallel to the wire axis, while in the transverse geometry it is in-plane and perpendicular to the wire axis. After [50].

ture are presented in Figure 2.4. The extrapolation of the high field MR data shows that at T_{comp} the transverse and longitudinal resistivity at zero field become equal. The remaining MR at low fields can then be attributed to domain walls. Perhaps surprisingly, it is found that the presence of domain walls *reduces* the resistivity of the Fe wire. MFM observations of the domain states of the Fe wires after longitudinal or transverse saturation were used to estimate the domain wall contribution to the resistivity as function of linewidth and temperature (See inset Fig. 2.4). Similar experiments by the same group on Co show a small additional contribution to the resistivity due to the presence of domain wall. However a Hall effect mechanism could also explain the observed results. FePt wires show a clear enhancement of the resistivity (of order 0.1%) due to domain wall scattering. It is worth noting here that due to the high magnetic anisotropy, FePt has the narrowest domain walls.

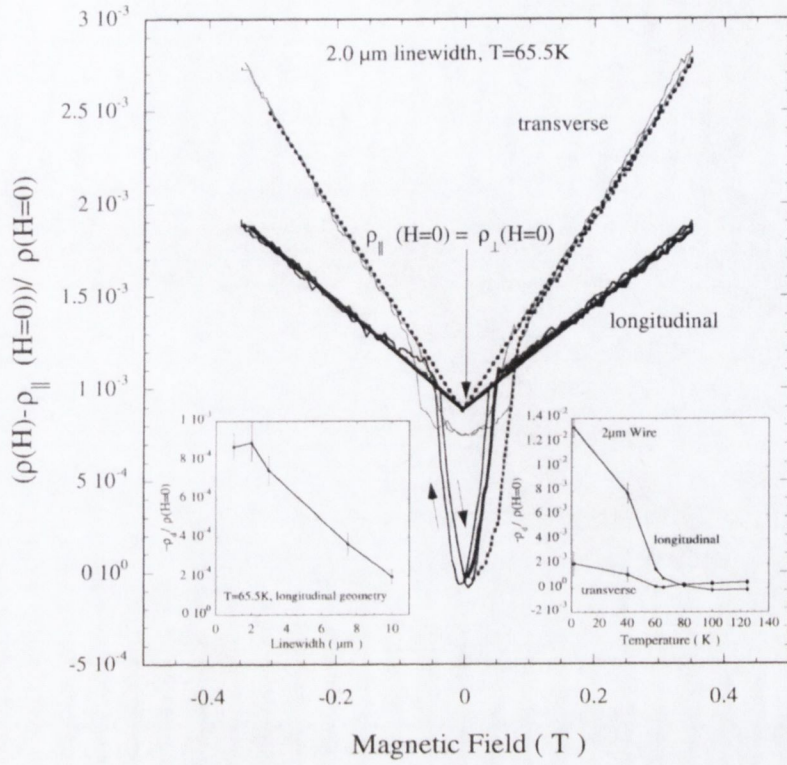


Figure 2.4. Magnetoresistance of a $2\ \mu\text{m}$ wide and $100\ \text{nm}$ thick Fe wire at $65.5\ \text{K}$. In the longitudinal geometry the applied field is in-plane and parallel to the wire axis, while in the transverse geometry it is in-plane and perpendicular to the wire axis. Insets show the negative domain wall contribution as function of linewidth (left), and temperature (right). After [50].

One of the earliest theoretical studies, by Cabrera and Falicov [66], used a free electron model to interpret transport through a domain wall as tunneling. The spin of an impinging electron will interact with the wall, partially reflecting it, leading to an *increase* of the resistance. It is found that this effect is small, when the exchange splitting is very small and when $\delta_w k_F > 1$, where δ_w is the wall thickness, and k_F the Fermi wave length. This condition is met in bulk where domain walls widths are of the order of tens of nanometers or more. Berger [67] noted that electrons traversing such a domain wall can adiabatically follow the spatial rotation of the magnetization, resulting in little scattering. When the exchange splitting is large, and the domain wall narrow, $\delta_w k_F \ll 1$, the increase in resistance becomes

appreciable. Cyclotron effects, ie. the "zig-zagging character of the electron orbits when going between up- and down-regions of the magnetization" [68], however, can explain the experimental magnetoresistance results found in iron whiskers [69].

Levy and Zhang [70] considered the role of scattering of impurities to explain the increase of resistance associated with the presence of a domain wall in the diffusive regime, as found by Gregg *et al.* [53]. Using the Boltzmann transport approach, they show that spin dependent scattering results in a mixing of the two heretofore independent current channels. It is this mixing that is the origin of the additional resistance of the domain wall, leading to a magnetoresistance of a few percent in bulk. When the current is perpendicular to the domain wall, the magnetoresistance is found to be

$$MR_{cpw} = \frac{\rho_{cpw} - \rho_o}{\rho_o} = \frac{\xi^2 (\rho_o^\uparrow - \rho_o^\downarrow)^2}{5 \rho_o^\uparrow \rho_o^\downarrow} \left(3 + \frac{10 \sqrt{\rho_o^\uparrow \rho_o^\downarrow}}{\rho_o^\uparrow + \rho_o^\downarrow} \right), \quad (2.5)$$

with $\rho_o^{\uparrow(\downarrow)}$ the resistivity for the spin up (down) channel, and ξ a measure for how well the electron can track the local magnetization. The parameter ξ can be seen as the ratio of the spin precession length and the domain wall width. Hence, the magnetoresistance varies as $1/\delta_w^2$. It should be noted that equation (2.5) is not valid for very narrow domain walls.

Tatara and Fukuyama [71][72], using linear response theory, identified two contributions to the resistivity caused by scattering off a wall in disordered systems. First, a (very small) positive Boltzmann resistivity, and secondly, a *negative* contribution of a domain wall to the total resistance, due to the decoherence of electrons in the presence of a wall, leading to a reduced weak localization of electrons, and an enhanced conductance. Calculations based on Landauer's formula [73] and computer simulations [74] are consistent with this dephasing effect of a wall. In a separate paper [75], Tatara *et al.* also studied how scattering by a domain wall modifies the electronic state, and found that "the wall in a ferromagnet with a

small exchange coupling can be squeezed to be very thin to lower the energy.”

Brataas *et al.* examined both ballistic and diffusive transport through a domain wall [76][77][78]. They find that in the 2-band Stoner model, ballistic wall scattering is very weak, but that it is enhanced significantly if realistic band structures are taken into account. For diffusive transport, it is found that by taking into account spin-dependent scattering times, qualitatively similar results as in Ref [70] are obtained. Further work by Van Gorkom *et al.* [79] showed that a contribution due to the spatial dependence of the magnetization in a wall should also be considered, and that the wall resistance can be negative as well as positive. Consider the Drude resistivity ρ of a two band Stoner ferromagnet:

$$\rho = \frac{m}{e^2} \frac{1}{n^\uparrow \tau^\uparrow + n^\downarrow \tau^\downarrow}, \quad (2.6)$$

with m the mass of the electron, e the electron charge, and $n^{\uparrow(\downarrow)}$ and $\tau^{\uparrow(\downarrow)}$ the spin-up (down) electron density and scattering relaxation time respectively. The change in magnetization in a wall is accompanied by a redistribution of the electrons between the spin-up and spin-down bands. Spin dependent scattering times lead then to a change in resistivity $\delta\rho$:

$$\delta\rho \approx -\rho^2 \frac{e^2}{m} \delta n^\uparrow (\tau^\uparrow - \tau^\downarrow), \quad (2.7)$$

where $\delta n^\uparrow = -\delta n^\downarrow$ is the change in spin-up electron density. The relaxation times, and therefore also the sign and magnitude of the resistivity change $\delta\rho$ depend on the type of impurities present in the material.

The work cited thus far concerned wide domain walls, ie. walls in bulk, $k_F \delta_w \geq 1$. Narrow walls were discussed by taking the limit $\delta_w \rightarrow 0$. Following the experimental work by García *et al.* [61] who found large magnetoresistance effects in ballistic Ni nanocontacts with $G \gtrsim 1G_0$, Tatara *et al.* [62] studied specifically the domain wall resistance in narrow

walls, and found a (ballistic) magnetoresistance effect MR :

$$MR = \frac{G_o - G}{G} = \frac{\pi^2}{4} \frac{\zeta^2}{1 - \zeta^2} F(\zeta, \delta_w), \quad (2.8)$$

where $\zeta = (k_F^\uparrow - k_F^\downarrow) / (k_F^\uparrow + k_F^\downarrow)$, and the function $F(\zeta, \delta_w)$ is given by

$$F(\zeta, \delta_w) = \frac{1}{2} \left[\frac{1}{\cosh^2(\pi k_F \delta_w)} + \frac{1}{\cosh^2(\pi k_F \zeta \delta_w)} \right]. \quad (2.9)$$

As expected, for values $k_F \delta_w \geq 1$, F becomes very small. It is interesting to note that for a narrow wall in a half-metallic ferromagnet ($\zeta \rightarrow 1$), the magnetoresistance goes to infinity. There is a reasonable comparison with the experimental data, given the uncertainties about the band structure in nanocontacts as compared to that in bulk.

As shown by Van Hoof *et al.* [78] for the ballistic transport case, the use of realistic band structures, as opposed to a free electron model, can give quite different results. Ab initio calculations [80] show that the (ballistic) domain wall magnetoresistance of Ni and Co, compared to that of a simple free electron model, is larger, and also shows a weaker dependence on wall thickness. For diffusive transport, it is shown that the effect of the additional wall scattering becomes weaker with increasing disorder. For both transport regimes it is found that the presence of a domain wall decreases the conductance.

Two papers [81][82] are concerned specifically with transport through walls in double exchange systems like the perovskites. It is found that the double exchange mechanism modifies the domain wall width, and that in bulk samples of manganese oxides a ballistic MR of 1-2% is expected. It is interesting to note that "the MR is bigger for small concentrations of electrons, where the Fermi energy is close to the band edge" [81].

2.3.3 Current-wall interaction

In recent years, there has been a great interest in the literature in the interplay between

a spin polarized current and the magnetic state of a ferromagnetic conductor. Several origins of the interaction between a current and a domain wall exist: momentum transfer, the field induced by a current (Oersted field); the 'hydromagnetic drag' force; and the spin transfer mechanism.

For high current densities the circumferential field induced by a current flow can be appreciable. In multilayer pillars a switching of the magnetization has indeed been observed and ascribed to this current-induced Oersted field [83][84]. In spin valves, the contribution of the current induced field has been demonstrated by Portier *et al.* [85].

The hydromagnetic drag force has first been studied theoretically by Berger [88], and later experimentally by [86][87]. It is associated with the Hall effect. Near a wall, where the internal field is reversed, charge carriers are deflected towards one end of the wall, due to the Hall effect. This non-uniform current distribution can be decomposed into a uniform current distribution and a eddy-loop current, and it is this eddy-loop current that produces a net force on the wall, which is always in the direction of the drift velocity of the carriers. The force per unit area of the wall is given by

$$F_x = 2J_s\mu_e^{-1}(R_1j - v_w), \quad (2.10)$$

where J_s is the saturation polarization, R_1 the anomalous Hall constant, j the current density, and v_w the wall velocity. The eddy current limited wall mobility μ_e , defined by the equation $v = \mu H$, has units m^2/C . For thin films, it is inversely proportional to the sample thickness, and the hydromagnetic drag force becomes negligible for films with a thickness $< 0.1 \mu\text{m}$.

The third mechanism, that of spin transfer ('*s-d* exchange drag'), is based on intra-atomic *s-d* exchange between spin polarized electrons and local moments, proposed by Berger [89]. The idea is that the injection of spin polarized electrons in the domain wall will lead

to a torque exerted by the wall on the polarization direction of the charge carriers. In turn, there is an s - d exchange interaction torque exerted by the polarized carriers on the wall, which may cause the wall to move. The expression for the total s - d exchange force per unit area of the wall is similar to that for the hydromagnetic drag force (2.10):

$$F_x = 2J_s\mu_i^{-1}(\beta_1 v_e - v_w), \quad (2.11)$$

with μ_i the intrinsic wall mobility, β_1 a parameter of order unity that describes the relation between change in carrier concentration and conductivity, and v_e the drift velocity. This force is significant for thin domain wall where the electrons cannot follow the local magnetization completely. It is expected to dominate over the hydromagnetic drag force for films with thickness $< 0.1 \mu\text{m}$. Experimental evidence of displacement of domain walls by this exchange drag force has been found in thin permalloy films [90][91][92].

The idea of the transfer of spin, or angular momentum, from a spin polarized current has since then been used as a basis for further theoretical and experimental studies. For magnetic multilayers, if the current density is large enough ($> 10^7 \text{ A cm}^{-2}$), the torque exerted on the magnetic moments will cause them to precess, leading to the emission of spin waves [93][94][95][96][97]. Because of the relatively high current densities needed, these spin waves have been studied experimentally through point contacts [98][99][100][101] and patterned devices [102]. Techniques used include electron spin resonance [98][101], Brillouin light scattering [99] and transport measurements [98][100].

An important effect predicted by Slonczewski [93], also based on the spin transfer idea, is the switching of the magnetic state of a ferromagnet by a (pulsed) current. This mechanism has been used to explain observed transport properties in manganite trilayer junctions [103], where it is proposed that a ferromagnetic cluster is switched by transfer

of angular momentum from the carrier spin to the conductor's moment. Current induced magnetization switching has also been observed in Ni nanowires [104][105] and Co/Cu/Co nanopillars [106].

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Chapter 3

Experimental Set-up

In this chapter a description is given of the experimental set-up, electronics and data acquisition software employed to obtain the data presented in this thesis. In the case of the Scanning Tunnelling Potentiometry and the Point Contact measurements, these had to be built and developed to make the required measurements possible, and are therefore presented in greater detail.

3.1 Sample Characterisation

3.1.1 X-ray Diffraction

X-ray diffraction was used to determine the structural characterization of our samples. This technique probes the crystal lattice with Cu K_α radiation (wavelength $\lambda = 1.5418\text{\AA}$). The X-rays are specularly reflected from the successive parallel planes of atoms in the crystal (see Figure 3.1). At certain angles the diffracted beams interfere constructively, leading to a peaks in the detected intensity. The position and intensity of these peaks is unique to each

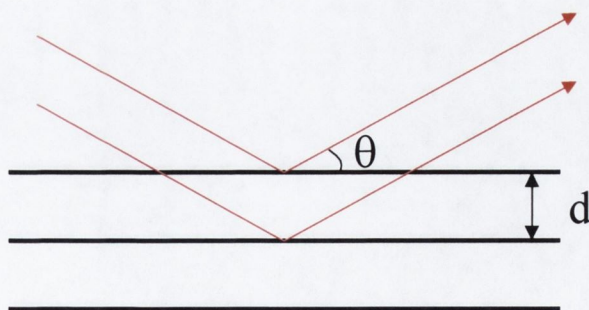


Figure 3.1. Principle of X-ray diffraction.

crystalline material. X-ray diffraction is based on Bragg's condition:

$$n\lambda = 2d \sin(\theta) \tag{3.1}$$

stating that a diffraction pattern is observed only when the path difference between the reflected waves (the product of twice the lattice spacing d and the sine of the diffraction angle θ) equals an integral number of X-ray wavelength λ . Spectra were recorded on a Siemens D500 diffractometer, with sample mounted on a glass substrate in the $\theta - 2\theta$ configuration.

3.1.2 Scanning Electron Microscopy

Scanning Electron Microscopy (SEM) is a powerful technique to determine the surface morphology and chemical constituents of a sample. Electrons generated by a tungsten filament, accelerated by a voltage of a few kV and focussed into a narrow beam by magnetic lenses onto the sample, will interact with the material. As a result of these interactions, secondary electrons, back-scattered electrons and X-rays are emitted. (See Figure 3.2). The

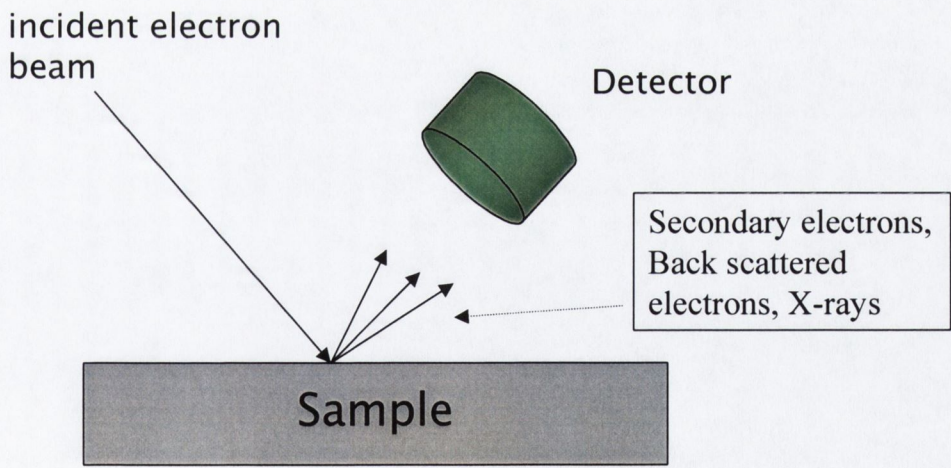


Figure 3.2. Principle of scanning electron microscopy.

low energy secondary electrons can be used to obtain an image of the surface morphology, while the high energy back-scattered electrons provide good atomic number contrast, mak-

ing it possible to detect different phases present in a sample. Heavier atoms produce more backscattered electrons and therefore lead to brighter areas in the image, whereas lighter atoms lead to darker areas. The SEM is used is the Hitachi S-3500N which also allows for Energy Dispersive X-ray (EDX) analysis. Using EDX, the produced X-rays can reveal information about the elements present, and can therefore be used to determine the chemical composition of the sample and the eventual presence of impurities.

3.1.3 Atomic Force Microscopy and Magnetic Force Microscopy

The surface morphology of our samples was characterised using an atomic force microscope (AFM). See Figure 3.3. In this technique an atomically sharp tip at the end of a cantilever is brought into contact with the sample. By shining a laser at the far end of the cantilever, and detecting the reflected beam with a four quadrant photo detector, the deflection of the cantilever is measured. When scanning the tip over the sample, the cantilever will be deflected due the hills and valleys of the sample. The output of the photo detector is fed to a controller which in its turn moves the sample up or down, to maintain a constant deflection of the cantilever. The amount which the sample has to move is then a measure of the local sample height. High precision sample movement in x, y and z direction is made possible using a four quadrant piezo tube. The AFM used is Digital Instruments MultiMode AFM with a Nanoscope III controller, which also allows for obtaining topographic information using Tapping modeTM. In this mode the cantilever is oscillating at its resonant frequency (usually a few hundred kHz), by acoustically pumping it with a piezo crystal which is located just underneath the cantilever holder. As the cantilever approaches the surface of the sample, the amplitude of the oscillation will change, and this change is monitored by the photo detector. Again, the controller will adjust the sample position to

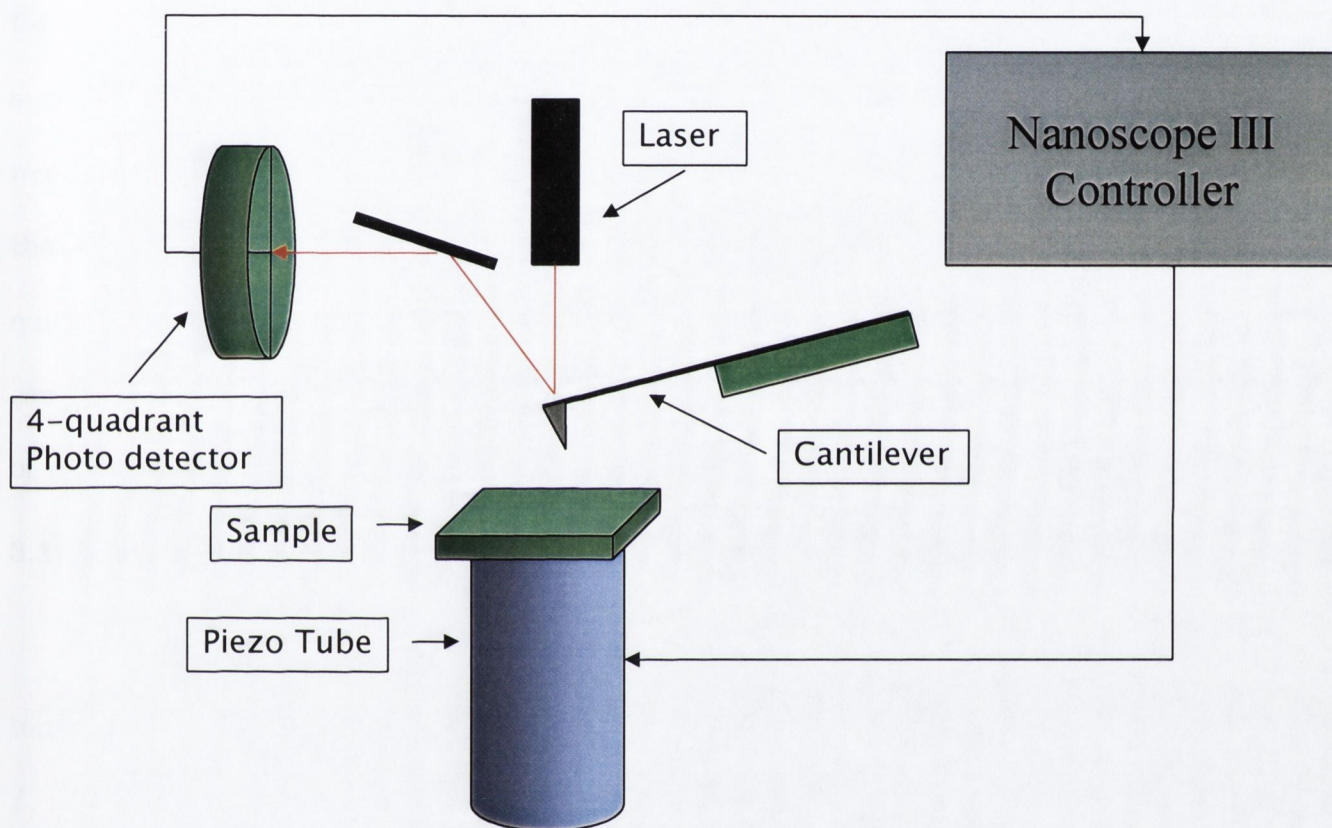


Figure 3.3. Schematic of the AFM set-up. The deflection of the cantilever while scanning over the sample is measured using a photo detector. The Nanoscope III controller then moves the sample up or down to compensate for this deflection.

maintain a constant amplitude as the cantilever is scanned over the sample. Tapping mode has the advantage over contact mode in that there are no frictional forces present.

Magnetic Force Microscopy (MFM) is used to obtain information about the domain structure of ferromagnetic materials. This technique employs LiftmodeTM, where a tip coated with magnetic material is first scanned over the surface to obtain the surface topography in tapping mode. The tip is then raised a few tens of nanometres above the surface, and the surface topography is scanned while monitoring for the influence of magnetic forces. The resonant frequency f_0 of the cantilever is shifted by an amount Δf which is proportional to the vertical force gradients in the magnetic forces acting on the tip. These shifts are usually quite small, in the order of a few Hz, compared to a resonant frequency $f_0 \sim 100$ kHz. They can be detected by monitoring the change in amplitude as the resonant frequency is shifted, or by directly measuring the shift in frequency.

3.1.4 Vibrating Sample Magnetometry

The magnetization M of a sample can be determined using Vibrating Sample Magnetometry (VSM). In this technique, the sample, mounted on a rod, vibrating at a frequency f , is placed in between two pick-up coils (see figure 3.4). The displacement of the sample gives rise to a change in magnetic flux Φ over time. This induces a voltage V in the coils:

$$V = C \frac{\partial \Phi}{\partial t} \tag{3.2}$$

where C is a constant. Integrating the voltage over time gives the total change in flux which is proportional to the magnetization. A hysteresis curve is obtained when doing a series of magnetization measurements as function of applied external field. In our case, a field of up to 1.1 T is created by two Hallbach cylinders [1].

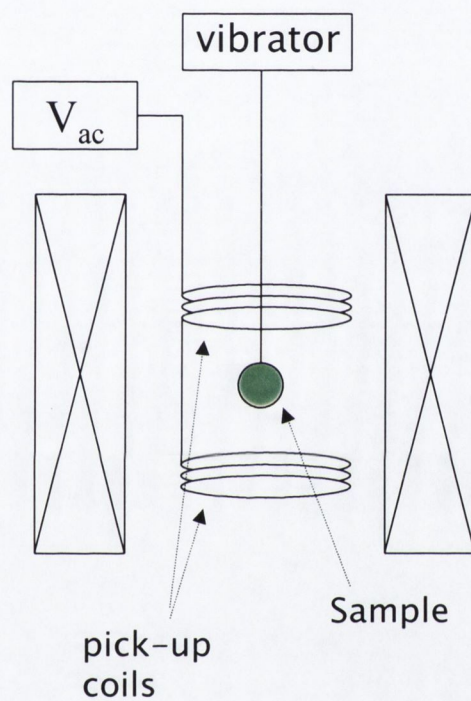


Figure 3.4. Schematic of the vibrating sample magnetometry set-up. A vibrating ferro-magnetic sample creates a change in flux over time, which results in a voltage differential in the coils.

3.1.5 SQUID

A SQUID (Superconducting QUantum Interference Device) is, like the VSM, used to measure the magnetization M of a sample. It has the advantage that it is much more sensitive than an VSM, so that much smaller samples can be measured. A set of superconducting detection coils is connected to the SQUID with superconducting wires, allowing the current caused by a moving sample inside the coils to inductively couple to the SQUID sensor. The SQUID itself consists of a superconducting loop interrupted by a so-called Josephson junction. Quantum interference effects in the SQUID make the device extremely sensitive to small changes in magnetic field. In a fully calibrated system, the variations in output voltage of the SQUID sensor are directly proportional to the sample's magnetic moment.

The SQUID measurement system used here is a Quantum Design MPMS, with capability of measuring the magnetization of samples in fields of up to 5 T, and in a temperature range of 2 – 400 K, or up to 800 K when using the oven.

3.1.6 Transport Measurements

The resistance of a sample as a function of temperature could be measured using a standard 4-point geometry in a APD closed-cycle cryostat, down to 10 K. The sample is mounted on a copper block using a heat sink paste, and the four contacts are made using silver wire with Agar Scientific quick drying silver paint.

3.2 Scanning Tunnelling Potentiometry

To be able to perform Scanning Tunnelling Potentiometry (STP) we started with an existing STM, PC and control electronics for scanning and feedback. We had to modify the sample holder, add additional electronics, and write the data acquisition software.

3.2.1 Set-up

The scanning tunnelling microscope used was a Burleigh ARIS 3500 Benchtop STM, with some additional electronics to apply a bias voltage over the sample. All the experiments are carried out in ambient air at room temperature. Our scanning tunnelling potentiometry setup (see Fig. 3.5) is based on that first used by Kirtley, Washburn, and Brady [2]. It allows for simultaneous potential and topographic measurements at each point of the scan range. In the following we will refer to the tip-sample voltage by V_{bias} , the tip-sample separation d , the tunnelling current will be designated by I_t . We recall that the tunnelling current is proportional to the bias voltage V_{bias} but depends exponentially on the tip-sample distance d : $I_t \propto V_{bias} \exp(-\alpha d)$. An STM can be used in two ways: in a "constant current mode"

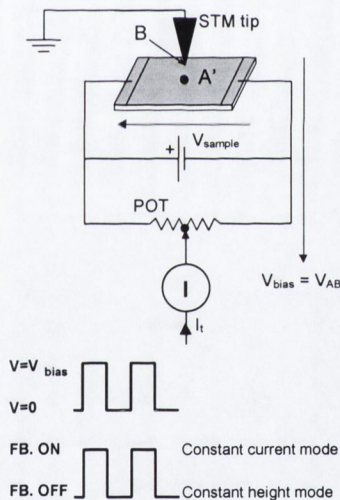


Figure 3.5. Schematic drawing of the STP set-up. Adjustment of the trimpot POT will ensure that the potential of point A' will be equal to that of A, so that a macroscopic V_{sample} only introduces a small perturbation to V_{bias} .

in which the tunnelling current is kept constant by letting the tip follow the contours of the sample surface. This is done by adjusting the voltage applied to a piezo crystal, to keep the tip-sample distance d constant. This voltage is then directly proportional to the surface

topography. The other way of operating an STM is in a "constant height mode", in which the tip is maintained at a constant height and the tunnelling current is recorded during the scan over the surface. The topography of the sample can be deduced since the tip-sample height varies logarithmically with the tunnelling current. For a scanning tunnelling potentiometry recording, we have to apply a voltage V_{sample} across the sample to make a current flow through the sample (see figure 3.5). It is then possible to adjust a potentiometer POT so that below the tip scanning area the potential $V_{AA'}$ is equal to zero, the voltage V_{sample} thus introducing little perturbation in the tip-sample bias potential V_{AB} . In this way it is possible to apply a large voltage over the sample, while being able to control the sample-tip potential V_{AB} to within a few hundreds of microvolts. The actual tip-sample bias varies slightly as the tip of the STM is scanned over the surface, because of the applied potential V_s over the sample: $V_{bias}(x, y) = V_{bias} + V_s(x, y)$. This leads to a change of the tunnelling resistance as a function of the position. The subsequent error in the measured surface height, however, is negligible because the tip height above the sample depends only logarithmically on the tunnelling resistance [2]. For a typical scanning area of $250 \times 250 \text{ nm}^2$ and a typical bias voltage of 30 mV, a 20 V/cm field applied on the sample only introduces variations of less than 2% in the bias voltage.

During a potentiometric scan one first senses the topography, at each position over the scanning area, via a constant current measurement. The bias voltage V_{bias} is set to a value of a few tens of mV (typically 30 mV) and the feedback loop is closed. The microscope adjusts the tip height relative to the sample so that the tunnelling current I_t is set equal to the "set point" I_{t0} . The feedback loop is then opened and the bias voltage set to 0. The residual voltage between the sample and the tip is then equal to $V_s(x, y)$ but the tip-sample distance

is unchanged. The measured tunnelling current I is thus proportional to $V_s(x, y) \exp(-\alpha d)$. Since the tip-sample distance is kept constant and equal to d , the tunnelling current map is proportional to the potential $V_s(x, y)$. The proportionality factor is the tunnelling resistance (usually ranging from 1 to 10 M Ω) and which depends on the distance d . The typical measurement conditions are $V_{bias} = 30$ mV, $I_t = 3$ nA, giving a tunnel resistance of 10 M Ω . Since the noise in the measured potential is dominated by fluctuations in the tunnelling resistance, higher values of V_{bias} tend to give poor signal to noise ratios. The typical potential resolution we obtain in our set-up is of order 10 μ V.

The Burleigh STM consist of three parts: the STM itself, the feedback electronics with the controls for magnification, scan range, reference current, bias voltage and feedback settings, and finally a PC with the data acquisition software. The PC controls the control electronics via an ADA3100 ADC card. A TTL signal is used to enable or disable the feedback loop. In the home-built additional electronics (See Fig 3.6), this TTL signal is

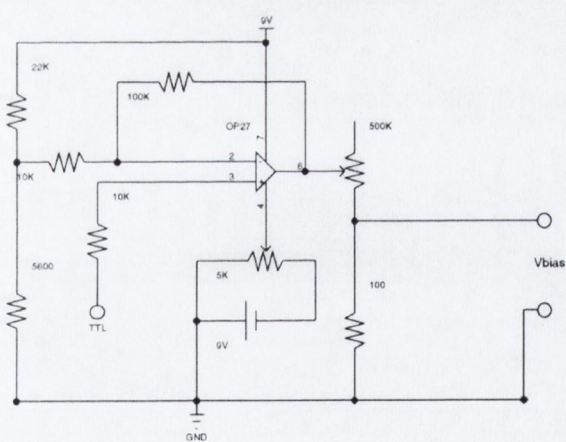


Figure 3.6. Schematic drawing of the electronics that allows bias voltage V_{bias} to be changed simultaneously with the opening and closing of the feedback loop (TTL signal).

also used to switch the bias voltage: a closed feedback loop will supply a few tens of mV (depending on the 500K Ω trimpot) to the V_{bias} output, while an open feedback loop gives a

0V output. The 5K Ω trimpot, together with the 9V battery, is used to compensate for any zero-voltage offset. This V_{bias} output is connected to the "external bias voltage input" on the control electronics.

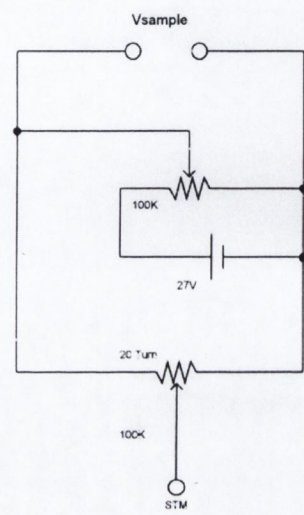


Figure 3.7. Schematic drawing of the electronics used for applying a macroscopic voltage over the sample while keeping the tip-sample voltage minimal.

For normal STM operation, the tip is grounded, while the sample, which is connected to the sample holder with silver paint, is biased. The sample holder therefore had to be modified to be able to perform STP, see Fig. 3.7. The batteries supply macroscopic potential difference V_{sample} over the sample, while the 20 turn trimpot is used to ensure that this only introduces a small pertubation on the tip-sample bias voltage which is supplied by the sample holder (labelled STM in Figure 3.7).

3.2.2 Software

The data acquisition software was written using Borland's Delphi 3 development tool, which uses the Pascal language. The program starts up with, after initializing the ADC card and the control electronics, with the main window, where the user can select a variety of parameters controlling the image acquisition, such as a log or linear preamplifier, gain, several

delay times and number of datapoints and substeps between those datapoints. Once these have been set, the program is ready to start acquiring both topographic and potentiometric data. With the feedback loop closed, and a tip-sample bias voltage applied, the piezo voltage output is recorded as a measure for surface height. The feedback loop is then opened, the tip-sample bias voltage is brought back to zero, and the tunnel current is recorded as a measure for the local potential. (It is also possible to average both the piezo voltage output as the tunnel current over several measurements). The feedback loop is then closed again, and the tip is moved to the next point, where the cycle repeats itself. During the scan, the user can observe both the piezo voltage and tunnel current signals, and make the necessary adjustments to the feedback settings. After completing one scanline, both the topographic data and the potentiometric data are plotted before continuing on the next scanline. After finishing the measurement the user has the option to save one or both datasets in the Burleigh IMG format. This format makes it possible to use the software developed by Burleigh for normal STM operation, for image processing.

3.3 Point Contact Measurements

To study of transport properties of nanocontacts one needs contacts that are stable for a sufficiently long time to perform the required measurements. Here we describe the set-up used to investigate the magnetotransport behaviour of point contacts at room temperature and 1-N₂ temperatures. Point contact phenomena like quantum conductance are most widely studied in nanocontacts formed when two macroscopic electrodes are put into contact and then slowly pulled apart. Ideally, one would like to have precise control over the movements of the electrodes, to obtain a stable nanocontact.

3.3.1 Room Temperature measurements

3.3.1.1 Set-up

In our set-up (see Figure 3.8), the nanocontacts are made by putting two $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ or Fe_3O_4 crystallites into contact and then slowly pulling them apart again. One crystal is mounted on a block of aluminium, while the other is attached to the tip of a P-820.30 low voltage piezo translator. This Physik Instrumente translator is made of stacks of piezo crystals and has a travel length of $45\mu\text{m}$ at room temperature. The voltage needed to fully extend the piezo crystals is supplied by a Physik Instrumente E-861.10 Single Channel Amplifier. It accepts up to 10V at the input and amplifies it to up to 100V for the full travel length of the translator.

The translator itself is mounted on a precision ball slide which can be moved by micrometer screw. This assembly is used for the rough approach of the two crystals. The actual making and breaking of the contacts is done by the piezo translator. A voltage V_{bias} , of the order of a few tens of mV, is applied over the two crystals (see Fig 3.9):

$$V_{bias} = \frac{R_1}{R_0 + R_1} V_{source}, \quad (3.3)$$

where V_{source} is the voltage supplied by the variable voltage source. The current flowing through the nanocontact is recorded by an I-V converter. The operational amplifier used is an OP37 low noise operational amplifier, suitable for applications requiring high speed at great gain. Its output, V_{output} , is given by:

$$V_{output} = \frac{-R_2}{R_{contact}} V_{bias}, \quad (3.4)$$

where $R_{contact}$ is the resistance of the nanocontact. The conductance of the nanocontact, in

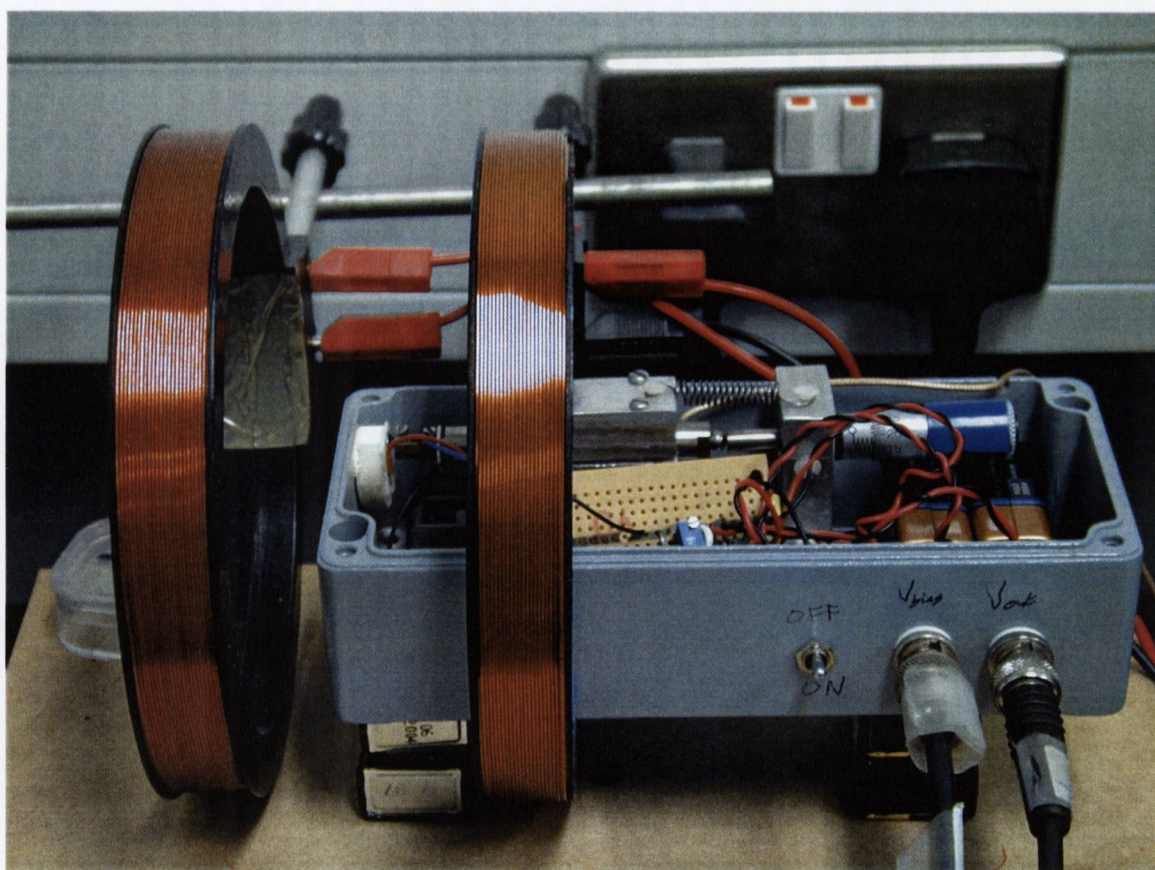
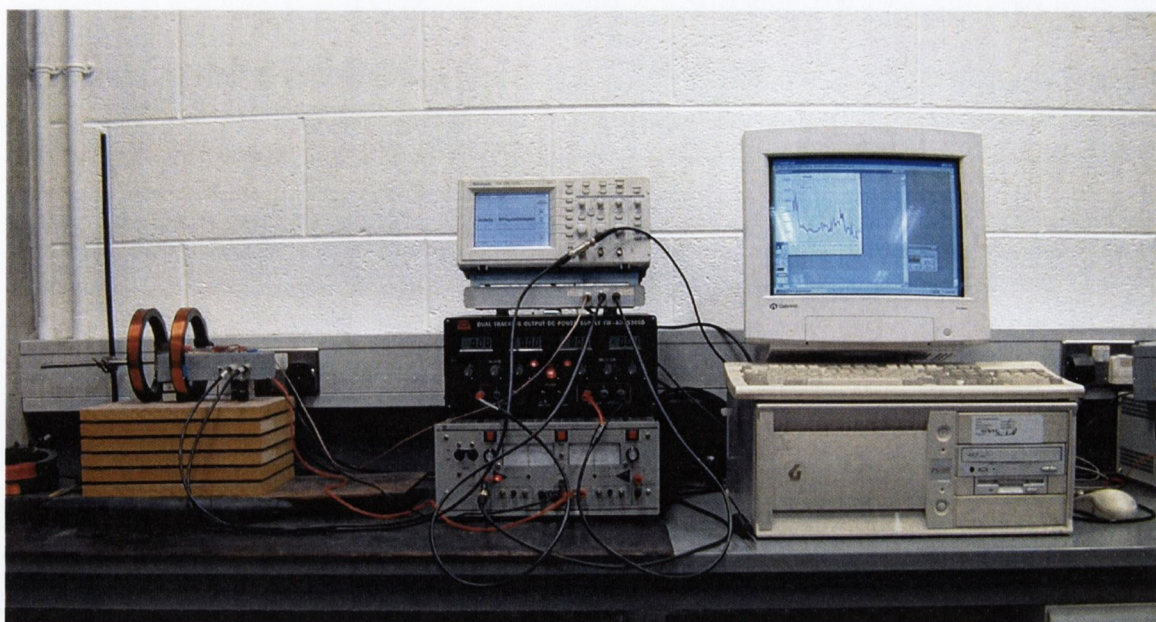


Figure 3.8. Photo of the tabletop set-up. a) General overview of set-up, b) Close-up of nanocontact set-up with piezo translator, micrometer and coils.

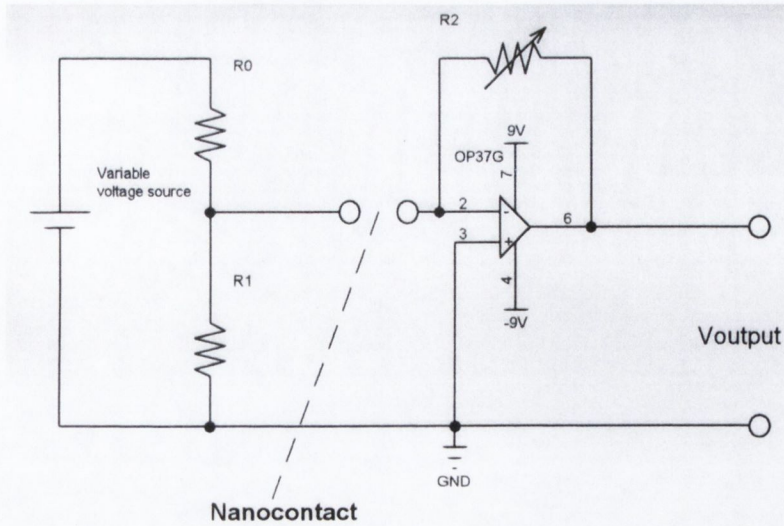


Figure 3.9. Schematic drawing of the electronics used to measure the conductance of a nanocontact. The voltage source supplies a voltage over the contact, while the current through the contact is converted to a voltage using a OP37 op-amp.

units of the quantum conductance G_0 , can then easily be deduced:

$$G = \frac{1}{R_{\text{contact}} G_0} = \frac{V_{\text{output}}}{R_2 V_{\text{bias}} G_0}. \quad (3.5)$$

The I-V convertor, and the nanocontact assembly are mounted in a die-cast box to shield from noise. For vibration insulation, the whole set-up is mounted on a stack of plywood plates separated by Viton O-rings, which are placed on heavy metal plate on top of an inflated tube.

The variable voltage source can be a battery, a power supply or the output from one of the DAC's from the ADA3100 data acquisition board, controlled by a PC, depending on the measurement required. The output from the I-V convertor can either be viewed (and stored) on a oscilloscope (Tektronix TDS220 digital storage oscilloscope), or fed directly into a PC using the ADA3100 card. The trace captured by the scope can be read into the computer using the RS232 serial port for further processing.

To study the transport behaviour of the nanocontacts in a small magnetic field, two

coils are placed around the die-cast box. The coils are driven by a home-built amplifier which has a -5V to +5V input and a -30V to +30V output which produces a field of up to 5mT. The field can be controlled by the ADA3100 card.

3.3.1.2 Software

All the data acquisition software that was developed for the quantum conductance experiments, was written in QuickBasic. Each experiment has its own program: measuring the conductance while slowly breaking the contact, measuring the current-voltage characteristics of a stable nanocontact, and the magnetoresistance of a stable nanocontact.

For measuring the conductance of a breaking nanocontact, the program first initializes the ADA3100 card and communications port. The scope is also initialized, setting it to single shot mode, and its trigger level to a few multiples of G_0 . The user has to manually bring the two LSMO crystal into contact, using the micrometer screw, after which the program will make and break the contacts automatically using the piezo translator. After a contact has been established, the program reduces the voltage at the piezo amplifier input through one of the ADA3100 analogue output ports, thereby retracting the translator, and checking the scope until it has been triggered. The scope stores the trace in its internal memory, which can be accessed through the serial port. The data is plotted on the screen and automatically saved as a file. The two crystals are then put into contact again, and the next trace is obtained and saved. In this way hundreds of breaking curves can be obtained automatically, which can then be analysed in the form of a histogram.

For measuring I-V curves of a stable nanowire, one of the analogue output ports of the ADA3100 has to be connected to the V_{source} input, and the V_{output} output has to be connected to one of the analogue input of the ADA3100. As before, after initializing, the

contact is made, and the translator is slowly retracted. The value of the conductance of the nanocontact is constantly measured (at 30mV), and when it reaches a certain level, the translator is stopped. The voltage over the nanocontact, V_{bias} , is then swept (via V_{source} and the voltage divider R_0 and R_1 , see figure 3.9) from about -0.8V to +0.8V and back, at a preset speed. V_{output} is monitored, plotted on screen as a function of V_{bias} and stored in a file. After thus obtaining an I-V curve, the V_{bias} is set again at 30mV, a new contact is made, and another I-V curve obtained.

The program for measuring the magnetoresistance of a nanocontact is much the same as that for obtaining I-V curves. The V_{source} input is now connected to a battery, giving a 30mV voltage difference over the nanocontact, while one of the analogue output of the ADA3100 is now connected to the input coils amplifier. Again, the contact is made and the translator slowly retracted. When the conductance of the nanocontact reaches the desired level, the magnetic field is swept in a certain pattern. This can be a square, a sine or a triangular waveform. The program plots V_{output} and the voltage output to the coil amplifier on the screen and saves the data in a file.

3.3.2 Low Temperature Measurements

3.3.2.1 Set up

Point contact measurement at low temperatures were performed in a custom made Oxford Instruments cryostat, based on an OptiStat model. See Figure 3.10. The continuous flow cryostat has a long tail which can be fitted inside the bore of a Multimag variable field permanent magnet system, capable of fields up to 1 Tesla, or placed inside a set of coils for fields up to 14 mT. A Cernox temperature sensor and heating element placed just after the needle valve between cryogenic reservoir and sample space controls the temperature in the

cold region of the cryostat, which is situated inside the tail. The sample rod is loaded into the cryostat from the top, with the KF flange sealing the sample space (see schematic drawing of the sample rod, figure 3.11). The two crystals making the nanocontact are mounted in copper block which contains another Cernox temperature sensor and heating element. A Mitutoyo 153-202 micrometer (precision $1\mu\text{m}$) at the top of the sample rod is used for a rough approach of the two crystals, while a piezo tube (EBL#2, Staveley Sensors, gold coated) controls the contact more precisely. Total travelling length at room temperature is $2\mu\text{m}$, but this is reduced dramatically at low temperatures, with only 20 % of the room temperature travel length remaining at 4 K [3]. A Physik Instrumente E-461.00 HVPZT Amplifier, which can be either controlled manually or via a computer, supplies the high voltages (up to 300 V) needed for the piezo tube.

The sample rod was wired up using copper wire and EPOTEK E4110 conductive epoxy, and silver wire with Agar Scientific quick drying silver paint. The sample holders and piezo tube were mounted using EPOTEK H77 nonconductive epoxy.

To measure the transport properties of the nanocontacts, a similar set-up to the room temperature one was used (See section 3.3.1). An I-V convertor outputs a voltage proportional to the resistance of the contact. This signal is fed into an oscilloscope and an ADC card. The software running the experiment also controls the movement of the piezo, the bias voltage applied to the contacts and field applied.

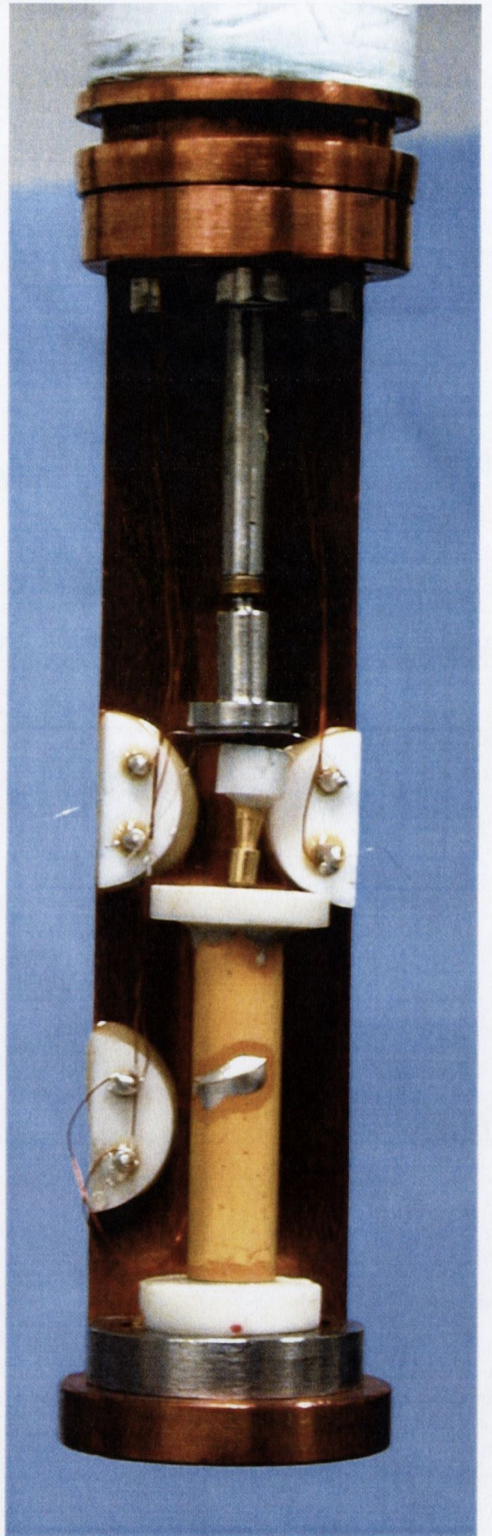
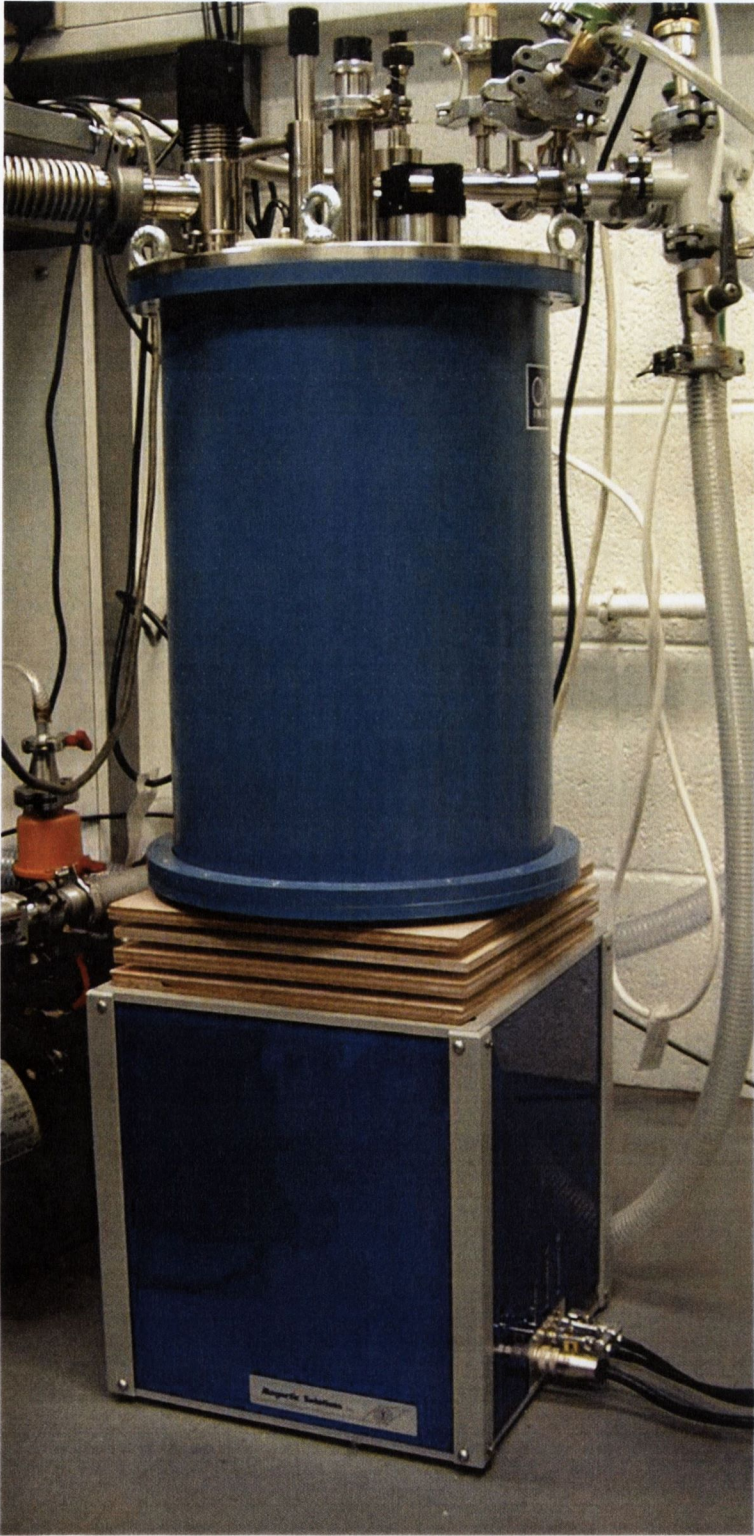


Figure 3.10. Photo of the low temperature point contact measurement set-up. a) Cryostat with its tail sitting inside a Multimag variable field source. b) Close up of the sample rod with piezo tube.

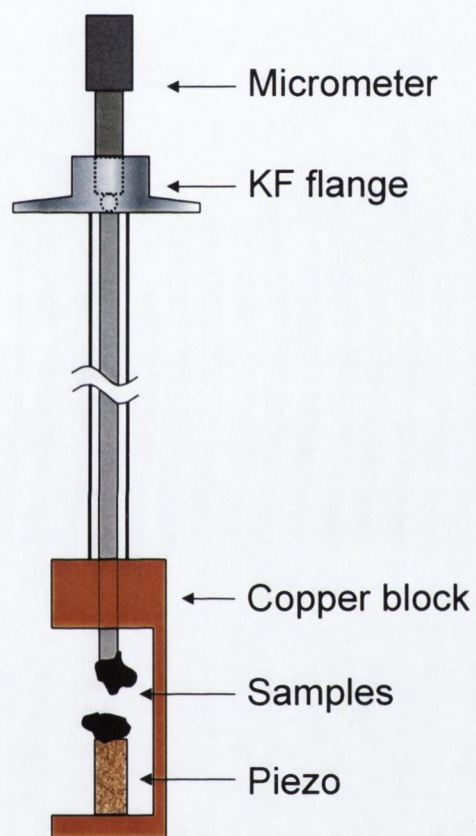


Figure 3.11. Schematic drawing of the sample rod used for low temperature nanocontact measurements.

References

- [1] O. Cugat, R. Byrne, J. McCauley and J.M.D. Coey, *Rev. Sci. Instrum.* **65**, 3570 (1994).
- [2] J. R. Kirtley, S. Washburn, and M. J. Brady, *Phys. Rev. Lett.* **60**, 1546 (1988).
- [3] Physik Instrumente, <http://www.physikinstrumente.com>.

Chapter 4

Results

4.1 Scanning Tunnelling Potentiometry

Polycrystalline $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films were deposited, on polycrystalline MgO substrates, using pulsed laser ablation (KrF laser, wavelength $\lambda=248$ nm). The films, with a thickness of about 75 nm, were grown in a 0.3 mbar oxygen atmosphere, with a substrate temperature of 680 °C, and cooled down in a 1 bar oxygen atmosphere. To prepare the substrates, MgO powder was pressed into pellets, and sintered at 1400 °C for 20 hours. The average grain size thus obtained was about 1 μm . The grain morphology of the LSMO film resembles that of the MgO substrate. The temperature dependence of the electrical resistivity of these polycrystalline films shows a broad maximum at about 150 K (Figure 4.1). Epitaxial

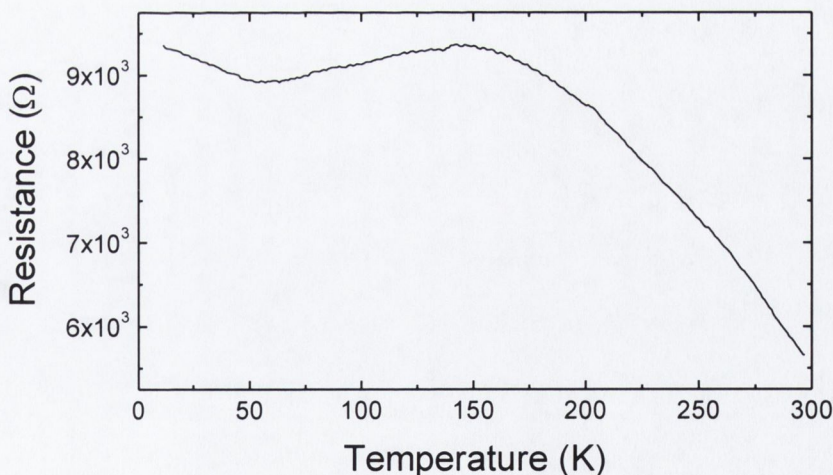


Figure 4.1. Resistance of a polycrystalline thin $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ film as a function of temperature.

$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films (thickness ca. 75 nm) deposited under the same conditions on (001)

oriented single crystal MgO substrates show typical metallic behaviour with a residual resistivity of $\rho_0=100\ \mu\Omega\text{cm}$.

Figure 4.2a shows a topographic image of an area of 250 nm by 250 nm of one of our polycrystalline $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films, while Figure 4.2b shows the simultaneously acquired potentiometric image of that same area, using the technique described above. An electric field of 20 V/cm was applied over the sample, in the horizontal direction, in such a way that the potential, on average, decreased from right to left. This would imply a drop of 500 mV over the 250 nm width of the scan. In contrast to this uniform drop, the potentiometric image shows essentially constant potential regions, separated by sharp steps. The step separating the region of nearly constant potential in the bottom left corner shows that the drop in potential is not monotonic; it actually shows an increase in potential, in contrast to the potential gradient, which is negative on average.

In pairs of images like Figure 4.2, it is generally possible to correlate the steps in the potentiometric image with distinct features on the surface, which we identify as grain boundaries.

Sometimes we see similar surface features that show no steps in the potentiometric image, but that can be explained if a metallic contact exists between those grains. On the other hand, steps are sometimes observed that seem to have no direct surface feature associated with them. It is possible that this is caused by grain boundaries that lie beneath the surface.

When no electric field was applied, the potentiometric images were invariably flat and featureless, see Figure 4.3. This, together with the fact that steps in the potential usually coincide with distinct topographical features leads us to believe that we are indeed measuring

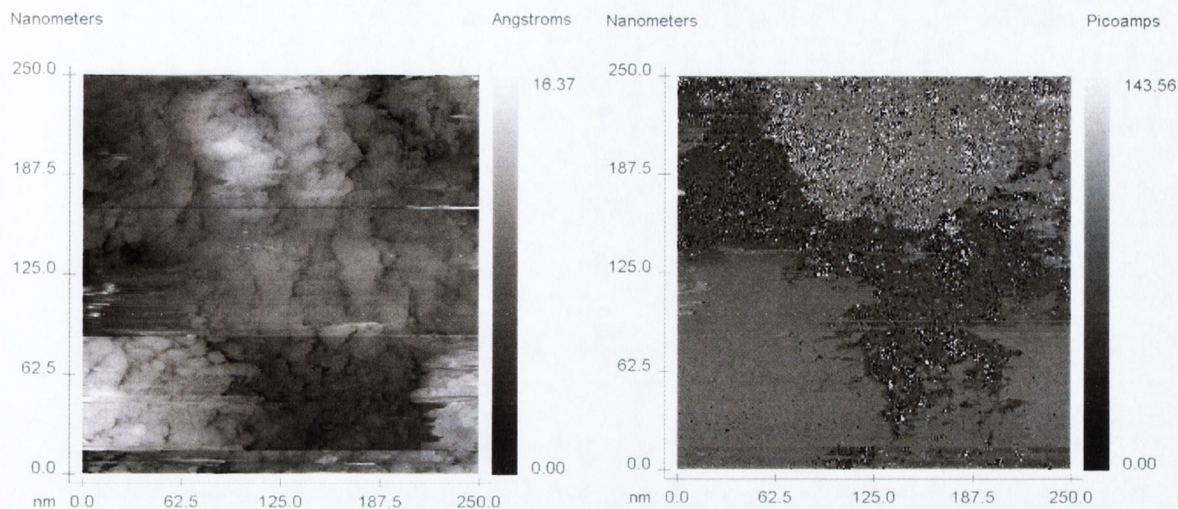


Figure 4.2. (a) Topographic and (b) potentiometric picture measured on a polycrystalline $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ thin film (75 nm thick) deposited on a MgO substrate. The images were obtained with a tunnel resistance of $10\text{ M}\Omega$.

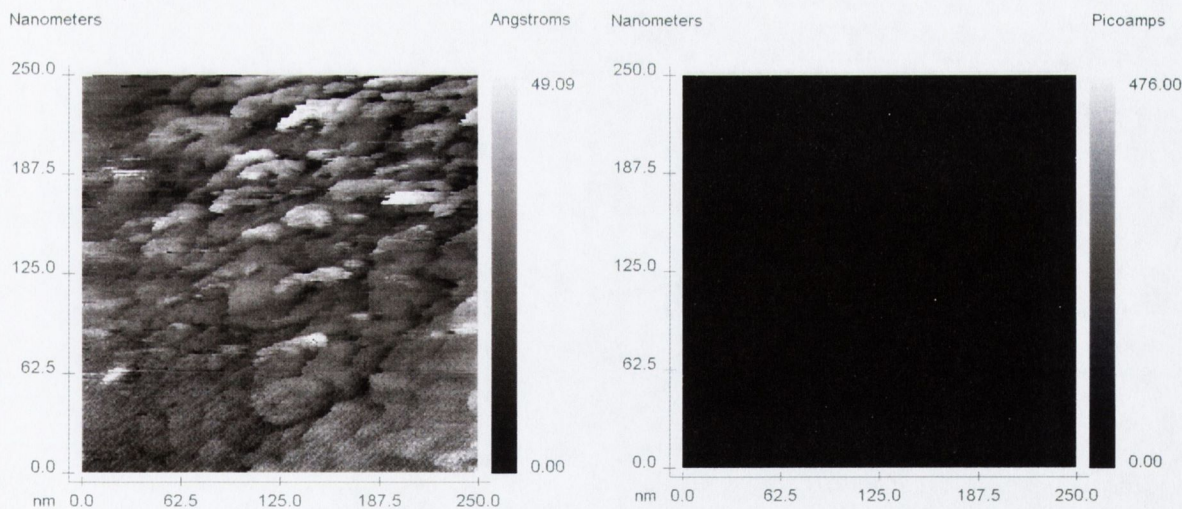


Figure 4.3. (a) Topographic and (b) potentiometric picture measured on a polycrystalline $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ thin film (75 nm thick) deposited on a MgO substrate. No bias voltage has been applied over the sample. The images were obtained with a tunnel resistance of $10\text{ M}\Omega$.

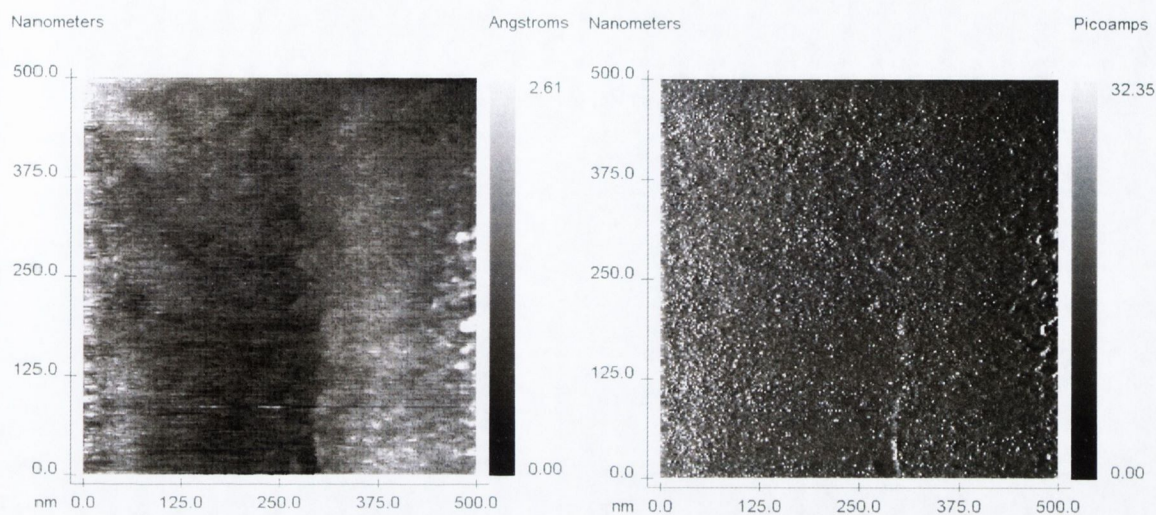


Figure 4.4. (a) Topographic and (b) potentiometric picture measured on an epitaxial $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ thin film (75 nm thick) deposited on a MgO substrate. The images were obtained with a tunnel resistance of $10\text{ M}\Omega$.

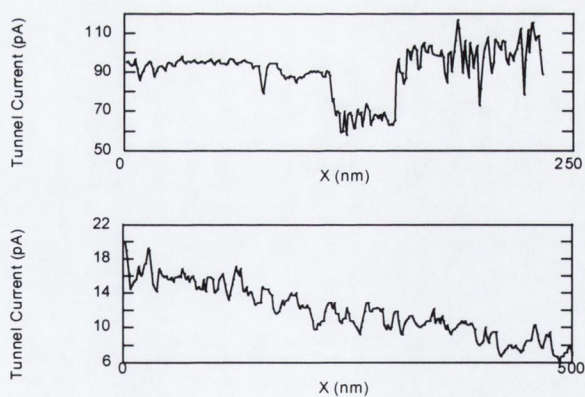


Figure 4.5. Potential cross sections. (a) on a polycrystalline thin film; (b) on a single crystalline thin film. Both were obtained with a tunnel resistance of $10\text{ M}\Omega$. The biggest step in the polycrystalline case corresponds to a voltage drop of $440\text{ }\mu\text{V}$.

the local surface potential.

Figure 4.4 shows the topographic and potentiometric images of an $500 \times 500 \text{ nm}^2$ area of an epitaxial $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ film, measured in the same conditions. No steps can be distinguished in the potentiometric image; only a smooth potential gradient is visible.

A representative cross section of the potentiometric images of both a polycrystalline and an epitaxial films are shown in Figure 4.5. No steps can be seen in the epitaxial film. Over the whole cross section the potential drops rather smoothly by about $120 \text{ } \mu\text{V}$. This constant potential gradient signifies that the resistance arises in the bulk of the film, rather than at the grain boundaries.

In the polycrystalline case, three distinct region can be noticed, separated by sharp changes in the potential. The biggest step corresponds to a 440 mV drop in the potential. A wide range in step heights is found, from the lower limit set by the resolution of our experimental set-up of about $10 \text{ } \mu\text{V}$ to steps as large as 1 mV . From the range in step heights observed experimentally we calculate grain boundary resistivities ranging from $r_{GB} = 3 \times 10^{-7} \text{ } \Omega \text{ cm}^2$ to $3 \times 10^{-5} \text{ } \Omega \text{ cm}^2$, with an average value of $r_{GB,ave} = 6 \times 10^{-6} \text{ } \Omega \text{ cm}^2$. If this is uniform, the resistance of a contact of area 60 nm^2 would therefore be $10 \text{ M}\Omega$.

4.2 Point contacts

4.2.1 $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$

4.2.1.1 Sample characterization

The $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystals (ca. $5 \times 3 \times 2 \text{ mm}^3$) used in this experiment were made using an infrared image furnace [1]. The topography and magnetic domain structure were studied using magnetic force microscopy (MFM), see figure 4.6. The topography shows

some preferential directions which seem to be reflected in the domain pattern. Bubble to stripe-like patterns with domain width of around a micron are observed. Magnetization as a function of applied field was investigated at room temperature, 150 K and 77 K, using a SQUID, see figure 4.7. At 300 K there is still a high field slope present in the magnetization, whereas for temperatures $T \leq 150$ K, the LSMO crystal's magnetization is saturated in a field of 1 T. The inset shows that the coercivity is around 1 mT for all three temperatures measured.

Resistance as a function of temperature is shown in Figure 4.8. As expected for this material, the resistance shows a metallic behaviour below the Curie temperature ($T_C = 380$ K). A sharp drop in the high temperature region is gradually replaced by a smaller decrease in resistance for lower temperatures. The low temperature resistivity is approximately $1.5 \times 10^{-4} \Omega \text{ cm}$.

4.2.1.2 Quantum Conductance

Figure 4.9(a) shows a typical variation of the conductance when breaking the contact between the two crystals using a relay. Steps at values close to multiples of G_0 are clearly visible. It is noteworthy that the time scale during which the contact between the crystals breaks is much longer than for metals. To explain this, Ott *et al.* [2] have suggested that the breaking mechanism for a ceramic like $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ is similar to brittle fracture whereas for ductile metals the break is accompanied by plastic deformation [3]. Figure 4.9(b) shows a conductance histogram obtained by accumulating 60 contact breaks between two $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystals. There are well defined peaks, again at values close to multiples of the conductance quantum. Peaks up to the seventh order can clearly be distinguished [2]. A closer inspection of Figure 4.9(a) and (b) shows that the steps are not

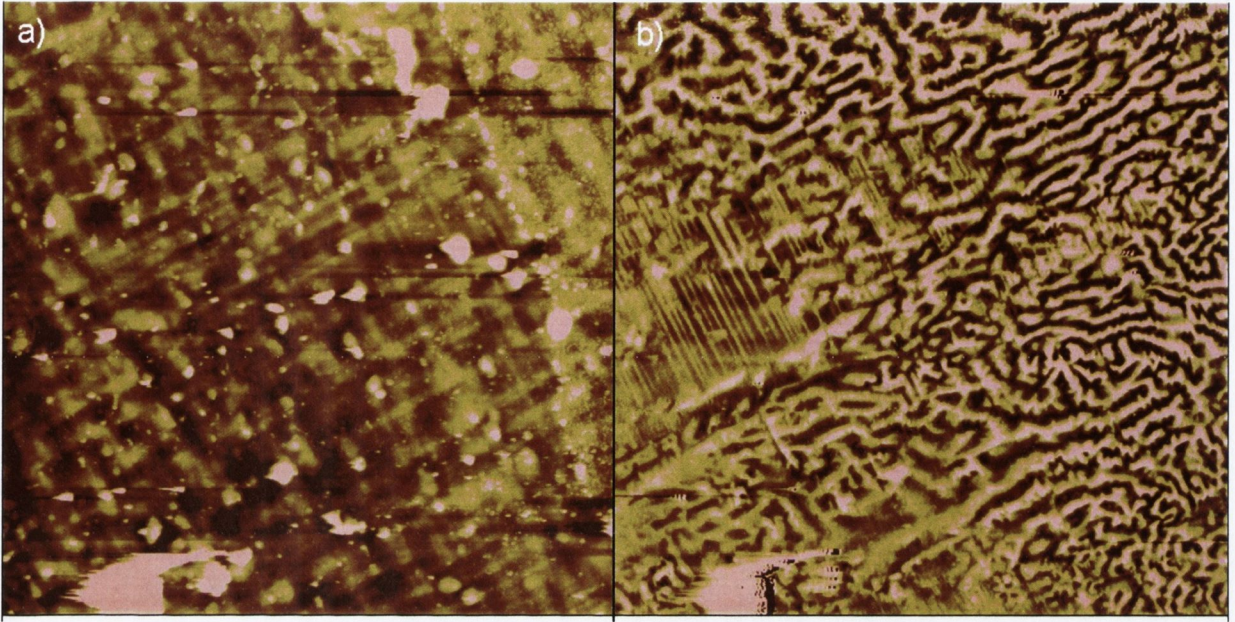


Figure 4.6. a) Topography of a $35 \times 35 (\mu\text{m})^2$ area of $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystal obtained by AFM. Z-scale is 200 nm. b) MFM picture of the same area.

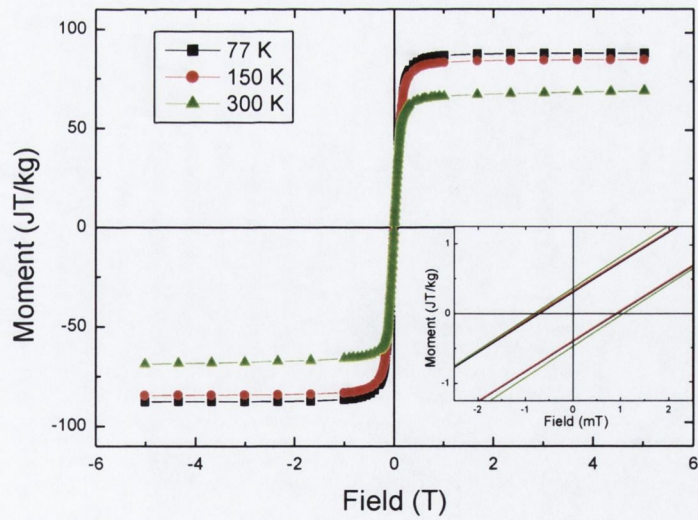


Figure 4.7. Magnetization of a LSMO single crystal as a function of applied field for three different temperatures: 300 K, 150 K and 77K, respectively. The inset shows a close-up of the region around the coercive field.

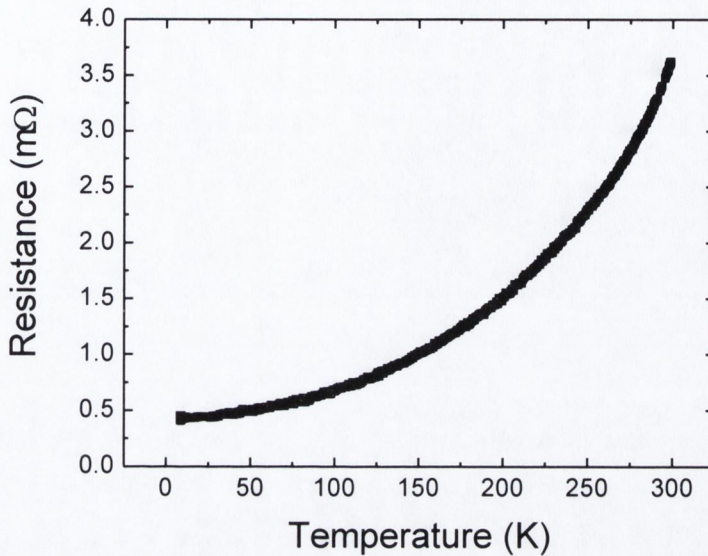


Figure 4.8. Resistance of an $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystal as function of temperature.

located at integer values of G_0 .

The use of a relay offers little control over the speed with which the contact between the two crystals breaks, and makes it impossible to stabilize the conductance of the contact at a few G_0 . We have therefore repeated the above experiment using a piezo crystal controlled by a computer to make and break the contact. With the piezo crystal fully extended, the two crystals are brought into contact. The voltage over the piezo crystal is then decreased, causing the two crystals to be pulled apart, and the current is recorded as described above. Figure 4.10 shows a typical variation of the conductance when breaking the contact between two $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystals using a piezo crystal. Twenty or more steps are visible, with the steps reflecting the digitized output of the ADC card that controls the piezo movement. The conductance of the contact in between steps often takes on values close to nG_0 .

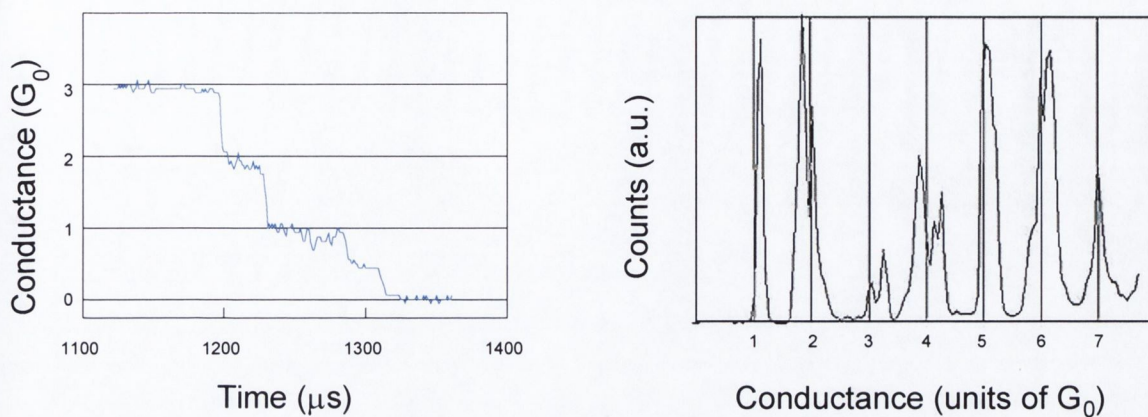


Figure 4.9. (a) Quantized conductance during the breaking of a contact between $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystals using a relay. (b) Conductance histogram accumulated from 60 breaking contacts of $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystals using a relay. After Ott et al. [2].

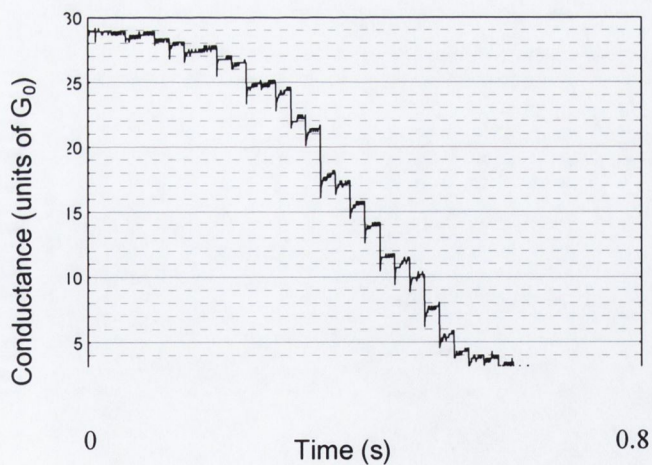


Figure 4.10. Typical variation of the conductance during the breaking of a contact between two $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystals using a piezo crystal.

By using the piezo crystal, we have been able to stabilize these nanocontacts for relatively long time periods, of the order of seconds. This has enabled us to investigate the electronic transport properties across the nanocontact in more detail. We have measured the I-V characteristics for a range of nanocontacts with different conductances in the following way. Biasing the two crystals with a small voltage of 30mV, the contact is slowly broken using the piezo crystal. The conductance is monitored continuously until it decreases to the desired value. The bias voltage is then ramped from -0.3V to 0.3V and back, while measuring the current through the contact. In Figure 4.11 we present typical I-V characteristics of six different nanocontacts obtained in this way. These contacts had, at a bias voltage of

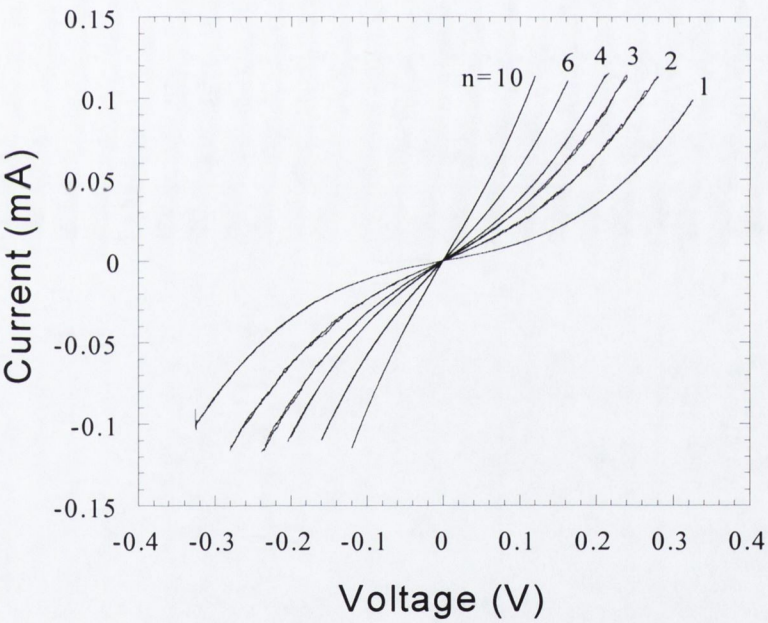


Figure 4.11. I-V characteristics for six different nanocontacts which had, at a bias voltage of 30mV, a conductance of nG_0 with $n \approx 1, 2, 3, 4, 6$ and 10. The curves show the data obtained by sweeping the voltage both up and down.

30mV, a conductance of close (within a few percent) to nG_0 , with n respectively 1, 2, 3, 4, 6 and 10. The data is remarkably reproducible, even after many consecutive breaks. For low voltages ($<0.05V$) the current is linear with the applied bias voltage. However, at higher

voltages, all the curves show a non-linear behaviour, which is most pronounced for lower values of n . For higher values the curves seem to approach the ohmic regime.

As in the case of Fe_3O_4 point contacts, the current-voltage characteristics can be fitted with a simple $I = GV + cV^3$ law. The non-linearity of these contacts in terms of their conductance can be analyzed by plotting the ratio of the non-linear and linear term. This is shown in Figure 4.12. We find that at room temperature there is a wide spread of the data, and no clear trend is observed, but for all the contacts the ratio has the same order of magnitude.

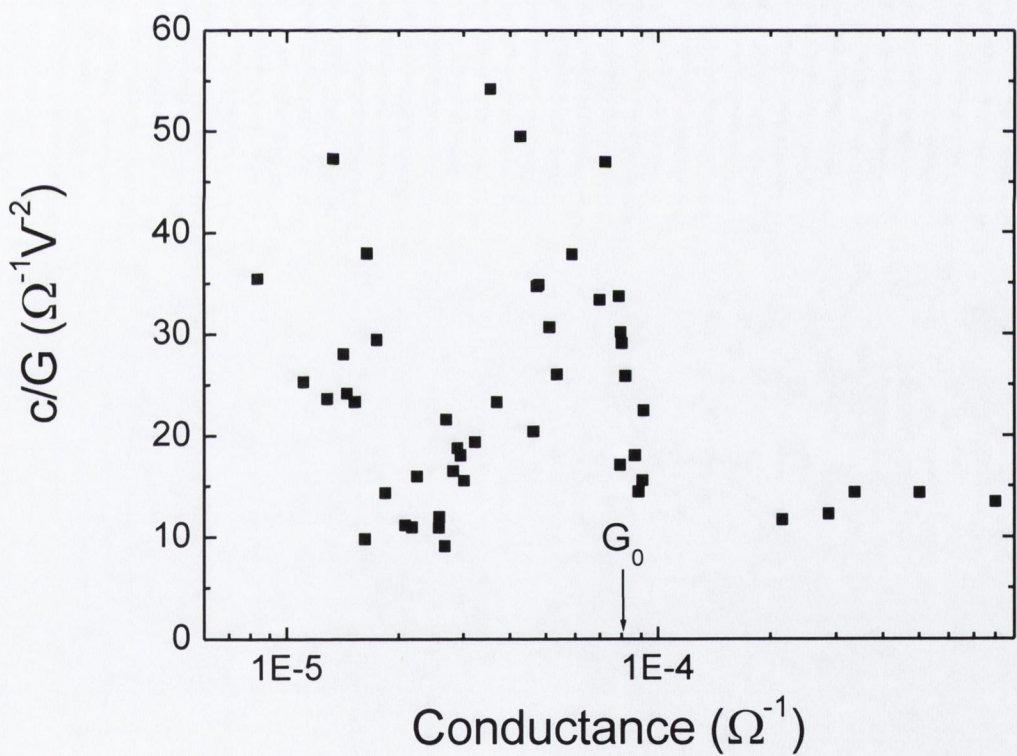


Figure 4.12. The ratio of the non-linear and linear contributions to the total current as function of conductance for LSMO point contacts at room temperature in zero field.

The transport properties of the LSMO point contacts are dramatically altered by ap-

plying a small magnetic field. Shown in Figure 4.13 are the current-voltage characteristics of a LSMO point contact at room temperature with and without a field of 7 mT. Both current-voltage characteristics, obtained by sweeping the voltage up and then downwards, are non-linear. The zero bias, zero field resistance of the point contact is about 39 k Ω , which is reduced by almost half to 21 k Ω by application of the field. This corresponds to a magnetoresistance of about 45 %. By taking the slope of the current-voltage characteristics, we can also determine the magnetoresistance at finite bias voltages. We find that at 0.3 V the magnetoresistance drops to 39 %.

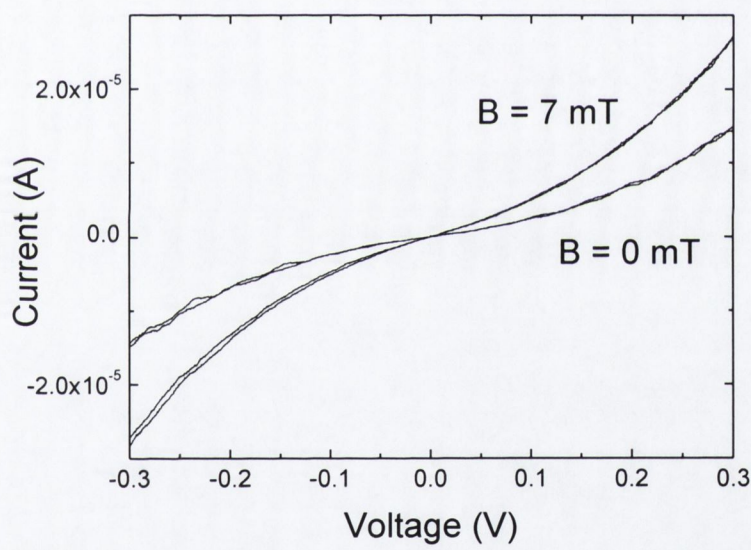


Figure 4.13. Current-voltage characteristics of a LSMO point contact with and without an applied field of 7 mT. The data is obtained by sweeping the data up and down. Zero bias resistance in zero field is 39 k Ω . Applying a 7 mT field changes this to 21 k Ω .

The non-linearity of point contacts measured in a field is, just like the zero field case, different from contact to contact. Figure 4.14 shows the ratio c/G as function of the conductance. A wide spread is observed, but all points lie within the same order of magnitude.

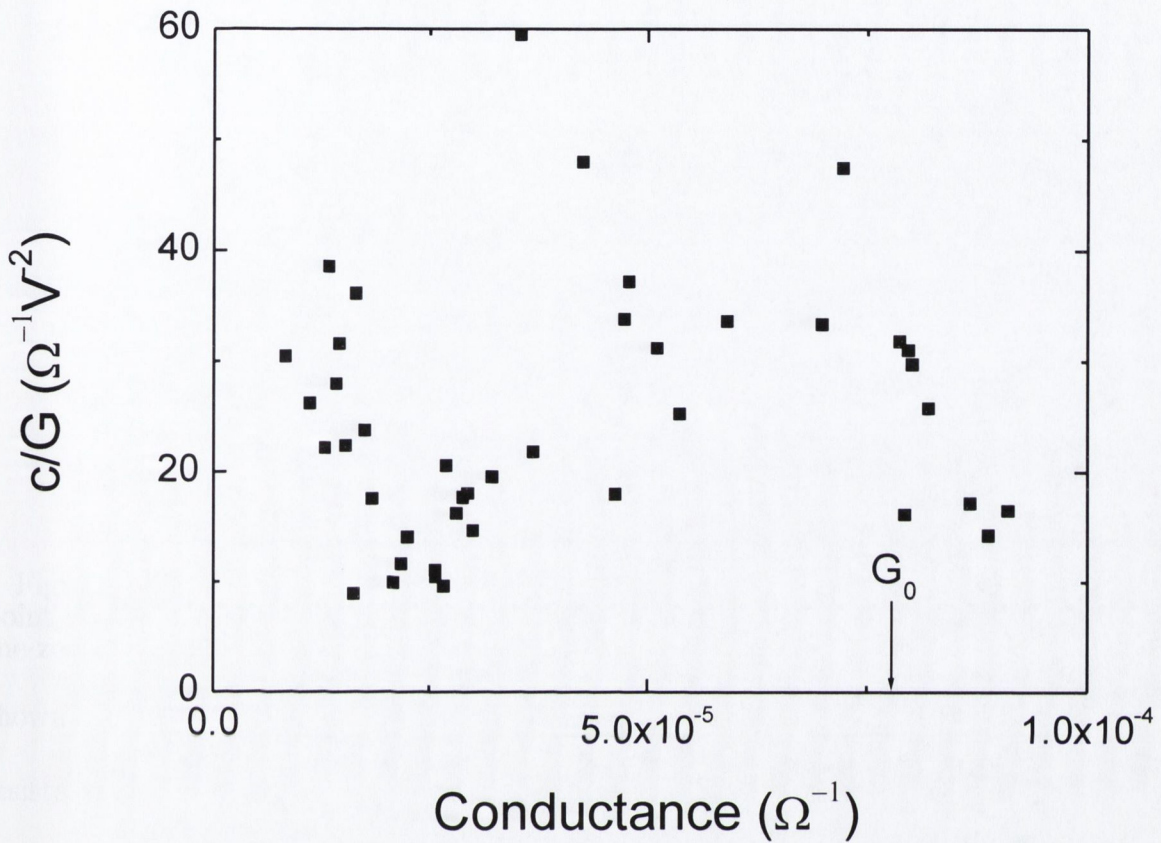


Figure 4.14. The ratio of the non-linear and linear contributions to the total current as function of conductance for LSMO point contacts in a field of 7 mT.

To compare the non-linearity of the current-voltage characteristics of point contacts with and without field, we plot the ratio $\frac{c/G(7mT)}{c/G(0mT)}$ as a function of the (zero field) conductance. See Figure 4.15. There is a wide spread of values, but for the contacts with low conductance values there is tendency for a reduction of the non-linearity with applied field.

We have measured current-voltage characteristics of many point contacts, and find that the magnetoresistance is in most cases negative, i.e, the resistance of a point contact drops when a field is applied. Sometimes we have to sweep the field a number of times before a sizeable magnetoresistance is observed. This "training" of the point contacts is

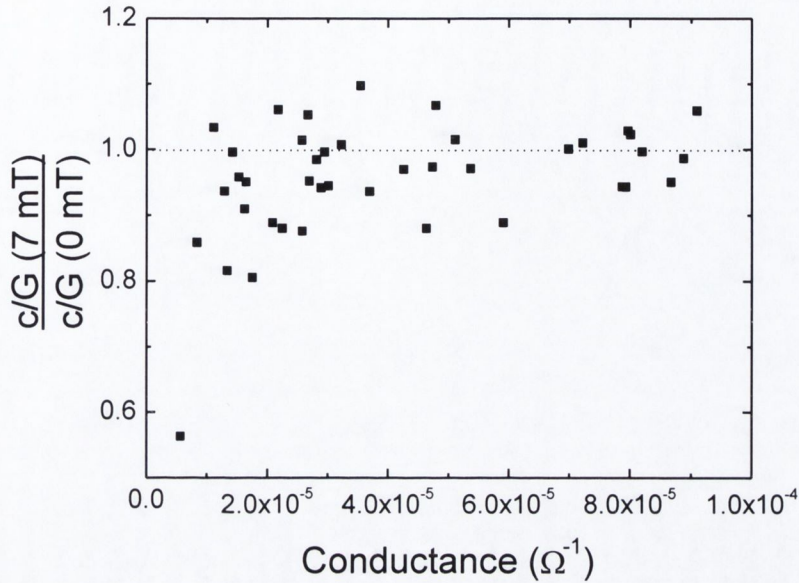


Figure 4.15. The ratio of the non-linearity of current-voltage characteristics of LSMO point contacts with and without a 7 mT applied field at room temperature, as function of the zero field conductance.

shown in Figure 4.16, and is similar to what is observed in Fe_3O_4 (See Section 4.2.2.3). The resistance of the point contacts is not stable, varying from $\sim 50 - 65 \text{ k}\Omega$, but the observed magnetoresistance becomes larger as the field is being swept.

A summary of the magnetoresistance observed in LSMO point contacts is shown in Figure 4.17. Here we plot the magnetoresistance for bias voltage $V = 0$ and $V = 0.3 \text{ V}$ as function of the conductance of the point contacts. Some point contacts show a (small) increase of resistance when a field is applied, leading to a magnetoresistance value which is negative. The majority however behave like the point contact shown in Figure 4.13, where the resistance drops in a field. For point contacts with a large conductance, i.e. a low resistance, the magnetoresistance is low, a few percent. For smaller conductances, the magnetoresistance increases, up to values as large as 45 %. For even smaller conductance values the magnetoresistance seems to drop again. The magnetoresistance values for finite

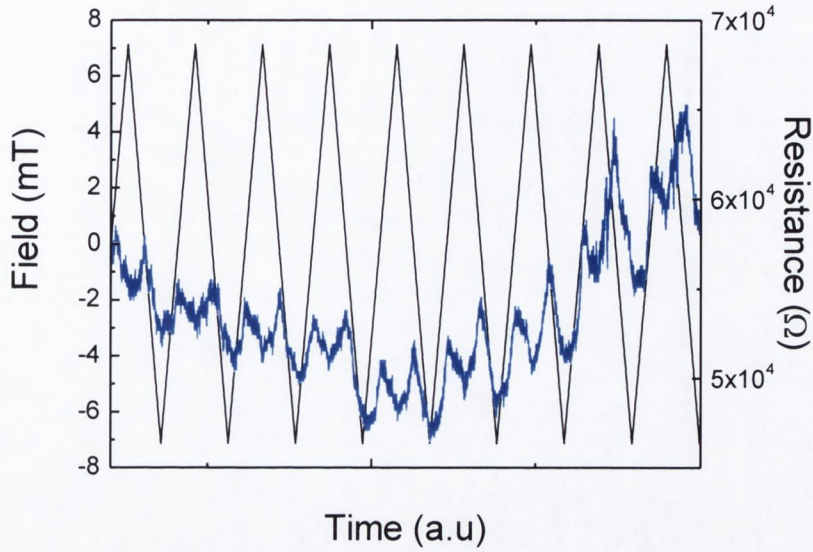


Figure 4.16. Evolution of the magnetoresistive behaviour of a LSMO nanocontact with cycling of the applied field. Resistance and applied field as a function of time are denoted by the solid black line and blue line, respectively.

bias voltages is usually lower than that for zero bias voltage.

A further idea of the magnetoresistive behaviour as function of bias voltage can be obtained by plotting the data from Figure 4.13 following Equation (4.2), as shown in Figure 4.18. The magnetoresistance shows a smooth decrease from its maximum value of around 45 % at $V = 0$. It appears to be somewhat asymmetric with respect to the sign of the applied bias voltage, with slightly higher values of magnetoresistance obtained for negative bias voltages than for positive bias voltages.

The results shown so far were all obtained in air at room temperature. Using the cryostat, with liquid nitrogen, current-voltage characteristics have also been obtained for point contacts at lower temperatures. In Figure 4.19 we show data obtained at $T = 150$ K. At this temperature, it becomes rather difficult to obtain stable point contacts. Contacts tend to break very suddenly, and it becomes very hard to obtain contacts with values of

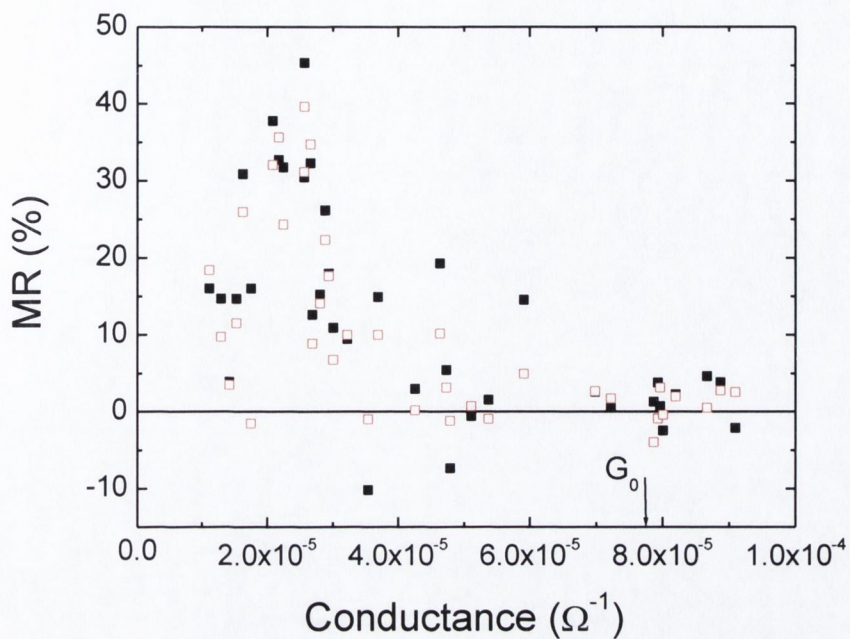


Figure 4.17. The magnetoresistance of 40 LSMO point contacts as function of the (zero field, zero bias) conductance. Magnetoresistance at zero bias is denoted by (■), that at 0.3 V by (□). Note that the magnetoresistance value cannot exceed 100 % in the definition used here.

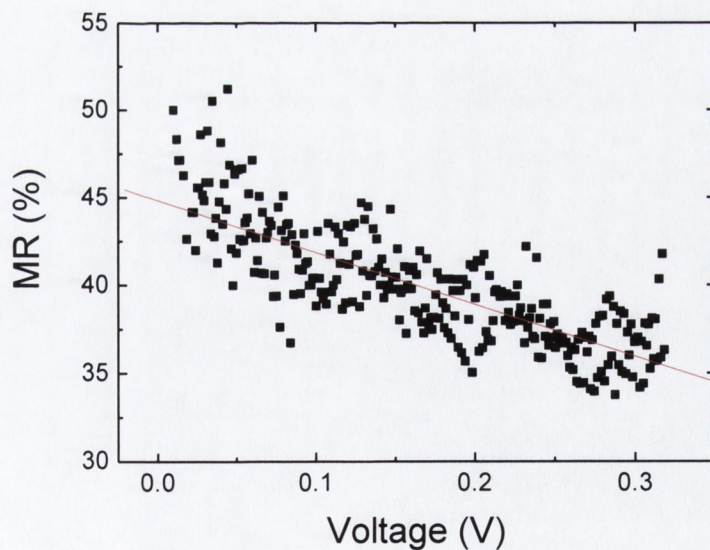


Figure 4.18. Magnetoresistance of an LSMO point contact as a function of applied bias voltage in a field of 7 mT. The solid line is a linear fit to the data.

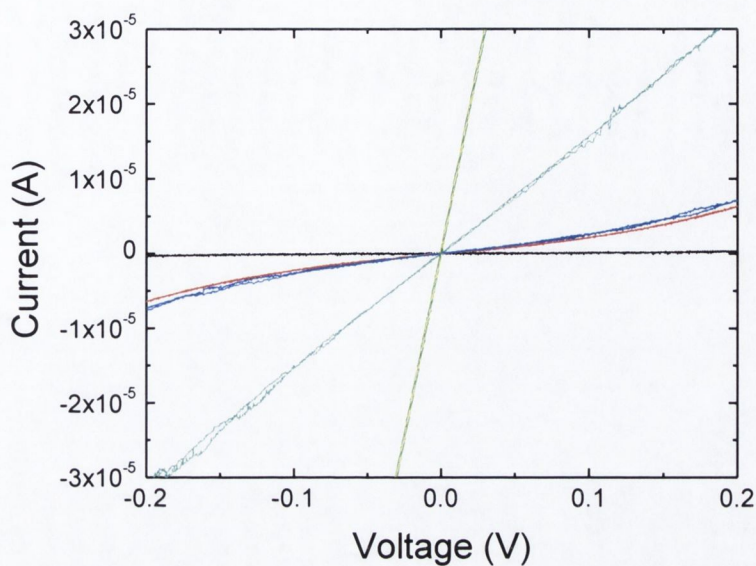


Figure 4.19. Current-voltage characteristics of several LSMO point contacts at 150 K. The resistances range from 3 k Ω to 5 M Ω .

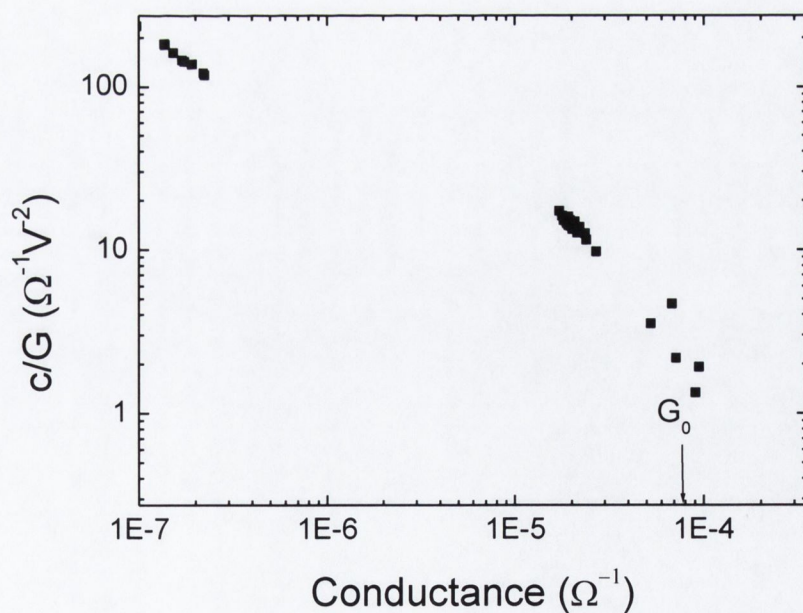


Figure 4.20. The ratio of non-linear and linear contributions to the current in LSMO point contacts at 150 K, as function of the conductance.

resistance larger than about 50 k Ω . This is reflected in Figure 4.19 with resistances ranging from 5 M Ω down to 3 k Ω . The contacts obtained are all non-linear, but again this depends on the value of resistance, as shown in Figure 4.41. The lack of data for intermediate values of conductance illustrates once more the difficulty of getting point contacts with resistances in this range. Measuring current-voltage characteristics with and without an applied field of 7 mT shows a negligible magnetoresistance at this temperature.

A remarkable phenomena observed in current-voltage characteristics at this temperature is that of telegraph noise. Figure 4.21 shows the current-voltage characteristics of a point contacts that seems to switch between a high and a low resistance state as the voltage is swept. The dotted lines are fits to an $I = GV + cV^3$ law. We have observed several point contacts where this phenomena occurs, with sometimes only two resistance states, sometimes more. In the same figure the ratio of current in the high and low resistance states is plotted, which is the inverse of the ratio of the resistances. As can be clearly seen, this ratio is not constant, but drops off with increasing bias voltage.

Figure 4.22 shows current-voltage characteristics of several LSMO point contacts at 77 K. The point contacts have resistances that range from 17 – 95 k Ω . The non-linearity of the current-voltage characteristics is shown in Figure 4.23. Again it shows that the contacts with the highest resistance are the most non-linear.

Finally, we compare the non-linearity of current-voltage characteristics of point contacts measured at three different temperatures (room temperature, 150 and 77 K). The room temperature data is showing a wider range of values, and no clear trend is observed, as in the lower temperature data. However, the values measured at room temperature are generally higher than the 150 K data, which in turn are generally higher than the 77 K data,

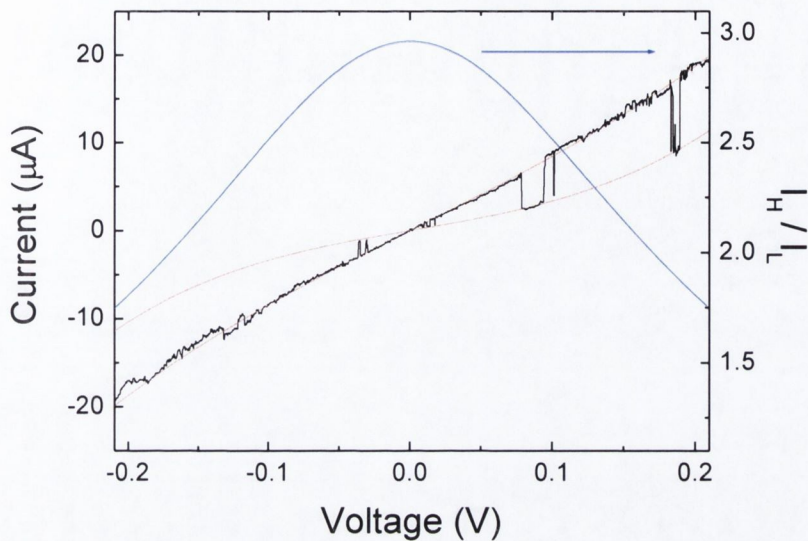


Figure 4.21. Current-voltage characteristics of a LSMO point contact at 150 K. The point contact switches between high and low resistance states as the voltage is swept. The dotted lines shows the fits to an $I = GV + cV^3$ law. The blue line shows the ratio of the currents in the high and low resistance states.

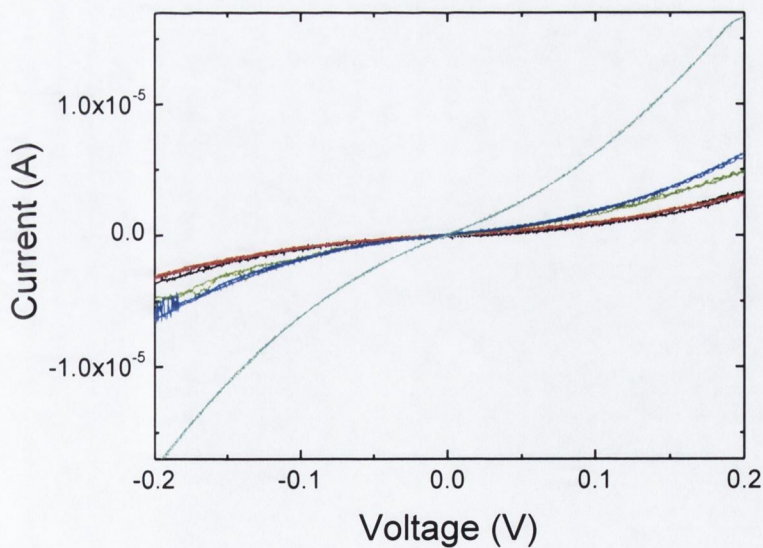


Figure 4.22. Current-voltage characteristics of several LSMO point contacts at 77 K.

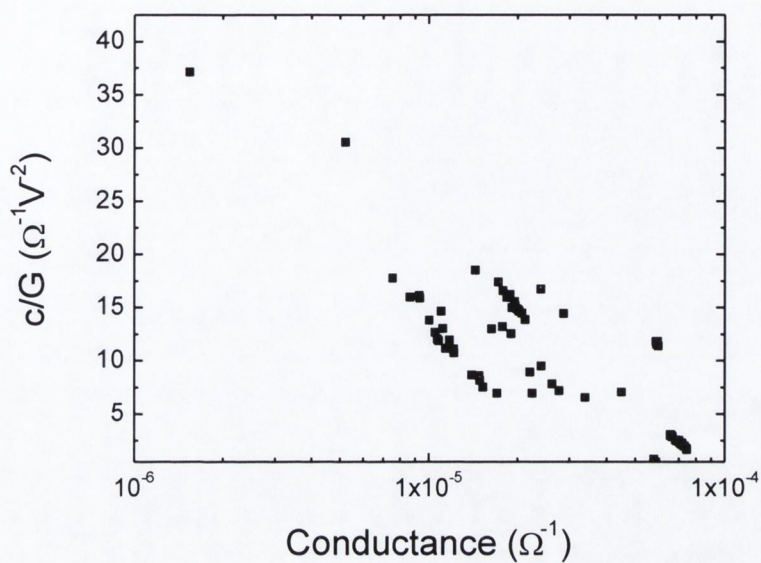


Figure 4.23. The ratio of non-linear and linear contributions to the current in LSMO point contacts at 77 K, as function of the conductance.

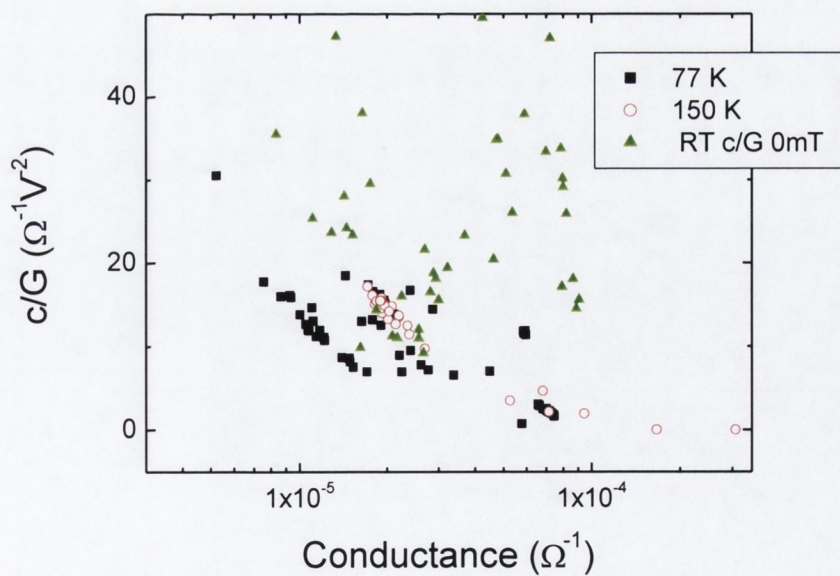


Figure 4.24. The ratio of non-linear and linear contributions to the current in LSMO point contacts at room temperature, 150 K and 77 K, as function of the conductance.

indicating that the non-linearity becomes less for lower temperatures.

4.2.2 Fe_3O_4

4.2.2.1 Sample characterization

The Fe_3O_4 crystals used in the experiments were grown by chemical vapor transport [4]. Figure 4.25 shows scanning electron microscope pictures of these tiny magnetite crystallites (usually $< 0.1 \text{ mm}^3$), with the typical octahedral structure clearly visible. Their composition was confirmed using Energy Dispersive X-ray Analysis and X-ray diffraction. The magnetic properties were investigated using SQUID and VSM. Magnetization of a single crystallite as function of applied field was measured for three different temperature: room temperature, 150 K (well above the Verwey transition temperature), and below (77 K), see Figure 4.26. The sample magnetization is saturated in a field of 0.5 T for all three temperatures. The slight diamagnetic contribution, i.e. the decrease in magnetization at high field at room temperature, can perhaps be ascribed to the presence of some vacuum grease on the sample. The inset, with a close-up of the region around the coercive field, shows the coercivity to be around 1 mT for temperature $T \geq 150 \text{ K}$, and around 5 mT for $T = 77 \text{ K}$. The temperature dependence of the magnetization in a field of 0.3 T (below saturation) is shown in Figure 4.27. Around the Verwey transition the magnetization reaches a maximum, with a gradual decrease of magnetization with increasing temperature. Magnetization as a function of field for a group of crystallites at room temperature is shown in Figure 4.28. The crystallites are saturated in a field of a few hundred milliTesla, with a negligible coercive field. The saturation magnetization value for this assembly of crystallites is lower than that for the single crystallite measured by SQUID (Figure 4.26). Using a multimeter and a copper plate, it is possible to make a two point measurement of the resistance of these tiny crystallites at

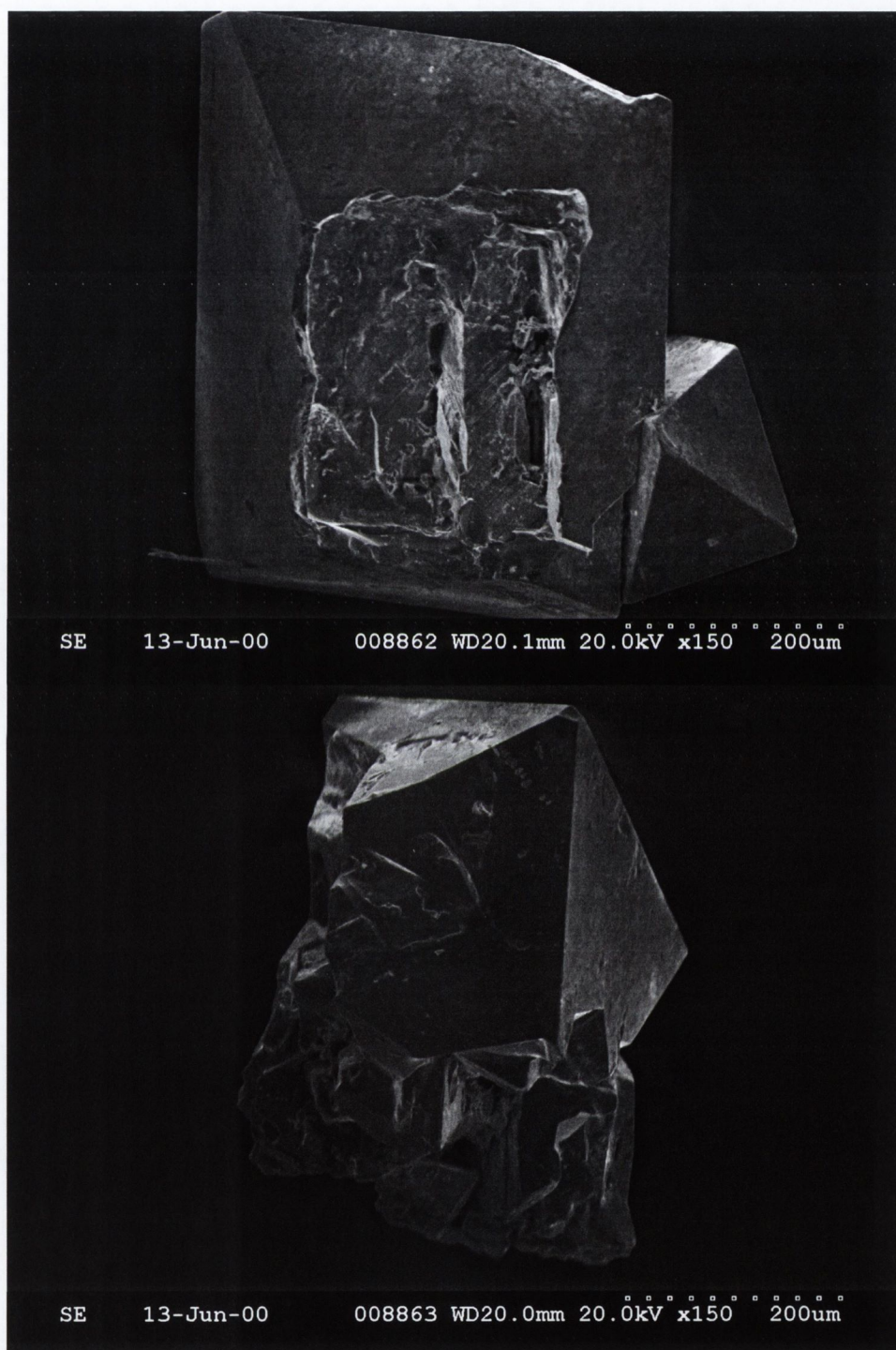


Figure 4.25. Scanning Electron Microscope pictures of the Fe_3O_4 crystallites.

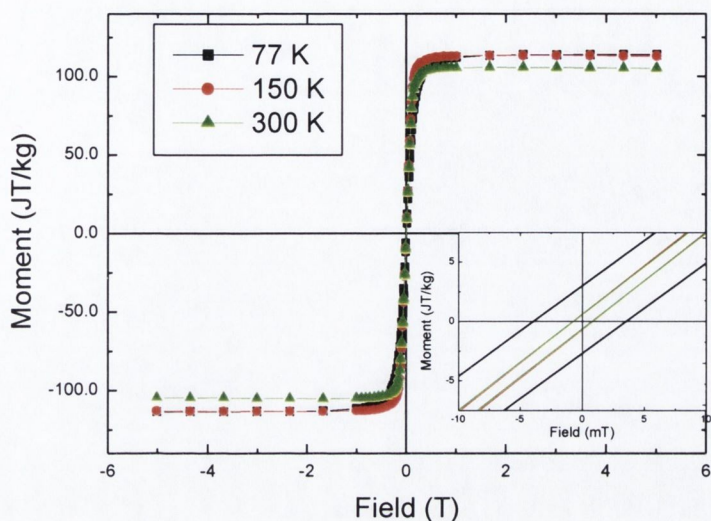


Figure 4.26. Magnetization of a single Fe_3O_4 crystallite as a function of applied field for three different temperatures: 300 K, 150 K and 77K, respectively. The inset shows a close-up of the region around the coercive field.

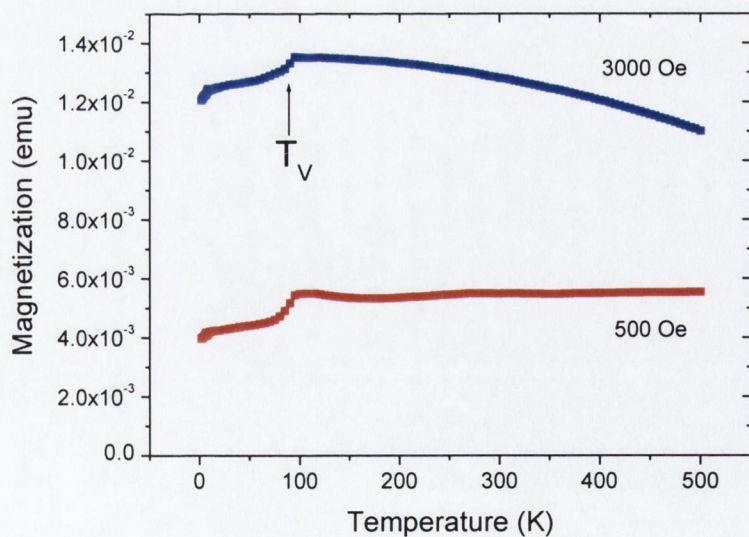


Figure 4.27. SQUID measurement of magnetization of Fe_3O_4 crystallites as a function of temperature, at fields of 3000 and 500 Oe. The Verwey transition is at 100 K.

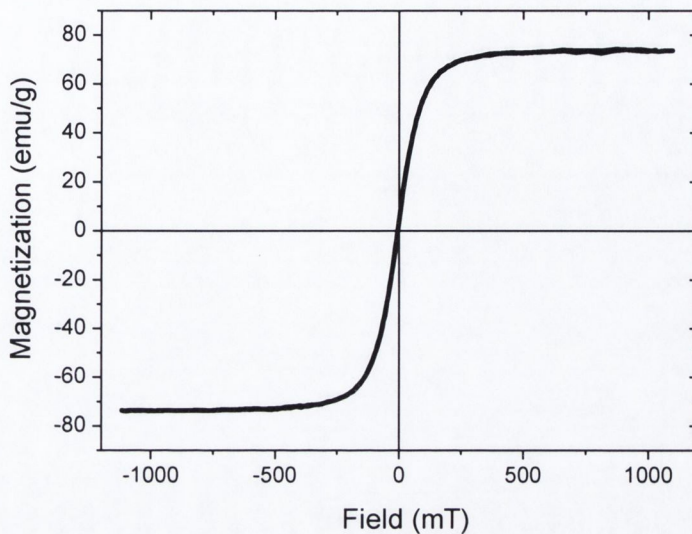


Figure 4.28. VSM measurement of the magnetization of Fe_3O_4 crystallites as a function of applied field.

room temperature, giving values of a few hundred Ohm. The room temperature resistivity of single crystal magnetite is $5 \text{ m}\Omega \text{ cm}$, which should give a resistance of order $1 \text{ }\Omega$ for our crystallites. This discrepancy might be attributed to the difficulty in placing the point probes on the tiny crystallites and making a good contact.

4.2.2.2 Quantum conductance

Using a relay set-up, breaking contacts between Fe_3O_4 crystallites were investigated at room temperature. A histogram based on 500 breaks shows only a weak peak near G_0 , which is suppressed in an applied field of 7 mT (Figure 4.29). This behaviour is different from that of LSMO (See Figure 4.9), where clear signs of quantized conductance are found.

4.2.2.3 Transport properties

The piezo set-up gives more control over the point contact. By slowly varying the piezo voltage, the conductance of the contact changes continuously, close to three orders

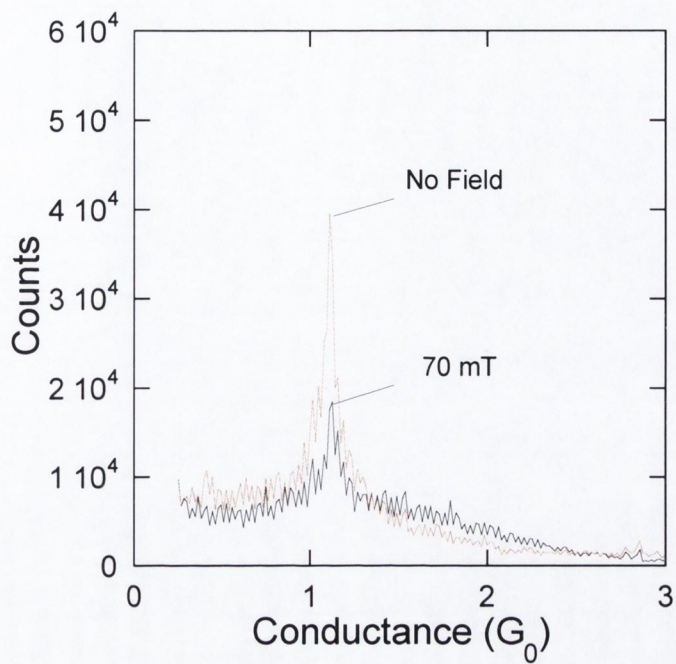


Figure 4.29. Histogram based on 500 breaks of Fe_3O_4 crystallites contacts, with and without an applied field of 70 mT.

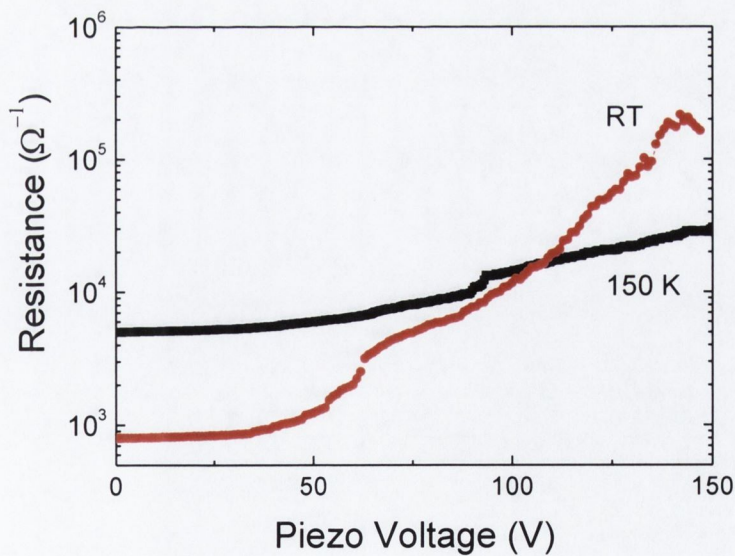


Figure 4.30. Resistance of a Fe_3O_4 point contact as a function of applied piezo voltage, at room temperature and 150K.

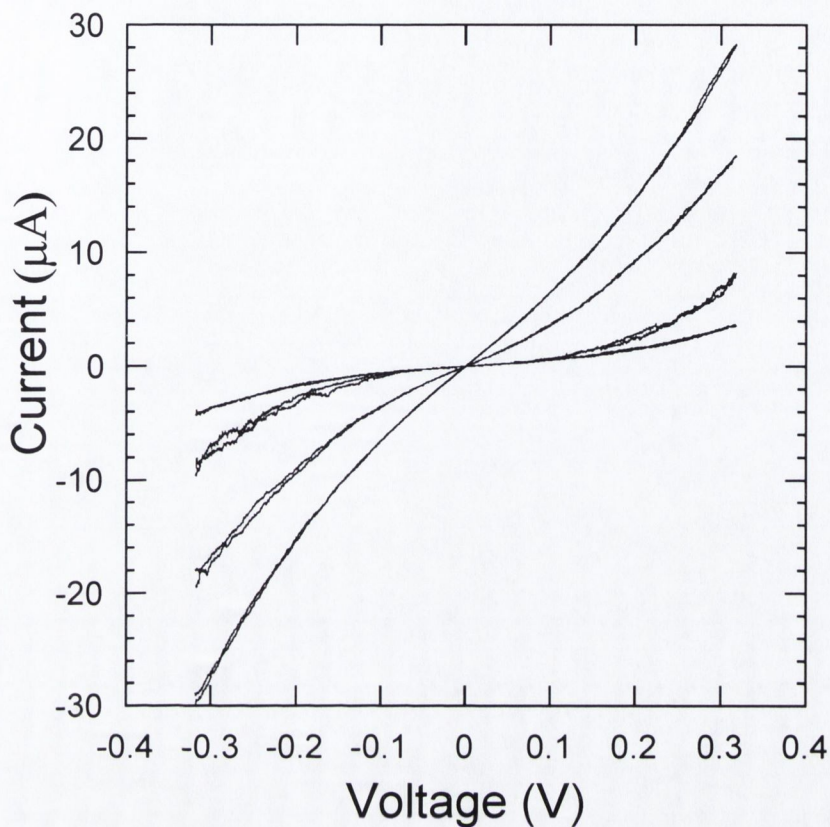


Figure 4.31. Fe_3O_4 point contacts' current-voltage characteristics for different values of conductance. The data is obtained by sweeping the voltage up and down. Zero bias resistance ranges from $\sim 10 - 200 \text{ k}\Omega$.

of magnitude, at room temperature, as shown in Figure 4.30. At low temperatures the resistance varies much slower with piezo voltage, reflecting the temperature dependence of the piezo coupling constant. By monitoring the resistance of the point contact while varying the piezo voltage, it is possible to stabilize contacts at different values of conductance G . These contacts are stable for up to a minute or more, long enough to measure the magnetotransport properties of these contacts. Figure 4.31 shows the current-voltage characteristics of point contacts stabilized in this way, with (zero bias voltage) resistances ranging from $\sim 10 - 200 \text{ k}\Omega$. The data presented is obtained by sweeping the voltage up- and downwards. The current-voltage characteristics all show a strong non-linearity, but they become more ohmic

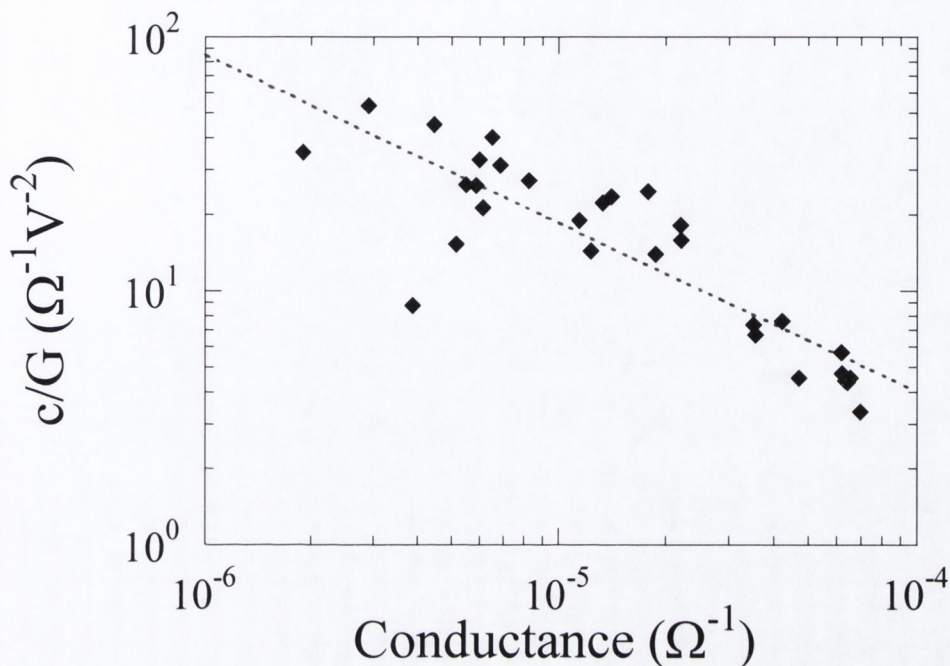


Figure 4.32. The ratio of the non-linear and linear contributions to the total current as function of conductance for Fe_3O_4 point contacts in zero field. The dotted line is a power-law fit of the data.

the lower the resistance. This can be shown in the following way. The current-voltage characteristics can be fitted with the following expression:

$$I = I_{\text{linear}} + I_{\text{non-linear}} = GV + cV^3, \quad (4.1)$$

where I is the measured current, V the applied bias voltage, and G and c the fit parameters. Plotting the ratio of the non-linear part and linear part, c/G , as a function of the (zero bias) conductance G (Figure 4.32), it can be seen that the non-linear part becomes less important for point contacts with a higher conductance. Fitting the data to a power law (dotted line) leads to the empirical relation $c \propto G^{0.3}$.

The application of a magnetic field dramatically changes the transport properties of the point contacts. Using the piezo set-up, the point contacts are stable for long enough to

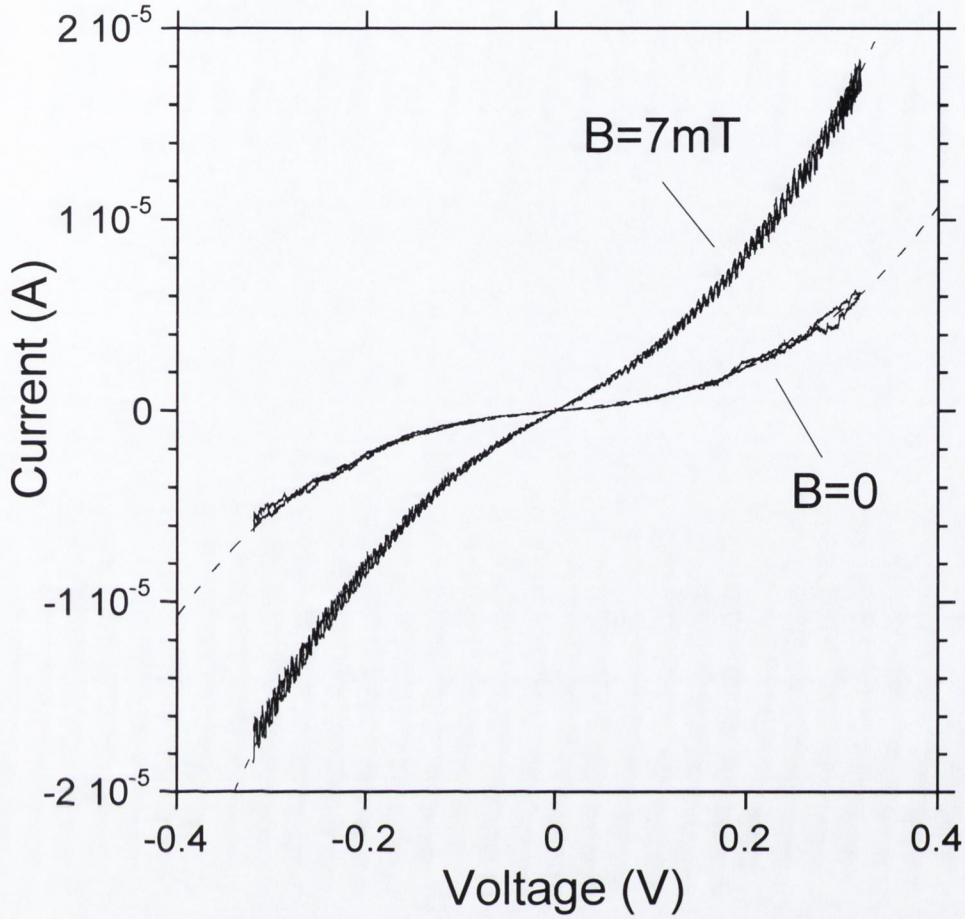


Figure 4.33. Current-voltage characteristics of a Fe_3O_4 point contact with and without an applied field of 7 mT. The data is obtained by sweeping the data up and down. Zero bias resistance in zero field is 170 k Ω . Applying a 7 mT field changes this to 28 k Ω .

measure the current voltage characteristics with and without a small applied field. Figure 4.33 shows how the conductance is dramatically increased, by a factor of 5.4, when a field of 7 mT is applied. This corresponds to a magnetoresistance $MR = -\frac{R(H)-R(0)}{R(0)} = 85\%$. The non-linearity of the current-voltage characteristics as function of conductance of point contacts in 7 mT is shown in Figure 4.34. Again it is found that the non-linearity is larger for contacts with a higher resistance, but it is smaller in a field compared with the zero field case: $c \propto G^{0.5}$. The influence of field on the non-linearity of the current-voltage characteristics can

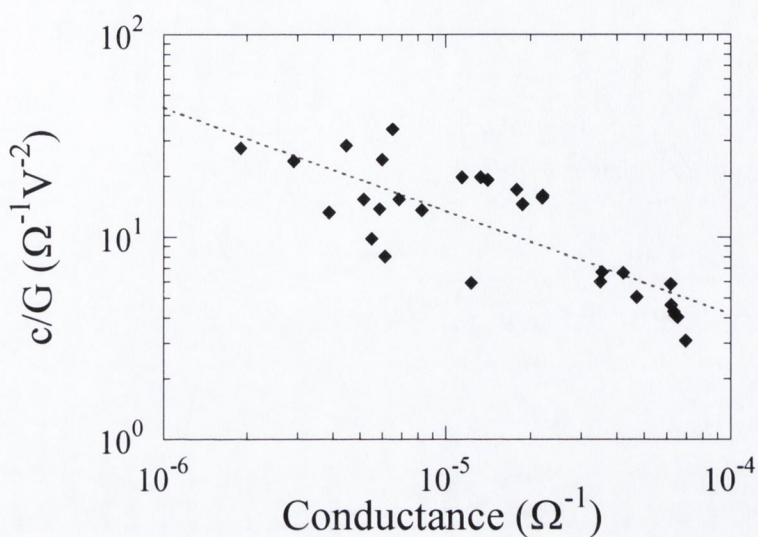


Figure 4.34. The ratio of the non-linear and linear contributions to the total current as function of conductance for Fe_3O_4 point contacts in a field of 7 mT. The dotted line is a power-law fit of the data.

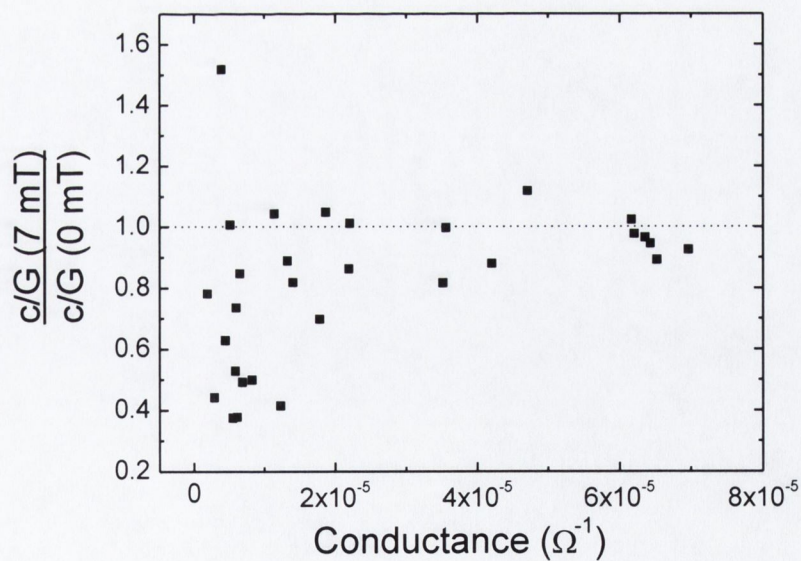


Figure 4.35. The ratio of the non-linearity of current-voltage characteristics of Fe_3O_4 point contacts with and without a 7 mT applied field at room temperature, as function of the zero field conductance.

also be shown by plotting the ratio $\frac{c/G(7mT)}{c/G(0mT)}$ as a function of the (zero field) conductance. See Figure 4.35. It can be seen that the non-linearity for low conductance contacts is generally reduced by applying a field.

During the course of our study of oxide point contacts presented here, we have measured the transport properties of thousands of contacts, and in particular the magnetotransport properties. In the vast majority of cases it is found that the magnetoresistance is negative, i.e. the resistance is reduced in a field. A feature often encountered in these experiments on point contacts is that the magnetoresistive effect becomes larger after cycling the applied magnetic field many times. A typical evolution of the magnetoresistive behaviour of a Fe_3O_4 nanocontact is shown in Figure 4.36. The breaking of a contact was arrested when the contact resistance reached $\sim 20 \text{ k}\Omega$. At this point, an $\pm 6 \text{ mT}$ oscillating magnetic field, with triangular waveform, was applied, and the contact resistance monitored. At first, no magnetoresistance was observed in this point contact, but after about 25 cycles of the field, a magnetoresistance becomes apparent. The magnetoresistance becomes larger as the contact becomes more resistive. We refer to this cycling of the field to increase the magnetoresistance as the 'training' of the point contact. Not all point contacts need training, for some the magnetoresistance is apparent from the outset. Others contacts break before any magnetoresistance can be observed. It should be noted that the magnetoresistance does not only appear after the contact becomes more resistive. It is also seen in contacts that become less resistive, but in general, the magnetoresistance of these point contacts is larger for more resistive contacts.

For stable point contacts we can study the resistance as a function of field. Figure 4.37 shows how the resistance exhibits hysteretic behaviour when applying a variable field,

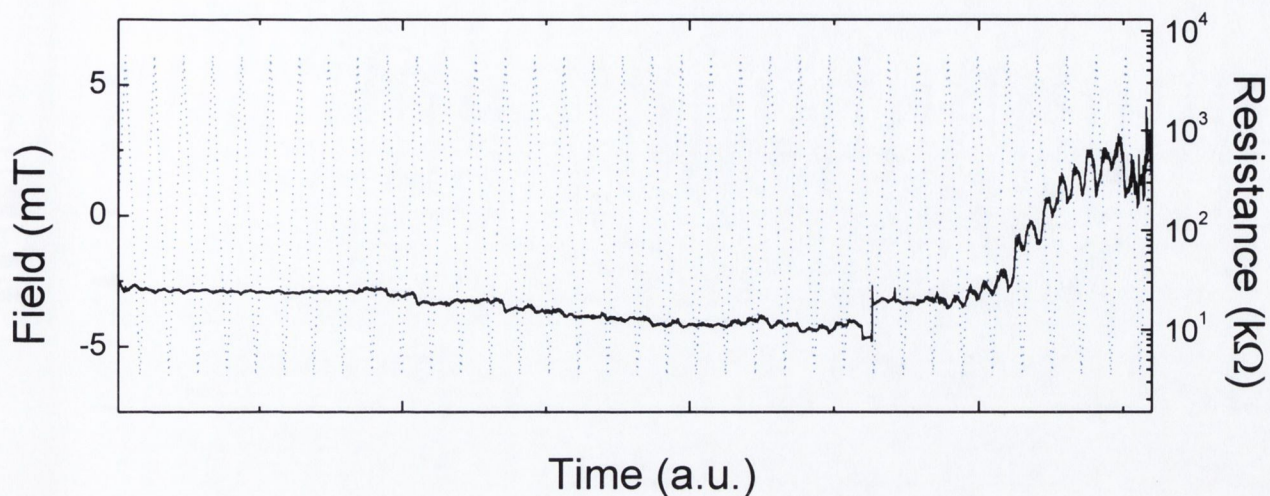


Figure 4.36. Evolution of the magnetoresistive behaviour of a Fe_3O_4 nanocontact with cycling of the applied field. Resistance and applied field as a function of time are denoted by the solid black line and dotted blue line, respectively.

leading to a typical 'butterfly' curve. It is found that the resistance of the contact is reduced in a field, leading to an MR of about 70 % in 6 mT. After cycling the field the resistance does not quite reach its original value again, but has increased by about 10 kΩ. The peak in the resistance occurs at around 1 mT, which is the value found for the coercivity of the Fe_3O_4 crystallites (See Figure 4.26).

Keeping the training of the nanocontacts in mind, our current-voltage measurements were carried out in the following way. The contact under investigation is slowly broken while monitoring its resistance. At the desired value of resistance this is stopped, and a current-voltage characteristic is obtained. A magnetic field is then swept quickly from +6 mT to -6 mT for forty times before another current-voltage characteristic is measured in a 6 mT field. This is then followed by another one without a field and one in a field.

We have measured the current-voltage characteristics of point contacts with a wide range of conductance values. Figure 4.38 shows how the magnetoresistance depends on the

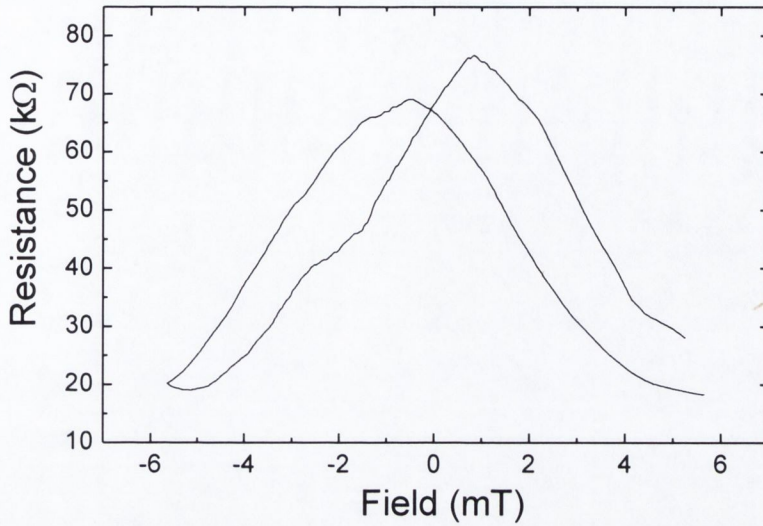


Figure 4.37. Resistance as a function of applied field for a Fe₃O₄ point contact at room temperature.

conductance of the point contacts. Plotted are the magnetoresistance values, at 0.3 V (○) and zero bias voltage (■), for thirty point contacts. Point contacts with similar conductance values exhibit different values of magnetoresistance, but the general trend is clear: the lower the conductance value the higher the magnetoresistance. For the least conducting point contacts the zero bias magnetoresistance can be as high as 85 % (out of a maximum possible 100 % in the definition used here), but it drops quickly, to around 10 %, for conductance values near G_0 . The zero bias voltage magnetoresistance value is higher than the 0.3 V one.

By plotting the current-voltage characteristics of Figure 4.33 in a different way (see Figure 4.39) we can measure the magnetoresistance of this point contact, in 7 mT, as function of the applied bias voltage. Here we calculate the magnetoresistance MR as follows:

$$MR(V, 7 \text{ mT}) = \frac{I(V, 7 \text{ mT}) - I(V, 0)}{I(V, 7 \text{ mT})} \times 100\% = \frac{R(V, 0) - R(V, 7 \text{ mT})}{R(V, 0)} \times 100\% \quad (4.2)$$

with $I(V)$ the current as function of the bias voltage V . Again we see that the zero bias

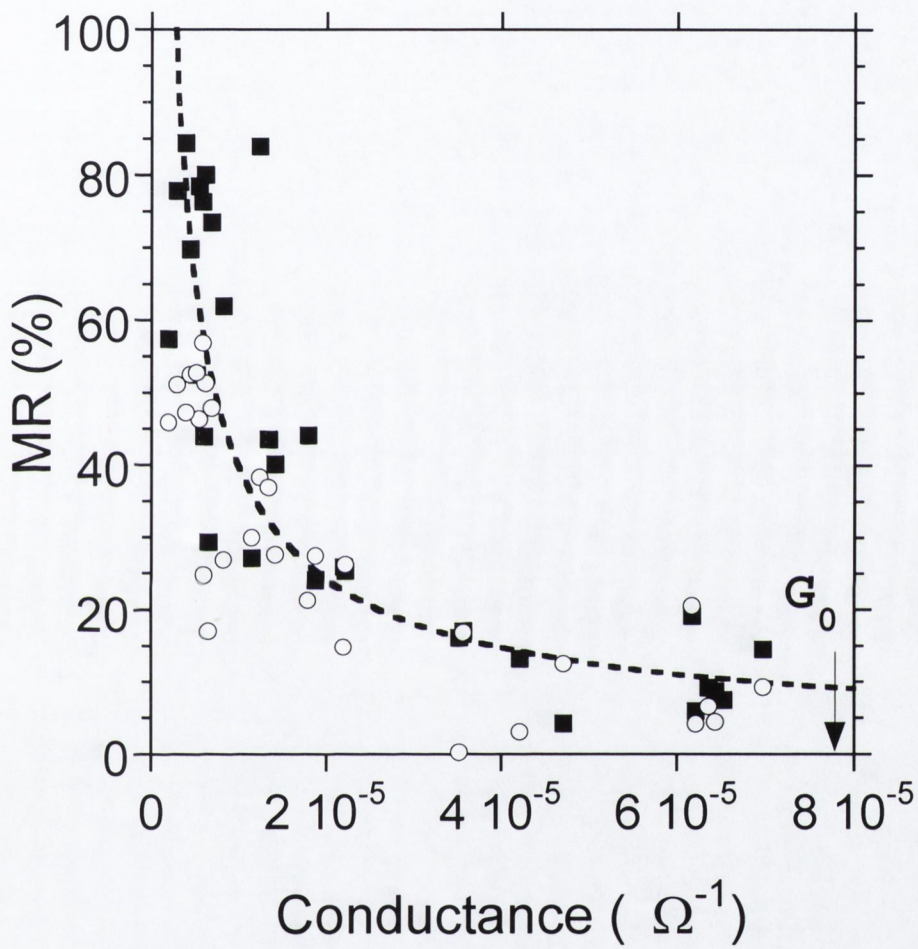


Figure 4.38. The magnetoresistance of 30 Fe_3O_4 point contacts as function of the (zero field, zero bias) conductance. Magnetoresistance at zero bias is denoted by (■), that at 0.3 V by (○). The quantum of conductance value is indicated by the arrow. The dotted line is a guide to the eye. Note that the magnetoresistance value cannot exceed 100 % in the definition used here.

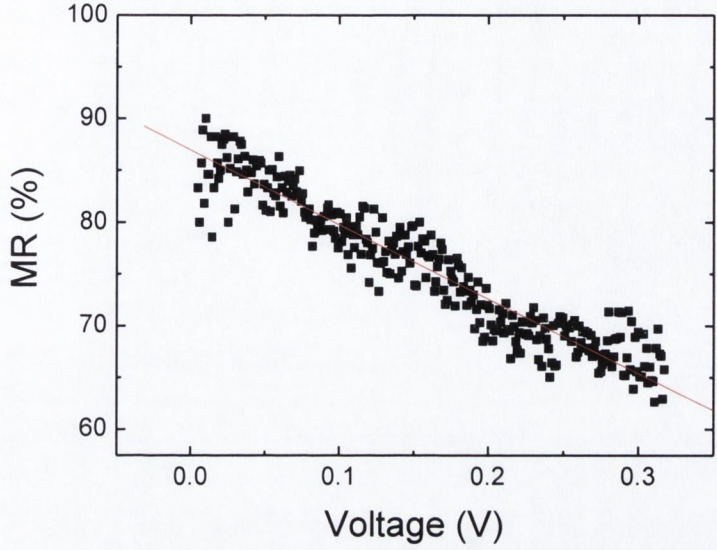


Figure 4.39. Magnetoresistance of an Fe_3O_4 point contact as a function of applied bias voltage in a field of 7 mT. The solid line is a linear fit to the data.

magnetoresistance has the highest value, of around 85 %, which drops to around 65 % at 0.3 V. The noise in current-voltage characteristic makes it difficult to determine the exact nature of the dependence of magnetoresistance on bias voltage, but it seems to be linear.

Using the cryostat set-up we have been able to measure current-voltage characteristics at lower temperatures as well. Figure 4.40 shows current-voltage characteristics at a temperature of 150 K, well above the Verwey transition temperature. They are all non-linear, and can be fitted using a simple $I = GV + cV^3$ term, as used for the room temperature data. It should be noted that at this temperature it becomes much harder to measure stable point contacts. Apart from the problems due to the reduced piezo travel length at this temperature, we sometimes observe that even though a mechanical contact between the crystallites can be made, it is not possible to establish an electrical contact. Furthermore, when it is possible to establish an electrical contact, the full contact resistance (that is, the resistance measured when the two crystallites are pressed strongly against each other) is often

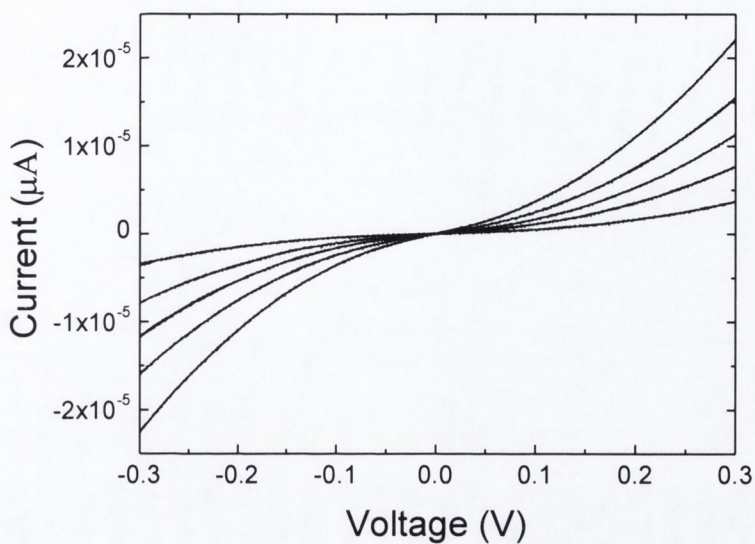


Figure 4.40. Current-voltage characteristics for several Fe_3O_4 point contacts at 150 K. Zero bias resistances range from $\sim 25 - 240 \text{ k}\Omega$.

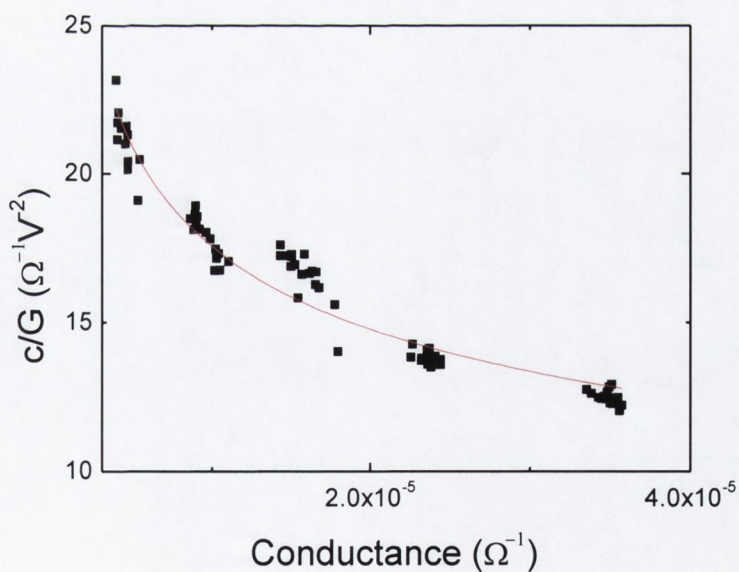


Figure 4.41. The ratio of the non-linear and linear contributions to the total current as function of conductance for Fe_3O_4 point contacts in zero field and 150 K. The line is a power-law fit of the data.

much higher than what can be expected from the room temperature full contact resistance and the temperature dependence of resistivity of magnetite.

Analyzing the non-linear part in terms of the conductance of the point contact shows again that the transport properties become more non-linear for the more resistive contacts. Figure 4.41 shows the non-linearity of the current-voltage characteristics as a function of the conductance of the contact. The power law fit (solid line) yields $c \propto G^{0.75}$, compared with $c \propto G^{0.3}$ for the room temperature data (See Figure 4.32).

We find that at a temperature of 150 K, the application of a 6 mT magnetic field does not have a noticeable effect on the conductance of the contact. This field is higher than the coercive field at this temperature, as measured in a SQUID (See figure 4.27).

At a temperature of 77 K (below the Verwey temperature), it becomes even more difficult to obtain stable point contacts. A few current-voltage characteristics of highly resistive point contacts have nonetheless been obtained. Again they are all non-linear, with the more resistive contacts becoming more non-linear.

To conclude this section, we plot in Figure 4.43 the non-linearity of current-voltage characteristics measured at room temperature and 150 K. The most non-linear characteristics are found in low conductance point contacts at room temperature.

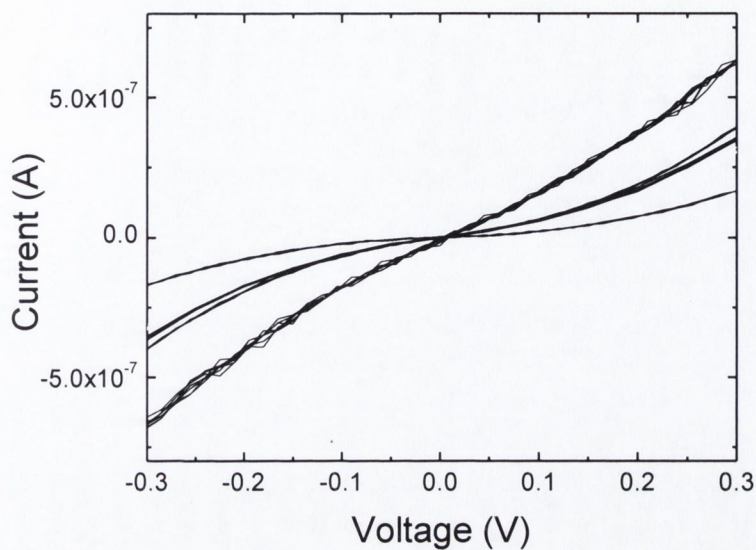


Figure 4.42. Current-voltage characteristics for several Fe_3O_4 point contacts at 77 K. Zero bias resistances range from $\sim 0.6 - 4 \text{ M}\Omega$.

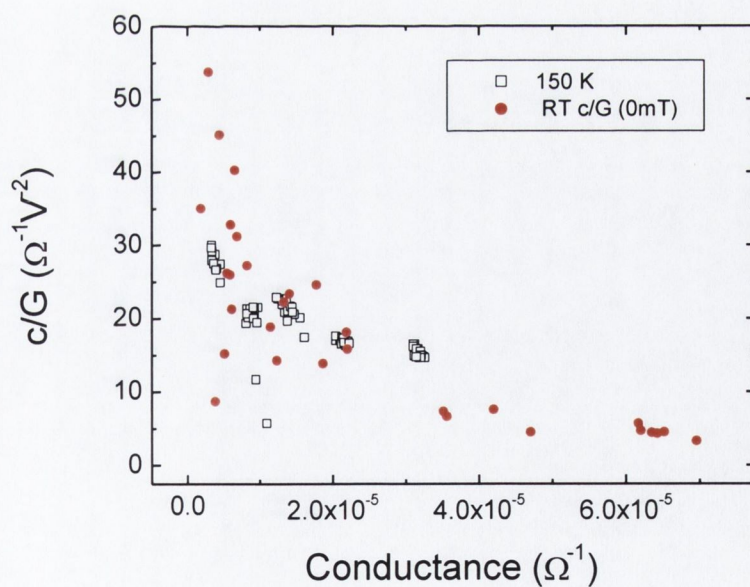


Figure 4.43. The ratio of the non-linear and linear contributions to the total current as function of conductance for Fe_3O_4 point contacts in zero field at room temperature and 150 K.

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Chapter 5

Discussion & Further Work

5.1 Scanning Tunneling Potentiometry

Scanning Tunneling Potentiometry on polycrystalline LSMO thin films shows a wide range of values for the grain boundary resistance, r_{GB} . This can be explained in terms of a distribution of grain boundary structure and angle between the grains [1]. An average for these films is $r_{GB} = 6 \times 10^{-6} \Omega\text{cm}^2$. Table 1 shows how this value for the grain boundary resistivity compares with other reports in the literature [2][3][4].

	Material	GB resistivity (Ωcm^2)	Temp. (K)
Gupta <i>et al.</i> [2]	$(\text{La}_{0.67}\text{Ca}_{0.33})\text{MnO}_3$	6×10^{-5}	10
		2×10^{-5} (Field indep. comp.)	10
Mathur <i>et al.</i> [3]	$(\text{La}_{0.7}\text{Ca}_{0.3})\text{MnO}_3$	$\sim 1 \times 10^{-7}$	50
Steenbeck <i>et al.</i> [4]	$(\text{La}_{0.8}\text{Sr}_{0.2})\text{MnO}_3$	4.1×10^{-6} (Field indep. comp.)	32
		2.3×10^{-6} (Field dep. comp.)	32
This study	$(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$	6×10^{-6}	RT

Table 1. Grain boundary resistivity of ferromagnetic manganite thin films as reported in the literature. The last column denotes the measurement temperature.

Recently, Grevin *et al.* [5][6] have also used scanning tunneling potentiometry to study transport in $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ thin films. In ref [6], they find potential steps located near grain boundaries, while for epitaxial films [5], a continuous variation of the potential is found.

Two unexpected features of our data are: i) A typical potential step at a grain boundary, $200 \mu\text{V}$, is significantly smaller than expected from the applied potential gradient ($20 \text{ V/cm} = 2 \text{ mV}/\mu\text{m}$). ii) Regions appear where the potential steps are in the opposite sense to the average potential gradient. Both features can be understood on the basis of simulations we have performed of the local potential in polycrystalline thin films using mesh

resistor networks with a normal distribution of the resistances. The discrepancy between

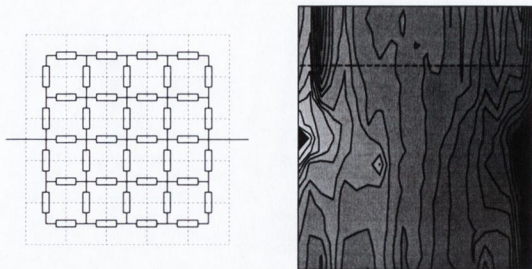


Figure 5.1. Simulation of potential distribution in polycrystalline thin films using a resistor mesh network (left). Shown on the right is the result of a simulation on a 20 by 20 network. The gray scale denotes the potential. The resistances are the absolute values of normal distribution with zero mean, and a standard deviation $\sigma = 1$. The dashed line in the simulation shows regions where the local potential gradient is of opposite sign to that of the average potential.

observed and expected potential steps is explained both by the use of point contacts and the presence of a highly resistive surface layer which causes most of the potential gradient to occur in the vicinity of the contacts. Evidence for a resistive surface layer about 15 nm thick is provided by measurements on thin continuous films of $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ on MgO [7], and extrapolation of the resistivity measured on films of different thicknesses deposited on LaAlO_3 [8]. We estimate the resistance of the surface layer to be of the order of 1 k Ω . When a local distribution of surface resistances is present it is indeed possible to have a local potential gradient that is of opposite sign to the average potential gradient. Figure 5.1 shows some simulations that exhibit the effect.

5.1.1 Future work

For future work on Scanning Tunneling Potentiometry, a visit for a few months was planned to the Institute für Physikalische HochTechnologie in Jena, Germany. After an earlier visit to the institute and the available equipment and a discussion, updated software was acquired, and a STM sample holder for use in a vacuum can designed. This will allow us

to perform STP with a higher resolution, making it possible to study grain boundaries and their electronic properties in greater detail. Especially the question if the grain boundary has a sharp potential drop or one that is spread out over a few nanometers is of great interest. Hopefully it will allow us to increase our understanding of the nature of the grain boundaries and their transport properties.

Another interesting experiment we hope to carry out is the study of the grain boundary resistance as a function of magnetic field. The field will be produced by a pair of Helmholtz coils, set up around the vacuum can which contains the STM. Placing this vacuum can in a bath of liquid nitrogen will also allow us to perform the experiments described above in a temperature regime where the low field magnetoresistance, associated with the presence of grain boundaries, is larger than at room temperature.

5.2 Point Contacts

What can explain the large room temperature magnetoresistance observed in point contacts of LSMO and Fe_3O_4 in small fields of 7 mT? The values found for magnetite (up to 85 %, using the definition $MR = -\frac{R(H)-R(0)}{R(0)}$) are much larger than anything found previously for magnetic oxide or metallic tunnel junctions at room temperature [9][10]. It is also larger than that reported for ballistic Ni nanocontacts [11]. The hysteresis in the resistance versus field curves proves that the magnetoresistance is related to the magnetization processes of the ferromagnetic crystallites, which, in view of their size, are certainly multidomain.

In the following sections we discuss three possible mechanisms that could be responsible for the observed magnetoresistance effects: 1) magnetostriction, 2) tunneling effects, and 3) electron transport through a narrow domain wall pinned by the point contact. We argue that we can discount the first mechanism as the main explanation, and focus on the idea of

tunneling and hopping transport through a narrow domain wall. We conclude with possible applications of point contacts in a fast-switching non-volatile memory element.

5.2.1 Magnetostriction

In general, materials with randomly oriented domains deform to some extent when the moments are aligned. This deformation is anisotropic, and depends therefore on the direction of magnetization, as is illustrated in Figure 5.2. The magnitude of this deformation is given by the magnetostriction coefficient λ , which is defined as $\lambda = \delta l/l$, where l is the length of the sample under investigation and δl the observed length change in the direction of magnetization. The length change in directions other than the magnetization direction is given by

$$\frac{\delta l}{l} = \frac{1}{2}\lambda_s (3 \cos^2 \theta - 1), \quad (5.1)$$

where λ_s is the saturation magnetostriction coefficient, and θ is the angle between the magnetization and measurement direction. For single crystals, there is also a dependence of λ_s on crystal directions. For cubic crystals the polycrystalline magnetostriction coefficient is then $\lambda_s = \frac{3}{5}\lambda_{100} + \frac{2}{5}\lambda_{111}$, with λ_{100} and λ_{111} the magnetostriction coefficients along the $\langle 100 \rangle$ and $\langle 111 \rangle$ direction, respectively. The saturation magnetostriction coefficients at room temperature for magnetite are $\lambda_{100} \simeq -20 \times 10^{-6}$ and $\lambda_{111} = 78 \times 10^{-6}$ [12], hence $\lambda_s = 19 \times 10^{-6}$. For LSMO the saturation magnetostriction is $\lambda_s \simeq 13 \times 10^{-6}$ [13].

Due to the small size of the magnetite crystallites, they are difficult to handle, and so we have little control over the crystal orientation. We can therefore not be certain about the sign of the length change. The magnetite crystallites used in the experiments reported here have dimensions of order $100 \mu\text{m}$ (See Figure 4.25). Therefore, alignment of the magnetic

moments in these crystallites can lead to length changes of order a nanometer. This could certainly influence the resistance of the point contacts, but we believe that magnetostriction alone is not the main explanation for our results, for reasons outlined below.

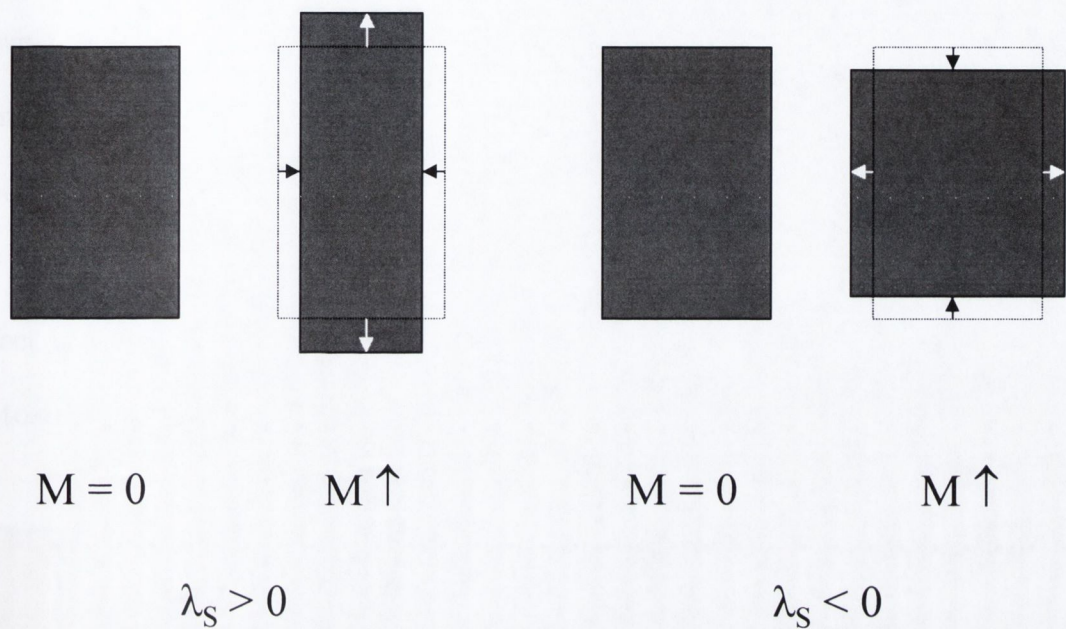


Figure 5.2. Schematic illustration of magnetostriction for positive and negative values of the saturation magnetostrictive coefficient λ_s .

- First of all, we have attempted to minimize magnetostriction by embedding the crystallites in silver paint when mounting them. The length free to expand is much less than the size of the crystal.
- Secondly, we almost always find the magnetoresistance to be of the same sign, i.e. the conductance of the contact increases when a field is applied. This is regardless of whether the field is applied parallel or perpendicular to the current direction. It could be argued that the shape anisotropy fixes the magnetization in the vicinity of the contact in a certain direction, but the crystallite as a whole is certainly multidomain, and so a bulk magnetostrictive elongation or contraction should be expected, leading to an increased or decreased contact area (see Eq. (5.1)). The fact that we find a magnetoresistance independent of field direction

is therefore a strong argument against magnetostrictive explanation for our observations.

► We have attempted to quantify the magnetostrictive contribution using the set-up shown in Figure 5.3. Using a two pairs of Helmholtz coils around the point contact, and supplying them with sinusoidal waveforms 90 degrees out of phase, we can apply a rotating field with frequency $f = 110$ Hz in the plane of the current direction. With a lock-in amplifier we can detect the f and $2f$ contributions to the resistance change in the contact. We find that the $2f$ contribution is no more than 5 % of the resistance change due to a static field. If the effect was magnetostrictive in origin we would expect a large $2f$ contribution, since the magnetostriction is independent of the sign of the field.

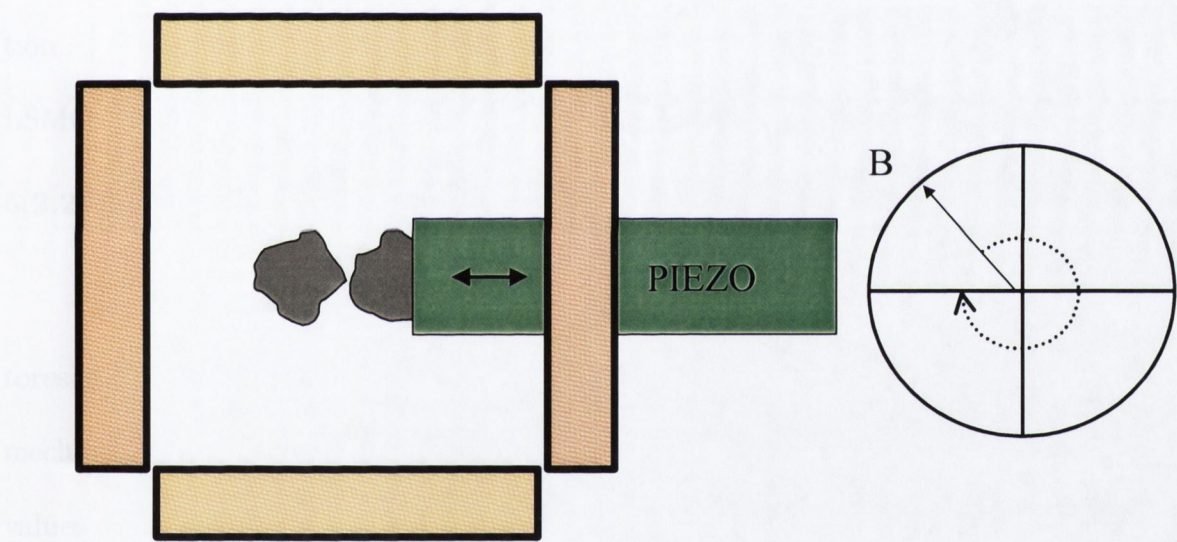


Figure 5.3. Set-up of rotating field experiment.

- In any case, there is no magnetostrictive change in length between configurations where the magnetization directions on either side of the contact are aligned parallel and anti-parallel (See again Eq. (5.1), with $\theta = 0^\circ$ and $\theta = 180^\circ$).
- At a temperature $T = 150$ K we observe no magnetoresistance for point contacts of both materials. Yet the magnetostriction coefficients for magnetite are still significant at

this temperature ($\lambda_{100} \simeq -20 \times 10^{-6}$ and $\lambda_{111} \simeq 65 \times 10^{-6}$ [12]; $\lambda_s = 14 \times 10^{-6}$), and the coefficient for LSMO is actually much larger: $\lambda_s \simeq 53 \times 10^{-6}$ [13]. If magnetostriction would be the explanation for the room temperature magnetoresistance, we should also observe a magnetostrictive magnetoresistance at $T = 150$ K.

► As a last point, we note that the magnetoresistance observed in point contacts has a bias voltage dependence (See for example Figure 4.39, where the MR drops from about 85 % at $V = 0$ to 65 % at $V = 0.3$). A magnetostrictive effect would lead to a magnetoresistance independent of voltage. Therefore, magnetostriction alone cannot explain our results.

Taking into account all the above arguments, we believe we can rule out magnetostriction as the main explanation for the large magnetoresistance values observed in Fe_3O_4 and LSMO point contacts.

5.2.2 Transport in point contacts

Since we can rule out magnetostriction as the main explanation for the large magnetoresistance values observed in point contacts, we have to turn our attention to the transport mechanisms in these contacts. To get a feeling for the numbers involved, Table 2 shows the values of the resistance of the contact, as calculated for three different transport mechanisms: ballistic transport, diffusive (or hopping) transport and tunneling.

Contact radius	ballistic	diffusive, hopping	tunneling
1 Å	94 kΩ	0.64 MΩ	3.2 GΩ
1 nm	940 Ω	64 kΩ	32 MΩ
10 nm	9.4 Ω	6.4 kΩ	320 kΩ
100 nm	94 mΩ	640 Ω	3.2 kΩ
1 μm	940 μΩ	64 Ω	32 Ω

Table 2. The resistance of a point contact, as function of its radius r , calculated for three different transport mechanisms. In the ballistic case, the Sharvin formula is used. For the diffusive case we assume a contact length $L = 4r$, and use the bulk resistivity of Fe_3O_4 , $\rho = 5 \text{ m}\Omega\text{cm}$. The tunnel resistance is determined in the low voltage regime, taking a tunnel barrier height = 4 eV and barrier width = 1 nm.

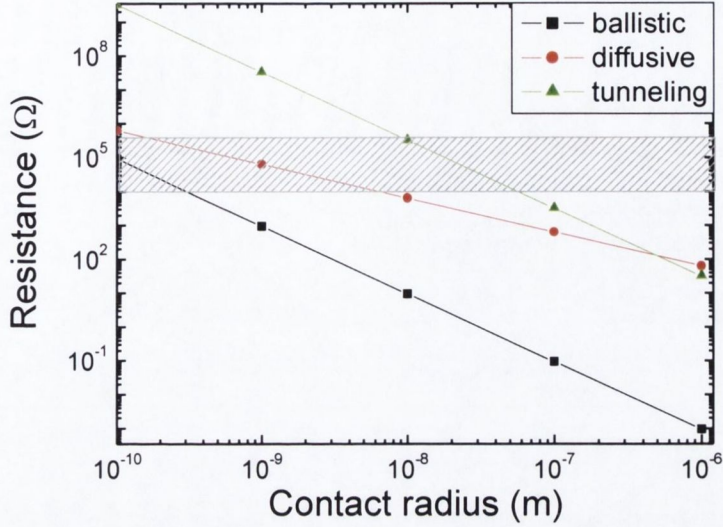


Figure 5.4. Resistance of a point contact as function of the radius of the contact, calculated for three different transport mechanisms. Details can be found in the text. The shaded area denotes the range of resistances found in our point contact measurements.

The ballistic resistance R_b is determined using the Sharvin formula [14]

$$R_b = \frac{h}{2e^2} \left(\frac{4\pi}{k_F^2 A} \right) = \frac{1}{G_0} \left(\frac{4\pi}{k_F^2 A} \right), \quad (5.2)$$

with A the area of the point contact, and $1/G_0 \simeq 12.9 \text{ k}\Omega$. An equivalent expression is $\frac{4\rho\lambda}{3A}$, where λ is the mean free path and ρ is the resistivity. In magnetite the electron density $n = 1.35 \times 10^{28} \text{ m}^{-3}$, and $k_F = (3\pi^2 n)^{1/3} = 7.4 \times 10^9 \text{ m}^{-1}$ ($\lambda_F = 0.85 \text{ nm}$). The diffusive (or hopping) resistance is simply determined by taking the bulk value of the resistivity, and assuming a contact length of four times the radius, $R_d = \frac{4\rho}{\pi r}$. The Simmons model [21] is often used to determine the barrier width and height from the current-voltage characteristics of well-defined tunnel junctions. Here we assume a barrier width (1 nm) and height (4 eV) to calculate the tunnel resistance. The tunnel resistance is strongly dependent on tunnel barrier height and width.

The values, even though they are rough estimates, do give an order-of-magnitude for

the resistance of the contact as function of its radius (Figure 5.4). The diffusive (or hopping) case seems to agree most with experimentally observed values.

5.2.2.1 Tunneling

Current-voltage characteristics Our experiments on $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ single crystals and Fe_3O_4 crystallites reveal a strong non-linear behaviour for the current-voltage characteristics. Similar non-linear behaviour has been observed in non-magnetic gold nanowires and magnetic Ni nanocontacts has been observed by Costa-Krämer *et al.* [16]. Following their work, we have fitted the measured current-voltage dependence to the following expression:

$$I(V) = I_{\text{linear}}(V) + I_{\text{non-linear}}(V) = cV + GV^3, \quad (5.3)$$

where I is the current, V the applied bias voltage, and c and G the fit parameters. Figure 5.5 compares the ratio c/G , which is a measure of the ratio of the linear and non-linear contributions, as a function of contact resistance for different materials. Except for Fe_3O_4 , all these materials show a strong signature of quantized conductance.

The non-linear current-voltage characteristics that we observe, however, can also be explained by tunneling. When the two crystallites are brought into contact, it is possible that there will be a thin layer of water, impurities or an otherwise insulating layer in between the crystallites. Transport could then be due to direct tunneling, from one crystallite, through the insulating layer, to the other crystallite, leading to non-linear current-voltage characteristics.

Another possibility could be contributions from indirect tunneling via one or more localised states in a barrier. The simplest process is resonant (elastic) tunneling, although inelastic processes could also contribute. When two crystals are put into contact, the in-

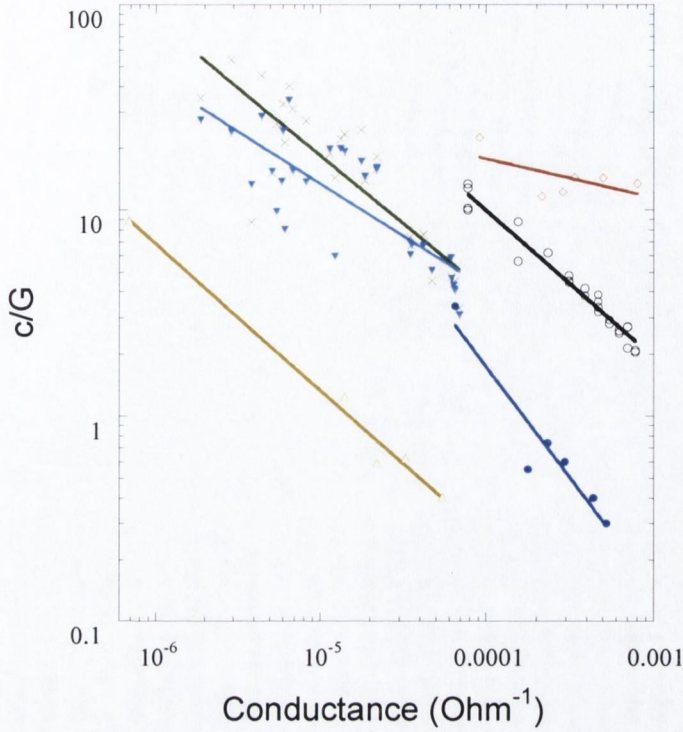


Figure 5.5. The ratio c/G as a function of the point contact conductance for a variety of materials. Shown are Fe_3O_4 with ($\blacktriangledown$) and without ($\times$) applied field of 7 mT, Au ($\triangle$) and ($\circ$) [27][16], LSMO ($\diamond$) [28], and Ni ($\bullet$) [29].

interface is likely to contain a disordered region with impurities and defects, something that is confirmed by theoretical calculation [18]. This region could act like a tunnel barrier with localised states. The conduction can then be expressed [19] as:

$$G_{tot} = G_{lin} + G_2(T, V) + G_3(T, V) + \dots, \quad (5.4)$$

where G_{lin} is the conduction due to direct tunneling and indirect tunneling via one localised state, and T and V the temperature and voltage respectively. Higher order terms are given by:

$$G_n(T, V) \propto V^{(n - \frac{2}{n+1})} \exp\left(\frac{-2\alpha d}{n+1}\right) \quad (eV \gg k_B T), \quad (5.5)$$

where α^{-1} is the radius of a localised state, and d the width of the tunnel barrier. As the

voltage is increased tunneling processes involving a higher number localised states become dominant.

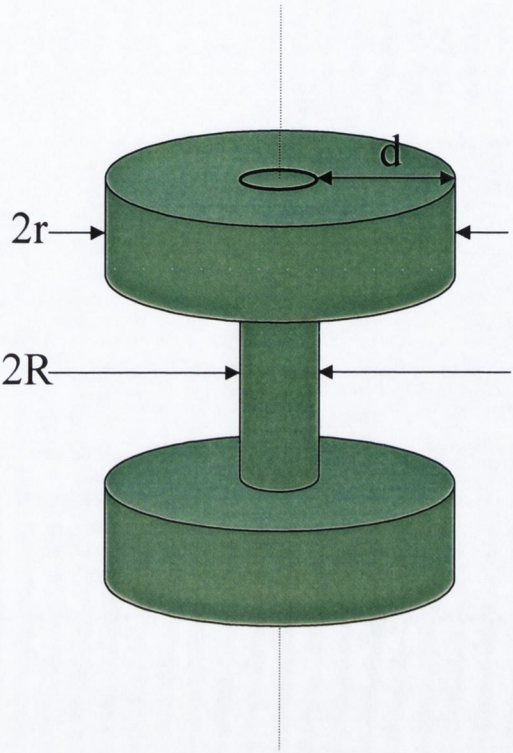


Figure 5.6. Model of a point contact. Ballistic transport takes place through the contact with radius R , while tunneling is possible in a ring with width $d = r - R$.

For magnetic tunnel junctions the picture becomes more complicated. Guinea [22] investigated the voltage dependence of the tunneling conductance due to the excitation of magnons, or spin waves. For bulk magnons, their creation leads to current $I(V)$ (at zero temperature) given by

$$I(V) \sim \frac{V}{R} \left(\frac{V}{J} \right)^{3/2} \quad V \ll J \frac{a^2}{d^2} \tag{5.6}$$

$$I(V) \sim \frac{V}{R} \left(\frac{a}{d} \right)^3 \quad V \gg J \frac{a^2}{d^2} \tag{5.7}$$

with J the exchange constant, d the barrier width, and a the lattice constant.

However, we don't find evidence for either tunneling through localized states, or for

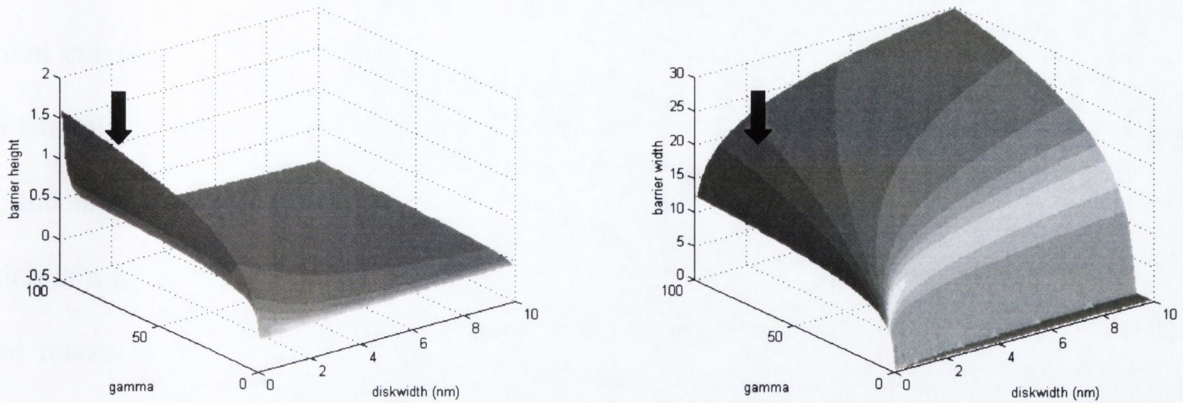


Figure 5.7. Barrier height (in eV) and width (in Å) as a function of both the width of the ring available for tunneling, and of gamma, which is a measure for the non-linearity of the IV curves.

magnon excitation when we analyze our current-voltage characteristics. The curves are fitted better with an $I = GV + cV^3$ law than with a $V^{4/3}$ or $V^{5/2}$ term.

Finally, an explanation could also lie in the existence of two parallel conduction mechanisms; ballistic transport and tunneling. This could be the case for LSMO, where quantized conductance is observed. If we consider a small nanocontact, it is reasonable to assume that the regions on both sides around the nanocontact are close enough to each other to contribute via tunneling to the total charge transport. To further investigate this possibility we have performed some simulations on the following model. Consider a curvilinear constriction as a nanocontact with a certain quantized conductance $G = nG_0$. Its radius, R , can be estimated from the semi-classical expression for a ballistic wire [20]:

$$n \approx \left(\frac{\pi R}{\lambda_F} \right)^2 \left(1 - \frac{\lambda_F}{\pi R} \right). \quad (5.8)$$

We now assume that only a ring with a thickness, $d = r - R$, is available for tunneling, giving a total tunnel area, $A = \pi(r^2 - R^2) = \pi(d^2 + 2dr)$. From our measured IV curves we take the low voltage data, and subtract the contribution due to ballistic transport, i.e.

$I_{ballistic} = nG_0V$, leaving the tunnel current contribution, $I_{tunneling}$. Converting this into a tunnel current density, $J = \frac{I_{tunneling}}{A}$, we can now use Simmons' model [15] to obtain both the tunneling barrier height (in eV) and width (in Å). In Figure 5.7 we show the results of simulations: barrier height and width as a function of both ring thickness d , and gamma, which is a measure of the non-linearity of the IV curves. For this model to be plausible we need reasonable values for both barrier width and height. Barrier height needs to be well above room temperature, while the barrier width has to be of the order of the nanocontact radius R . We see that for small values of d ($d \lesssim 1$ nm), these conditions are indeed met, as is shown in Figure 5.7.

Magnetoresistance We now turn to the magnetoresistance of our point contacts, and relate it to the concept of magnetoresistance in tunnel junctions. When a tunnel junction consists of two ferromagnetic materials, separated by a tunnel barrier, the tunnel conductance is also determined by the relative orientation of the magnetization in the two electrodes. Julliere [23] showed that the conductance in the parallel and antiparallel case depends on the spin-dependent density of states $n_{\uparrow,\downarrow}$

$$G_{\uparrow\uparrow} \propto n_{\uparrow}^1 n_{\uparrow}^2 + n_{\downarrow}^1 n_{\downarrow}^2 \quad (5.9)$$

$$G_{\uparrow\downarrow} \propto n_{\uparrow}^1 n_{\downarrow}^2 + n_{\downarrow}^1 n_{\uparrow}^2, \quad (5.10)$$

where the subscripts denote electrode 1 or 2. The tunnel magnetoresistance TMR is then

$$TMR = \frac{G_{\uparrow\uparrow} - G_{\uparrow\downarrow}}{G_{\uparrow\uparrow}} = \frac{2P_1 P_2}{1 + P_1 P_2}, \quad (5.11)$$

where the spin polarization P is defined as

$$P = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}}. \quad (5.12)$$

For a tunnel junction made of two ferromagnets with a spin polarization of the same sign, the tunnel magnetoresistance TMR is always negative. For half-metals (with $P = 100\%$), the TMR reaches its maximum value of 100% . Guinea [22] showed that the presence of magnetic impurities in the barrier will increase the tunnel resistance, but reduce the magnetoresistance. From the maximum of MR of 85% in magnetite we infer a room temperature polarization of 86% .

It is experimentally found that the tunneling magnetoresistance is dependent on bias voltage, with TMR decreasing for increasing voltage [24]. The voltage dependence of the effective barrier height is too small to explain the observed behaviour [25]. For high bias voltages, TMR is reduced due to hot electrons, leading to a linear dependence of TMR on bias voltage for voltages < 200 mV. A stronger voltage dependence is observed [24], which was ascribed to magnon scattering. This is predicted to lead to a $V^{3/2}$ behaviour [22].

Our measurements show a strong dependence of magnetoresistance on contact resistance, with the highest values of MR found for contacts with the highest resistance. If we assume that the contact consists of a tiny tunnel barrier, the resistance-dependent behaviour of the magnetoresistance could then be explained in terms of the presence or absence of impurities in the barrier. For the smallest contacts (those with highest resistance), the chance of impurities being present becomes small. For those 'clean' magnetite contacts, the magnetoresistance of 85% approaches the theoretical maximum. Since some of the smallest contacts, though, will contain impurities, a wide range of magnetoresistance values is found. Larger contacts, with smaller contact resistances, will have a strongly reduced magnetoresistance.

We also observe a voltage dependent magnetoresistance, which, for Fe_3O_4 , goes from

85 % at $V = 0$, to about 65 % for $V = 0.3$ V. The data in Figure 4.39 shows a linear dependence up to 0.3 V. For LSMO this dependence is smaller, from 45 % ($V = 0$) to about 37 % ($V = 0.3$ V) (Figure 4.18). In both cases, because of the spread of data points, it is hard to determine the exact nature of this dependence, but it seems linear.

Magnetic tunnel junctions show an increase of TMR with decreasing temperature [24]. The absence of magnetoresistance that we observe in point contacts at lower temperature cannot readily be explained, and more work is needed to clarify if this is indeed the case.

5.2.2.2 Hopping

Transport through a domain wall in a (conducting) contact can be either ballistic or diffusive, depending on whether the mean free path λ is larger or smaller than the domain wall width δ_w . This is shown schematically in Figure 5.8. In the diffusive case, when $\lambda \ll \delta_w$, it has been shown by Levy and Zhang [26], that the magnetoresistance MR varies as $1/\delta_w^2$. In magnetite, transport takes place through a hopping mechanism. Minority spin carriers hop from one B-site to another as they move through the material. Each hop can be seen as an internal tunneling process, with corresponding transmission T . If the mean hopping distance λ_{hop} is larger than the domain wall width δ_w , the carriers will hop from one side of the contact to the other side, 'tunneling' through the wall. In that case, the transmission is only determined by the misalignment of the magnetization on either side of the contact:

$$T \sim \cos^2 \left(\frac{\theta_{12}}{2} \right), \tag{5.13}$$

with θ_{12} the angle between the two magnetization directions. Full transmission is achieved when the magnetizations are parallel (i.e. $\theta_{12} = 0$), and no transmission when they are anti-parallel ($\theta_{12} = \pi$).

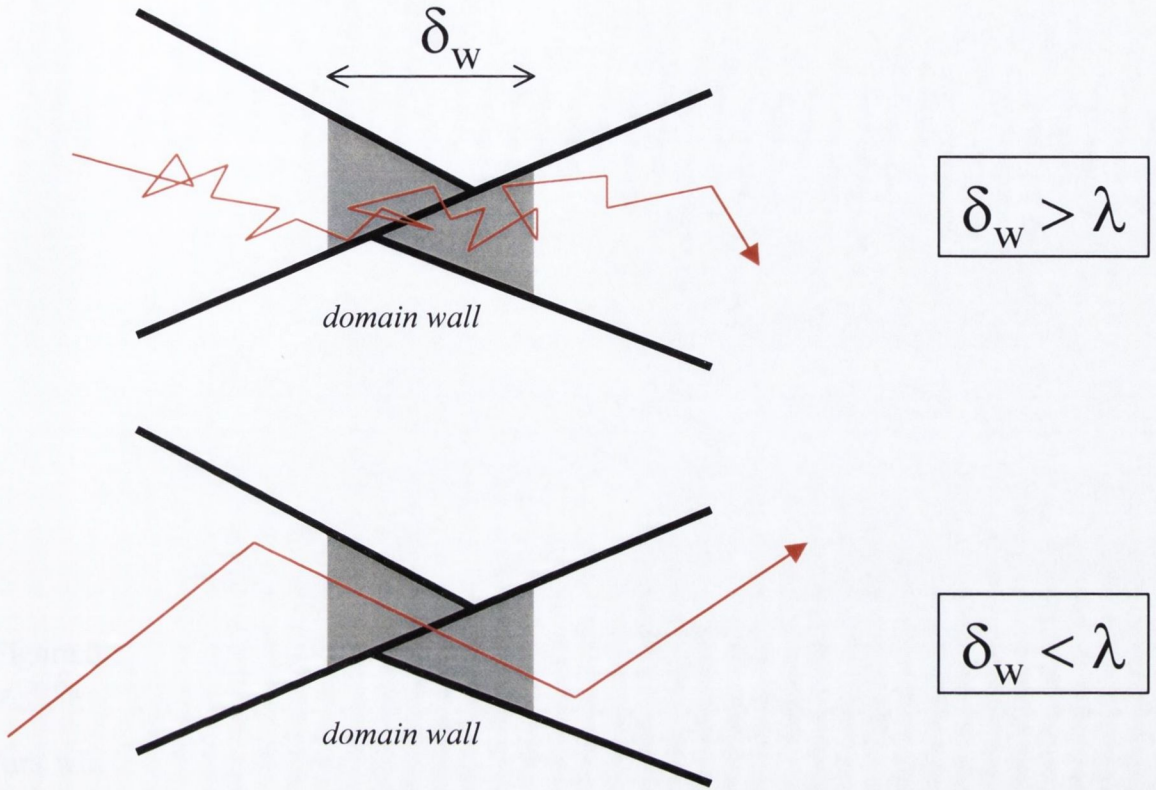


Figure 5.8. Diffusive (top) and ballistic (bottom) transport through a domain wall, with width δ_w , in a conducting point contact. The mean free path is denoted by λ .

In the other limit, when $\lambda_{hop} \ll \delta_w$, and the carriers who traverse the wall sample many sites inside it, the transmission T can be derived as follows. Assume that the carriers make $v = \delta_w / \lambda_{hop}$ hops inside a 180° wall. Each hop has a probability $T \sim \cos^2(\pi/2v)$, and so a total transmission of

$$T \sim \left[\cos^2 \left(\frac{\pi}{2v} \right) \right]^v. \quad (5.14)$$

For large values of v , the transmission T approaches unity.

Recall that the magnetoresistance is defined as $MR = -\frac{G(H)-G(0)}{G(H)}$, and that the conductance G of the contact is proportional to the total transmission, $G \propto T$. When the applied field is large enough, so that the magnetization on both sides of the contact are aligned, the magnetoresistance then reduces to $MR = 1 - T$. The maximum MR of 100 %

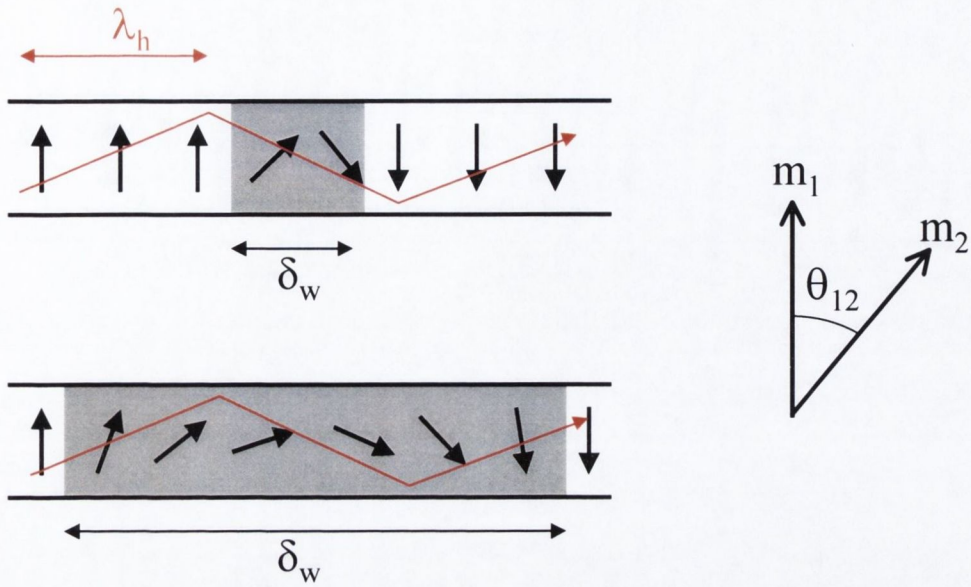


Figure 5.9. Hopping transport in through a domain wall. On top, transport in the limit $\lambda_{hop} > \delta_w$, at the bottom $\lambda_{hop} < \delta_w$.

occurs when the magnetizations are antiparallel, and $\lambda_{hop} > \delta_w$, and reduces rapidly with increasing v . In other words, the maximum magnetoresistance is found when the domain wall width is small compared to hopping distance.

In case the spin polarization P is less than a 100 %, due to the temperature dependence of the magnetization, or a particular (surface) band structure, the maximum magnetoresistance is given by the Julliere model [23], equation (5.11)

5.2.3 Spin Pressure

The large room temperature MR observed in some of the magnetite and LSMO point contacts is consistent with atomic-scale contact between crystallites with a narrow domain wall there. The field effect on the I:V curves of point contacts, illustrated in Fig. 4.32 and Fig. 4.34 is not simply to open the tunnel barrier at the contact to increased transmission by aligning the moments on either side of it. If that were the case, the ratio c/G would remain constant. In fact, c/G decreases by about a factor 2 for low conductance magnetite contacts

in 7 mT (Figure 5.5). This effect may be understood if the electric current exerts a pressure on the wall, tending to push it away from the nanocontact. Since the domain wall behaves like an elastic membrane with variable surface tension, depending on its position, we call this the 'magnetic balloon effect'. It is illustrated in Figure 5.10. Part of the nonlinear upturn in

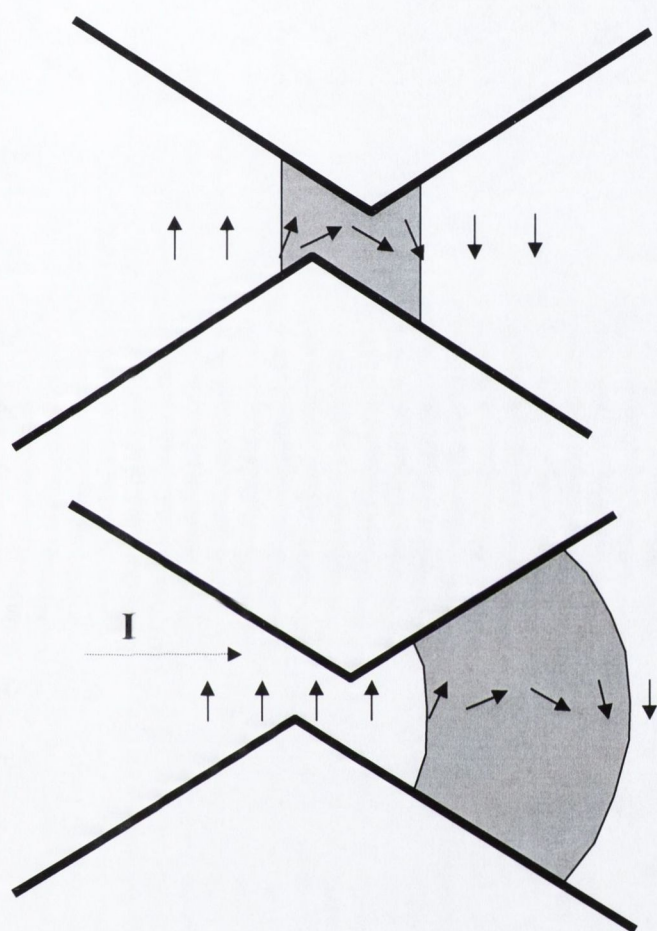


Figure 5.10. Illustration of the 'magnetic balloon' effect. A narrow domain wall, pinned by the constriction (top), is pushed out by the resulting pressure caused by an electrical current I .

the zero-field I:V curves may be attributed to current-induced displacement of the domain wall away from the contact. In the current-voltage characteristics of our point contacts we don't find any evidence for a threshold current for this effect.

Threshold pressure To see if this idea is feasible, we first need to have an idea of what

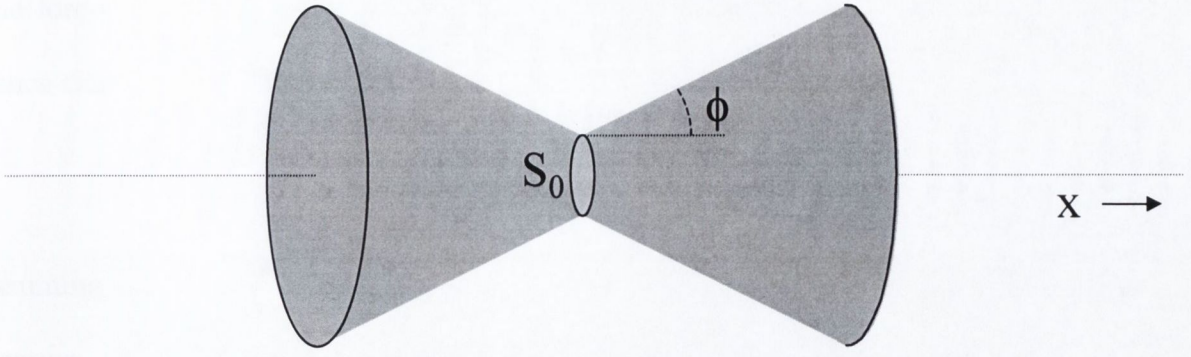


Figure 5.11. Sketch of the point contact geometry. The contact is modelled as two truncated cones with half opening angle ϕ , and common surface area S_0 .

pressures are needed to move the domain wall away from the constriction. We model the contact as two truncated cones with half angle ϕ and a common surface of area S_0 (See Figure 5.11). The energy density γ for a domain wall in bulk is given by

$$\gamma = 4\sqrt{AK}, \quad (5.15)$$

with A the exchange stiffness and K the anisotropy constant. In contrast, a domain wall pinned by a nanometric constriction (as shown in the top of Figure 5.10), will have a wall energy density γ_w given by

$$\gamma_w \approx \frac{\pi^2 A}{\delta_w}, \quad (5.16)$$

where δ_w is the domain wall width. We can use this expression to derive an expression for the threshold pressure needed to displace the wall. Assuming the wall width increases linearly with displacement x ,

$$\delta_w = \delta_0 + 2x \tan \phi \quad (5.17)$$

the wall energy E_w is then

$$E_w = \gamma_w S = \left[\frac{\pi^2 A}{\delta_w} \right] \cdot \pi \left(\frac{\delta_w}{2} \right)^2 = \frac{\pi^3 A}{4} [\delta_0 + 2x \tan \phi]. \quad (5.18)$$

The force F required to move the domain wall from its stable position is $-dE/dx \mid_{x=0}$.

Hence the threshold pressure is

$$P_t = \frac{F}{S_0} = \frac{\pi^3 A \delta_0^2}{4 \delta_0^2} \frac{2 \tan \phi}{S_0} = \frac{\pi^3 A \tan \phi}{2 S_0}. \quad (5.19)$$

Assuming $\tan \phi \approx 1$, $S_0 = 1 \text{ (nm)}^2$ and $A = 10^{-11} \text{ J m}^{-1}$, we can now estimate the threshold pressure, $P_t \approx 2 \times 10^8 \text{ N m}^{-2}$.

The question is now if there are physical processes capable of displacing a domain wall from the constriction, or in other words, what could produce pressures greater than the threshold pressure P_t calculated above? Below we will discuss various mechanisms, and attempt to quantify them.

Momentum pressure A first idea would be simple momentum scattering of minority spin electrons impinging on the domain wall. This is analogous to the electron wind effect in electromigration [30]. Large current densities can be obtained at the constriction, for example, a $10 \text{ }\mu\text{A}$ current through a contact of cross section 1 nm^2 corresponds to current density $j = 10^{13} \text{ A m}^{-2}$. The current density j can be related to the drift velocity v_d :

$$j = nev_d, \quad (5.20)$$

with n the electron density and e the charge of the electron. For Fe_3O_4 , with $n = 1.35 \times 10^{28} \text{ m}^{-3}$, this gives a velocity $v_d = 4.6 \times 10^3 \text{ m s}^{-1}$. An electron, with mass m_e , impinging on the wall and reflected by it undergoes a change of momentum $2m_e v_d$. The total force F exerted by the momentum scattering on the domain wall is simply the the total change of momentum per second:

$$F = \frac{I}{e} 2m_e v_d, \quad (5.21)$$

resulting in a pressure $P_m = F/S_0 = 5.2 \times 10^5 \text{ N m}^{-2}$. This is too small, by almost three orders of magnitude to displace the wall.

It can be argued that this underestimates the pressure due to momentum scattering. Electrons, with transmission T , will make $1/T$ attempts before successfully crossing the barrier. Taking this into account, the rate of change of momentum per unit area, or pressure should read

$$P_m = \frac{1 - T}{T} \frac{I}{e} \frac{2mv_d}{S_0}. \tag{5.22}$$

The transmission T can be estimated as follows. When the moments on either side of the contacts are incompletely aligned with the field, the Juliere formula yields

$$MR = \frac{2m^2}{1 + m^2}, \tag{5.23}$$

with m the reduced magnetization at room temperature. From Figure 4.27 we find $m = 0.95$. Since the conductance $G \propto T$, and therefore $MR = 1 - T$, we find $T \approx 0.05$, leading to $P_m = 1 \times 10^7 \text{ N m}^{-2}$. Again, this is too small a pressure to displace the domain wall from its stable position.

Angular momentum pressure Another mechanism envisaged is that associated with the electrons' angular momentum. An electron, coming from one side of the contact with magnetization up $\uparrow$, will have to change its angular momentum when traversing the domain wall to the other side of the contact, with magnetization down $\downarrow$. The rate of change of angular momentum produces a torque Γ , which we suppose translates into a force on the wall. Since each electron has angular momentum $\hbar/2$, the corresponding torque $\Gamma = \frac{jA\hbar}{2e}$, where

A is the cross section area of the wall. The pressure is $\Gamma/A\delta_w$ so

$$P_{am} = \frac{j\hbar}{2e\delta_w}. \quad (5.24)$$

If $j = 10^{13} \text{ A m}^{-2}$ and $\delta_w = 1 \text{ nm}$, then $P_{am} = 3 \times 10^6 \text{ N m}^{-2}$, which is too small to displace the wall.

Exchange drag pressure The exchange interaction between a domain wall and an electrical current was discussed theoretically by Berger[31] for the case of wide domain walls, and later the effect was observed in thin $\text{Ni}_{87}\text{Fe}_{13}$ films [32]. Berger used the adiabatic approximation where the spin of the electron essentially follows the rotation of magnetization in the wall; the broad domain wall does not reflect the electrons appreciably, but spin flip scattering (by impurities or phonons) does lead to force exerted on the wall.

His result (equation (2.11)) is that the force per unit area

$$P_{ed} \simeq \frac{2J_s}{\mu} v_e, \quad (5.25)$$

where $J_s \simeq 1 \text{ T}$ is the polarization of the ferromagnetic material, v_e is the electron drift velocity ($v_e = j/ne$) and μ is the domain wall mobility, which is typically $1 \text{ m}^2 \text{ C}^{-1}$ in normal ferromagnetic thin films. In our case, with a current density of 10^{13} A m^{-2} , $v_e \approx 5 \times 10^3 \text{ m s}^{-1}$, hence $P_{ed} \simeq 10^4 \text{ N m}^{-2}$. The problem with Berger's argument is the adiabatic assumption, which is not valid for very narrow walls such as we postulate.

We estimate the exchange drag pressure for the case of hopping electrons where $\lambda_{hop} > \delta_w$, i.e. for narrow domain walls. An electron hopping from one B-site to another will experience an increase in energy if the local moments of those atomic cores are misaligned. If J_{dd} is the exchange coupling of the electron with the atomic core, and θ the misalignment between the two cores, the energy increase is $J_{dd} \sin^2(\theta/2)$. The total change in energy per

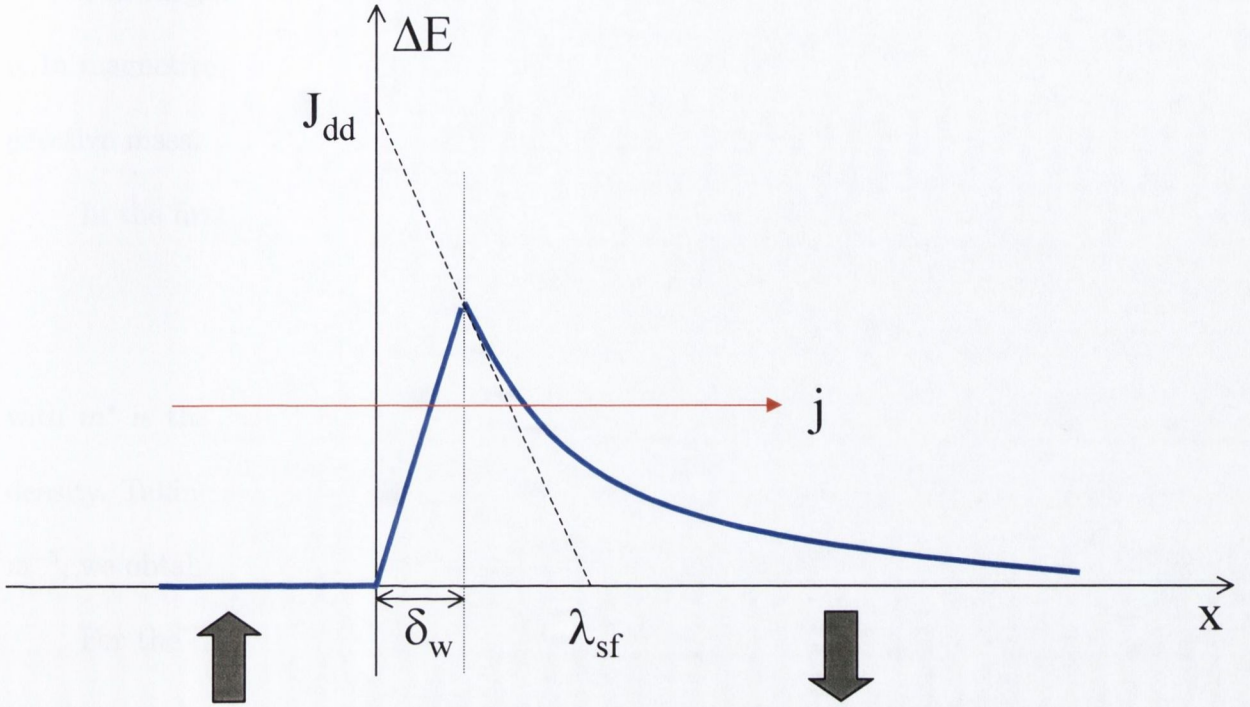


Figure 5.12. The excess energy due to the passage of a current of $\uparrow$ electrons through the domain wall at $0 < x < \delta_w$. The spin accumulation in high energy states on the right hand side is dissipated over a length scale λ_{sf} .

second is then $(I/e)J_{dd} \sin^2(\theta/2)$. The pressure on the wall arises from the excess energy when a current passes through it, as illustrated in Fig 5.12. The typical timescale for this process is the spin flip scattering time τ_{sf} . The flow of electrons across the wall is I/e per second, and if the spin flip time is τ_{sf} , the excess energy associated with a 180° wall is $(I/e)\tau_{sf}J_{dd}$. The expression for the pressure on the 180° wall is then

$$P_{ed} \simeq \frac{I}{e} \frac{\tau_{sf}}{S\lambda_{sf}} J_{dd} = \frac{j}{e} \frac{\tau_{sf}}{\lambda_{sf}} J_{dd}. \quad (5.26)$$

Now $\lambda_{sf} = \lambda_{hop} \sqrt{n_{hop}}$ and the spin flip scattering time $\tau_{sf} = \lambda_{hop} n_{hop} / v$, with λ_{hop} the hopping length, n_{hop} the number of hops before the spin is flipped, and v the conduction electron velocity. This leads to a new expression for the pressure

$$P_{ed} = \frac{j}{e} \frac{\sqrt{n_{hop}}}{v} J_{dd}. \quad (5.27)$$

The magnitude of the exchange drag pressure P_{ed} depends on which value we take for v . In magnetite, the current is carried by polarons, which we can view as electrons with an effective mass, or we can interpret polarons as hopping electrons.

In the first case, we can estimate v using the free electron model:

$$v = v_F = \frac{\hbar k_F}{m^*} = \frac{\hbar}{m^*} \sqrt{3\pi^2 n}, \quad (5.28)$$

with m^* is the effective mass of a polaron, k_F the Fermi wave vector and n the electron density. Taking an effective mass of three times the electron mass, $m^* = 3m_e$, $n = 1.35 \times 10^{28} \text{ m}^{-3}$, we obtain $v = 2.8 \times 10^5 \text{ m s}^{-1}$.

For the second case, we calculate the hopping frequency $v_{ph} = 1/\tau_{ph}$. At room temperature we have $\hbar v_{ph} = k_B T$, giving $v_{ph} = k_B T / \hbar = 6.7 \times 10^{12} \text{ s}^{-1}$. The hopping velocity v_{hop} can now be obtained; $v_{hop} = \lambda_{hop} / \tau_{ph}$, which, when we take the hopping mean free path as the interatomic distance, gives $v_{hop} = 2 \times 10^3 \text{ m s}^{-1}$.

We can now estimate the exchange drag pressure by taking $j = 10^{13} \text{ A m}^{-2}$, $J_{dd} = 0.1 \text{ eV}$. and $\sqrt{n_{hop}} = 10$, giving $P_{ed} \simeq 3.5 \times 10^7 \text{ N m}^{-2}$ for the first case, and $P_{ed} \simeq 5 \times 10^9 \text{ N m}^{-2}$ for the second. Only in the case of hopping polarons, is P_{ed} larger than the threshold pressure P_t . Transport in magnetite takes place via two conduction processes; a polaronic band conductivity together with a polaronic hopping conductivity [33], and it therefore seems that, in this case, exchange drag pressure is capable of moving the domain wall out of its equilibrium position.

The idea of exchange drag pressure on a narrow domain wall can explain the observed magnetoresistance and current-voltage characteristics of the point contacts. The presence of a wall pinned by a constriction such as a point contact leads to a higher resistance of the contact. Application of a field will align the magnetization on both sides of the

contact, and the wall is eliminated, resulting in a much lower resistance. The hysteresis in the magnetoresistance shows that the effect is indeed related to the magnetization in and around the contact.

The non-linear current-voltage characteristics are caused by the spin pressure of spin polarized electrons pushing the wall away from the contact. By doing so, the wall widens and it becomes less resistive, leading to the non-Ohmic behaviour observed in the contacts.

Why is the magnetoresistive effect present at room temperature, but absent at lower temperatures? At least for Fe_3O_4 , it can be argued that, at 77 K, hopping transport is greatly impeded since this is below the Verwey transition. But that leaves the question open why no magnetoresistance is observed at 150 K. We think this has something to do with the way the contacts are formed in our experimental set-up. At room temperature, we can completely break the contact and then make a highly conducting one before starting to slowly break it again. This way, we can be sure to always have a completely new contact to measure. As stated in the previous chapter, we often need to "train" the contact to observe an appreciable magnetoresistance. Even so, not all contacts show the effect, and only by taking many different contacts into consideration can we say something in general about the behaviour of the contacts.

At lower temperatures, the piezo tube loses most of its travel length, and it may therefore not be possible anymore to completely make and break the contact in between measurements. As a consequence, at these low temperatures, we don't measure many different contacts, but the same one over and over.

5.2.4 Future work

An application of the exchange drag pressure idea could lie in switching devices as

shown in Figure 5.13. Here, three ferromagnetic electrodes, or terminals, have their magnetization fixed in the directions shown. This could be achieved using antiferromagnetic pinning layers. The central island, separated from the terminals by constrictions (here denoted by A, B and C), has a magnetization that is free to change direction. Depending on whether the magnetization is up or down, there will be domain walls pinned in constrictions B and C, or in A. The state of the switch can be read by measuring the resistance of the constrictions B and C. If these constrictions contain domain walls, the resistance will be high, if not, it will be low. The device can be switched by flipping the magnetization of the island. This can be done by applying a large enough current pulse in the right direction to push the wall out of a constriction, and flip the magnetization of the island. Switching can be very fast, since the wall moves only about 10 nm, at a velocity of 100 m s^{-1} [35].

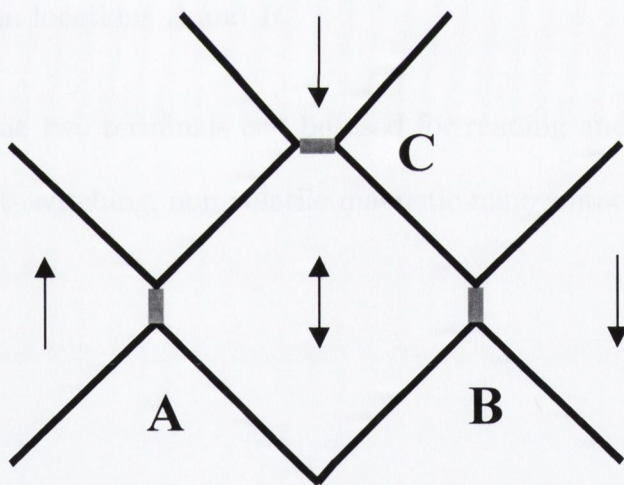


Figure 5.13. Three terminal magnetic switch based on current-induced toggling of a domain wall between two positions A and B. Readout is obtained by sensing the resistance across C.

An even simpler idea is the 'peanut device'[34], illustrated in Figure 5.14. This is a two terminal device, with two constrictions with the one (A) being narrower than the other (B). The two electrodes have their magnetizations pinned in opposite directions, meaning that

there will be a domain wall at either A or B. Since a wall at A will be narrower, the device in this state will have a high resistance, whereas a wider wall at B leads to a low resistance state. By measuring the resistance between the two terminals, the state of the device can be determined. Again, the device can be written by applying a large enough current pulse in the right direction, pushing the wall from A to B, and vice versa. This device has the

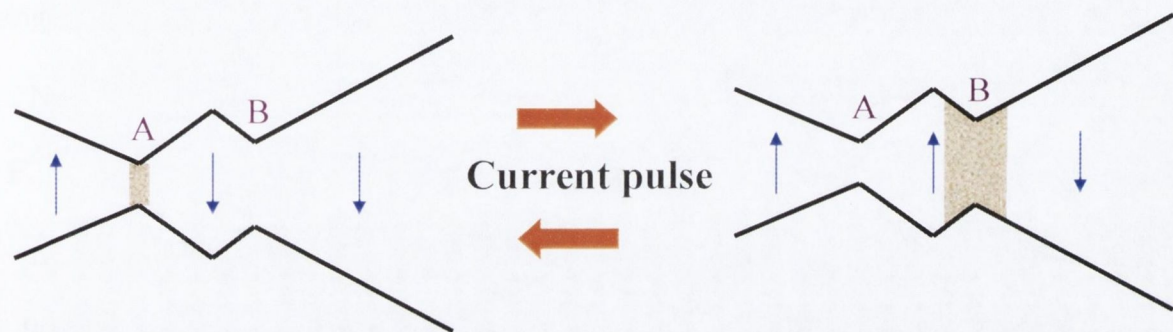


Figure 5.14. Two terminal switch, the 'peanut device'. The resistance of the device depends on the location of the domain wall. A current pulse in the appropriate direction toggles the wall between locations A and B.

advantage that the same two terminals can be used for reading and writing, and could be the basis for a new, fast-switching, non-volatile magnetic nanocontact memory architecture.

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Chapter 6

Conclusion

The study presented in this thesis has been concerned with mesoscopic transport in ferromagnetic oxides. Two techniques have been developed to gain insight into the extrinsic transport properties of these materials.

The first, scanning tunneling potentiometry (STP), was used to correlate topographic features of a thin film sample with the flow of current through the sample. Here, a Scanning Tunneling Microscope (STM) was modified successfully to simultaneously image the surface topography and map the potential distribution in a polycrystalline $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ thin film. The technique, together with required electronics and software has been described in some detail. Large voltage drops were observed to coincide with grain boundaries, whereas in the grains themselves essentially no change in voltage was found. From the voltage drop associated with the grain boundaries, an estimate of the resistivity of these grain boundaries can be calculated. It is found that it ranges from $3 \times 10^{-7} \Omega \text{ cm}^2$ to $3 \times 10^{-5} \Omega \text{ cm}^2$, with an average value of $6 \times 10^{-6} \Omega \text{ cm}^2$, which is comparable with other reports in the literature. It is concluded that for polycrystalline $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ thin films the resistance is mainly due to the grain boundaries.

The second method described in this thesis is the study of magneto-transport properties in point contacts. Two tiny single crystals of either magnetite (Fe_3O_4) or $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ are mounted in a device where their separation can be controlled using a piezo-electric actuator. The two crystallites are brought into contact, and the resistance of the contact between them is monitored while they are being pulled apart again. Using simple vibration

insulation ensures that the contacts are stable for up to a minute or more, which is long enough to obtain current-voltage characteristics and measure the magnetoresistance. The hysteresis of the magnetoresistance as a function of applied field establishes that the effect of the magnetic field is to modify the micromagnetic configuration of the contact.

$(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ shows clear conductance quantization when the contacts are broken rapidly ($100\ \mu\text{s}$), with peaks up to $7G_0$ present in the conductance histogram of 60 breaking contacts, but when broken slowly the contact can be stabilized at any value of conductance. Fe_3O_4 only shows a weak peak near $1G_0$, which is suppressed in a field. the difference is associated with the different conduction mechanism; metallic in $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$, hopping in Fe_3O_4 . For both materials, contacts with conductances smaller than G_0 , could be stabilized. Strong non-linear current-voltage characteristics are observed, which were fitted to an $I = GV + cV^3$ law. It is found that the non-linearity of the current-voltage characteristics increases for the more resistive contacts. Measurements at 150 and 77 K show that the non-linearity is also reduced for lower temperatures. Room temperature magnetoresistance (MR) values of up to 85 % for Fe_3O_4 and up to 45 % for $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ have been found for contacts with a low zero-bias conductance ($G \ll G_0$), in fields of 7 mT. For contacts with higher values of conductance the magnetoresistance falls off quickly. No magnetoresistance was observed in contacts at 150 and 77 K.

We find that to obtain large MR values contacts need to be 'trained', that is, sweeping the field many times can increase the magnetoresistance. The small magnetic hysteresis shows that the large magnetoresistance effect is related to the magnetization process of the crystallites.

The huge magnetoresistance values we find in magnetite point contacts at room tem-

perature and in small fields are larger than anything reported previously for ferromagnetic break junctions, including ballistic Ni nanocontacts, or tunnel junctions. We were able to rule out magnetostriction as the main explanation for these large effects, and advance two possible explanations; tunneling and spin pressure on a narrow domain wall pinned at the contact.

Seeing the contact as a magnetic tunnel junction can explain the dependence of the magnetoresistance on contact conductance. Assuming that the smallest contacts, with a low conductance value, contain fewer impurities in the insulating barrier, the observed maximum magnetoresistance value of 85 % for magnetite approaches the theoretical maximum of a 100 %. The bias dependence of the magnetoresistance found, can then be explained by magnon creation in the electrodes.

The other explanation, is based on the idea that a domain may be pinned at the contact. In contrast to the bulk, the width of the domain wall will then be small, of the order of the size of the constriction. For the current flowing through the contact, this narrow domain wall acts as an additional resistance. Application of the field moves the domain wall away from the contact, where it becomes broader (the magnetic balloon effect), leading to the large magnetoresistance observed. Interactions between electrons traversing the contact and the wall produce the force, or pressure, on the wall. This spin pressure on the wall causes it to be pushed away from the constriction, out into the electrode, and thereby widening it, leading to bias-dependent resistance of the contact. Order of magnitude calculations, modelling the contact as two truncated cones, shows that the spin pressure associated with the exchange coupling of the hopping electrons with the ion cores, is larger than the required minimum pressure to move the domain wall from its equilibrium. The domain wall will be

pushed out by a few nanometers.

At lower temperature no magnetoresistance is observed in either $(\text{La}_{0.7}\text{Sr}_{0.3})\text{MnO}_3$ or Fe_3O_4 . The reason for this are not fully understood, and more work is needed to investigate the temperature dependence of the MR. Both mechanisms, tunneling and spin pressure on a narrow domain wall, should lead to a magnetoresistance at lower temperature. Possible explanations for the absence of magnetoresistance in these contacts are the reduced travel length of the piezo crystal at lower temperatures, thereby making it impossible to create 'fresh' contacts, or the presence of a thin layer of ice on the crystallites, making good electrical contact more difficult.

The spin pressure idea may be exploited in a fast, current-activated magnetic switch, for use in non-volatile nanocontact memory devices, where the reading and writing of the state of the device requires only two terminals. The operation of this device depends on the magnetization reversal in the central region of the peanut.