PHYSICAL REVIEW B **66**, 220401(R) (2002)

# Magnetoresistance through a single nickel atom

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(Received 7 May 2002; published 12 December 2002)

The magnetoresistance (MR) of a nickel atomic contact has been measured using the break junction technique. When the contact is only between two atoms, the change of resistance with applied field reaches 40%. It is composed of a continuous bell-shaped curve on which discrete jumps are superimposed. The MR changes sign when the applied field is rotated, which we explain by a spin-orbit coupling change of orbital overlap between the Ni atoms forming the junction. Reproducible jumps in the MR curve are attributed to a field induced change of spin configuration within the few atoms composing the contact.

DOI: 10.1103/PhysRevB.66.220401 PACS number(s): 75.70.Kw

The effect of an external field on the resistivity of pure ferromagnetic metals (the magnetoresistance-MR) was the subject of intense research work in the middle of the 20th century. In homogeneous $3d$ elements, the internal field can affect the motion of the charge carriers (via the Lorentz force) as well as their scattering. In ferromagnets, part of the scattering is due to $3d$ localized atomic orbitals which are affected by a magnetic field through the spin-orbit mechanism. The resulting resistive effect is called anisotropic magnetoresistance (AMR) because it depends on the angle between the local moments and the electrical current lines. Charge carriers at the Fermi level are also composed of spin-up and spin-down electrons which provide two independent conduction channels with different resistivities: this is the two current model.[1] Any inhomogeneity in the local magnetization can induce some spin mixing which increases the total resistance. In bulk 3d metals at low fields (say below 2 T) the MR is typically of a few percent.

In the past fifteen years there has been a renewed interest in MR with the discovery of giant effects in systems composed of mixtures of magnetic and nonmagnetic materials. The giant-MR (GMR) effect results from the spin-dependent scattering of conduction electrons experiencing abrupt changes in the direction of the local magnetization vector. This is possible when nonmagnetic regions are smaller than the spin diffusion length, i.e., typically a few tens of nanometers. In unsaturated homogeneous $3d$ materials, the magnetization changes direction within the domain walls (DW) where the progressive canting of the local moments is too gradual for the conduction electrons to experience a reasonable potential of magnetic origin.[2] Moreover, the transverse component of the magnetization helps the electrons spins to track the local magnetization vector.[3] In order for the electron-spin mistracking to generate a reasonable MR effect, the magnetization would have to rotate significantly at the scale of the Larmor precession of charge carriers, i.e., a few nm.[3] In the limit of an infinitely narrow DW one recovers the GMR effect.

When sample dimensions are reduced below the mean-free-path, electronic transport can no longer be described by Boltzman statistics and one enters into the ballistic regime. In constrictions of the scale of the Fermi wave vector, boundary conditions enforce that transverse modes are quantized which results in the discreteness of propagating electron modes. In the Landauer formalism, the conductance of a mesoscopic contact is expressed as a function of the conductance quantum in the following manner :

$$G=\frac{e^2}{h}\left(\sum_1^{N_\uparrow} T_{i\uparrow}+\sum_1^{N_\downarrow} T_{i\downarrow}\right) \tag{1}$$

with $T_{i\uparrow}$ and $T_{i\downarrow}$ the transmission probabilities for each conduction channel $i$ with spin up or down and $e^2/h = 1/(26\ \mathrm{k}\Omega)$. The number of propagating modes depends on the constriction width $w$ and the Fermi energy of the relevant spin channel $E_{F_{\uparrow,\downarrow}}$ :

$$N_{\uparrow,\downarrow}=\frac{2w}{h}\sqrt{2mE_{F_{\uparrow,\downarrow}}} \tag{2}$$

Measurements in 2D electron gases (with Fermi wavelength around 40 nm) show conductance quantization in units of $2e^2/h$ because at zero field, the two spin species are degenerate and the Fermi surface is simple enough so that the transmission coefficients are close to unity. When a strong magnetic field is applied, spin degeneracy is removed and quantization in steps of $e^2/h$ is observed.[4]

In metals, because the Fermi wavelength is of the order of 2 Å, conductance quantization can only occur in wires of atomic size where conduction channels result from the overlap of the atomic orbitals. For a single atomic contact, the number of conduction channels cannot exceed the number of valence orbitals, but, as a rule of thumb, it is close to the number of valence electrons.[5] In transition metals, *s*, *p*, and *d* orbitals can potentially open $2+6+10$ channels. Ni is in the $4s^2\ 3d^8$ configuration and so should have around 4 spin down and 6 spin up channels whose transmittivity depends on the orbital in question as well as the distance between the two atoms, i.e., the local electronic structure. Tight-binding calculations with the use of Green functions give conductivities in good agreement with experiments.[6,5] *Ab initio* calculations are more powerful but difficult to carry out in that they require huge calculation power which restricts them to simple geometries. Chains of single Ni atoms have been investigated[7] and although the problem is oversimplified (colinear magnetization) it gives interesting indications about possible large conductance variations between different magnetic states of the Ni wire.

 

In ferromagnetic materials, up- and down- spin electrons experience a different exchange energy which removes the spin degeneracy. Because their Fermi wavelength is different, the number of transmitted channels for a given constriction is spin dependent. Indeed, conductance quantization of Ni contacts can change with their magnetic state.[8,9] Ono[9] clearly measured steps of $e^2/h$ in the ferromagnetic state of Ni under an applied field which could be switched to $2e^2/h$ when both ends of the atomic contact were magnetized antiparallel. This observation is understood[10] considering that introduction of a domain wall in an atomic contact reintroduces both spin species hence restoring spin degeneracy. It is also believed that in a narrow wire, the domain wall size should scale with the wire width.[11] The magnetization is then expected to fully rotate on an atomic length scale, which results in a potential step of height given by the exchange splitting. Because transport is ballistic, the main resistive effect comes from reflection on the wall induced potential step which can close some of the transmission channels. For contacts with a large number of opened channels, the induced change of conductance should scale with the ratio of the exchange splitting to the Fermi energy,[12] which is typically around 10 percent. Interestingly, when the contact gets narrower, the longitudinal kinetic energy of conduction electrons is reduced and can become smaller than the exchange splitting. It is then predicted that near the first jump to contact, large MR effects could arise because a thin DW can close the first opened channel.[13] However, for $3d$ elements it is known that several conduction channels are opened, even for a single atomic contact.[5,6] One can then wonder whether this last prediction can actually be realized. Experimentally, several articles report on MR as a function of the magnitude of the applied field. In nanowires around 10 nm in diameter, $R$ increases by less than one per cent in the presence of a DW.[15] However, when scaled by the expected wall width, the obtained MR within the wall reaches several 100%. In nanometer sized constrictions on the other hand, the MR was found negative and small.[16] Much larger effects were obtained in electrodeposited Ni nanocontacts where the resistance could be changed with an applied field by several 100%[17] and more recently by over 3000%.[18] However, the amplitude and sign of the effect seem to be rather unpredictable.

Here we report on measurements of the magnetoresistance as a function of field through only a few Ni atoms. We used the break junction technique[19] where a 100 nm wide 1 $\mu$m long bridge is defined by electron beam lithography onto a polyimide layer. The polymer is further etched isotropically by reactive ion etching which undercuts below the Ni structure (see Ref. 19 for more details). The end result is a suspended bridge attached to two electrodes of different shape presenting two distinct coercive fields. One can then bend the substrate with a micrometer screw fitted in a continuous flow cryostat where a 2 T field can be applied. During elongation or contraction of the bridge, the resistance is continuously monitored in a constant voltage mode with an ac measurement technique. The setup is particularly stable since most of the structure is attached to the substrate and only the narrow bridge is suspended. It is possible to mechanically stabilize the contact with a precision estimated below the picometre. The experimental procedure is as follows: the bridge is first cooled down to 17 K and then elongated until breaking. The two halves of the bridge are then brought back together and the transport goes from tunneling to contact when the first atom bridges the two electrodes.[6] When the contact is closed further, the conductance varies in a noncontinuous manner showing jumps as the atomic configuration of the contact changes. One can then go back and forth breaking and closing the contact while measuring the conductance. Not all the conductance plateaus can be observed each time and Fig. 1 shows the measurement with the largest number of steps (obtained while breaking under a 0.3 T transverse field). In particular, the lowest conductance level at $0.7e^2/h$ appears only rarely.

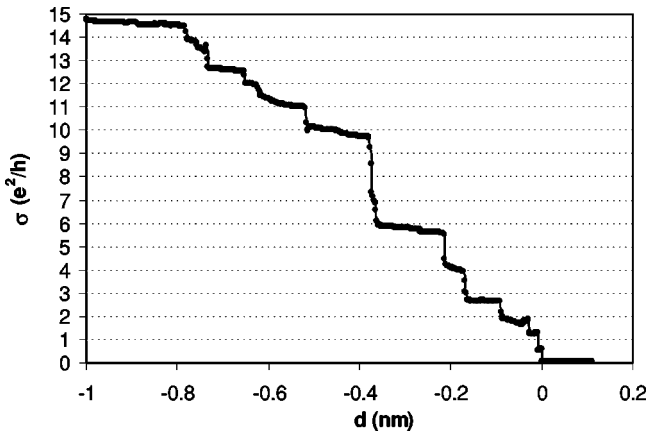


FIG. 1. Conductance steps as a function of distance between the electrodes while breaking the bridge under an 0.3 T transverse field.

The system can then be stabilized on these plateaus and the conductance is measured as a function of an applied magnetic field. The tunneling regime was first investigated when the two arms of the bridge were pulled apart and stabilized at a resistance of 350 k$\Omega$. An external magnetic field is applied perpendicular to the suspended bridge, and the resistance variation is recorded. Generally, the resistive signal is very noisy at first but reaches a stable value as the contact slowly relaxes. Then, the MR becomes reproducible and symmetrical as shown in Fig. 2. We have also checked that the conductance varies quadratically with the voltage as shown in the inset. At large voltages, strong electric fields make the atoms in the contact move and generate giant random telegraph noise.

This MR curve is surprising in two respects: the amplitude of the effect is small (5%) and the resistance is not lowest at saturation. This is in contrast to the effect obtained in conventional Ni/Insulator/Ni tunnel junctions, where the resistance depends on the spin polarization ($P$) of the electrodes and the angle $\theta$ between the electrodes' magnetization: $\Delta R \propto (\cos\theta - 1)P^2/(1-P^2)$. However, qualitative (and quantitative) differences can be expected because in break junctions the tunneling is between two atomic tips and in vacuum. The current is transmitted through the evanescent electronic wave functions of the two atoms of the contact whose local DOS could be very different from that of flat

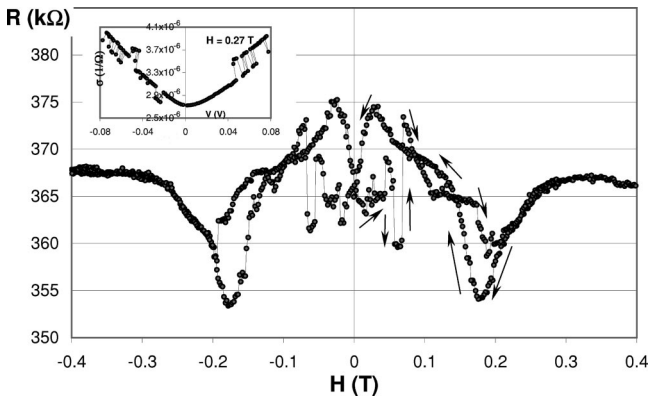


FIG. 2. Resistance in the tunneling regime as a function of an applied transverse field. In inset: the variation of conductance with applied voltage has the $V^2$ component characteristic of tunneling effects.

surfaces. It is also known that the spatial extension of *d* orbitals is short, which makes them less involved in the transport at large atomic distance.[14] Hence, in conventional ''trilayer'' experiments where the insulator is typically over 1 nm thick, *s* (and *p*) electrons dominate and impose their positive polarization. In our case, the tunneling distance is of the order of 3 Å, which makes *d* electrons more involved in the process. Their negative polarization is then likely to compete with that of the *s* and *p* orbitals to lead a lower MR. In any case, because the tunneling resistance varies exponentially with the distance between electrodes, it is very sensitive to any length change resulting, for example, from the field-induced strain in the suspended bridge. Ni has a negative magnetostrictive coefficient related to the magnetization direction. Hence, saturating our sample in a direction transverse to the bridge should increase its length, i.e., reduce the gap through which tunneling occurs. It is likely that the measured MR is influenced by this effect, but because the resistance varies only by less than 5%, we can estimate that the displacement of the apex atoms is below 3 pm.

The gap between the two Ni electrodes was then closed under 0.3 T until the first jump to contact obtained at $1.3e^2/h$ and the system was stabilized on this conductance plateau. There, the local atomic arrangement is likely to be one of two pyramids with overlap of the top Ni orbitals as calculated in Ref. 14 and shown in the inset of [Fig. 3(a)]. An external field was then applied transverse to the bridge and the MR curve was recorded as a function of the field magnitude (Fig. 3a). The MR effect (around 40%) is much larger than that in the tunneling regime and the resistance is lowest at saturation. The curve can be viewed as a bell-shaped MR on which discrete jumps are superimposed. In order to investigate the influence of field direction, we have carried out an other set of experiments with the field applied along the bridge, i.e., along the current lines. Surprisingly, on all the conductance levels studied, the resistance is maximum at saturation [see Fig. 3(b)]. The MR curve is also qualitatively very different, which probably reflects a different magnetization reversal process.

In order to get a clearer picture for the origin of MR in our atomic contact, it is important to understand how mag-

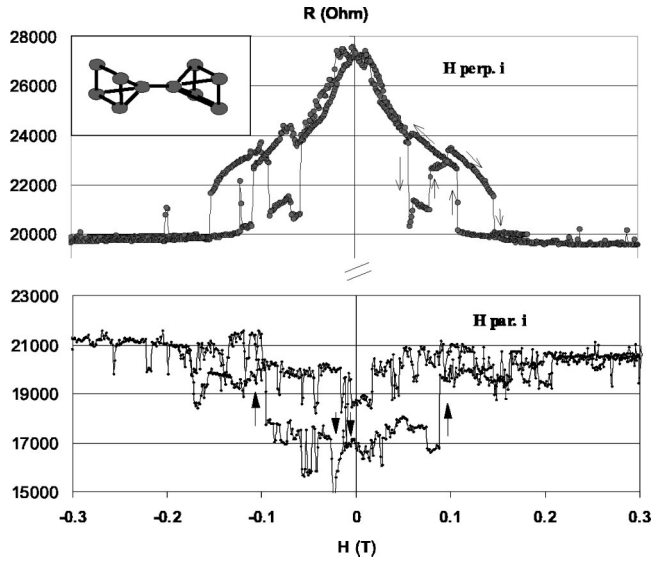


FIG. 3. Resistance as a function of applied field in the atomic contact regime. The field is applied transverse (a) and longitudinal (b) to the bridge (i.e., the current). The inset is the schematics of the expected geometry of the atomic constriction.[14]

netization reverses in the bridge. We carried out micromagnetic calculations which show that in a narrow constriction between two electrodes, the stable zero-field state for the magnetization is to generate two half (90°) walls on either side of the constriction. In our geometry of two 100 nm wide tips touching in a nm size contact, the main component of magnetization lies roughly along the bridge with a regular canting of spins because shape anisotropy in the bridge is affected by the crack. Under transverse applied fields, two vortices are formed at each side of the bridge and get pushed in by the field. Near the atomic contact, the spins align at higher field while no domain wall is left in the electrodes. The saturation process at the junction level is then likely to consist of a gradual decanting of the local magnetization. In any case, micromagnetic calculations do not lead to a configuration where a very thin domain wall is located at the junction even when the electrodes are antiparallel. At the scale of the contact a full atomic calculation of stable spin configurations would be very useful. This can be done *ab initio* for clusters composed of a few atoms but still represents a difficult problem when one wants to consider noncollinear arrangements.

In our MR curves, one can notice dips of a few percent around 0.06 T in both tunneling and atomic contact MR when the field is applied transverse to the bridge. These correspond to reversal of our big electrodes, far from the atoms of the contact. Since the voltage drop is almost exclusively within the few atoms at the constriction, we conclude that the magnetic configuration at the contact is only slightly affected (the amplitude of the effect is small and negative). Most of the resistance variation happens at higher field, where the magnetization in the contact gradually saturates. In magnetic atoms, the spin direction and the orbitals are coupled through the spin-orbit interaction. When the atomic spins are forced to rotate towards the direction of the external field, the relevant orbitals are affected and the electron scattering

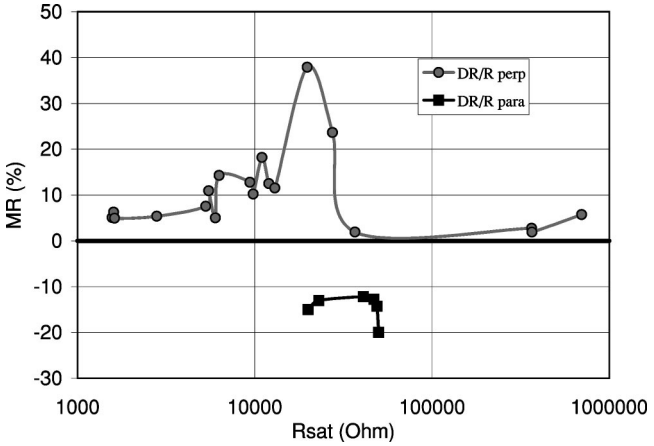


FIG. 4. Summary of the magnetoresistance measured on the different conductance plateaus showing a systematic change of sign between the field applied parallel and perpendicular to the bridge.

changes. The resulting effect in electrical transport is the AMR which is rather small in bulk $3d$ metals, but could be enhanced when dimensionality is reduced. The measured MR change of sign in the longitudinal geometry provides evidence that this effect is indeed important in atomic contacts where the orbital overlap plays a crucial role. In comparison, effects of reflection of the electrons on the magnetic potential seem to be weaker since the resistance at saturation can be larger or smaller than that at zero field depending on the field direction.

The results obtained in the whole range of conductances including the first few plateaus consistently show a change of sign between the MR obtained with the field parallel and perpendicular to the bridge as shown in Fig. 4.

When the contact is progressively stretched (without reaching the tunneling regime), $R(H)$ curves change continuously. The last resistance drop to saturation at high-field gets larger and dips appear at that field. It is then likely to correspond to a flip of the spin of the two apex atoms which are the most influenced by local anisotropy because of the reduced symmetry of their environment. All the large discrete jumps are obtained at fixed (symetrically positive and negative) values of the applied field. We propose that they are due to field-induced spin rearrangements of the few atoms in the contact. Transmission of the conducting channels changes abruptly because of the potential of magnetic origin determined by the atomic spin configuration. The complete MR curve could then be rationalized in terms of the combined effect of spin-orbit coupling induced changes of orbital overlap and reflection at the potential defined by the local spin configuration. The break junction geometry probably does not allow for the creation of an atomic size DW at the contact during field sweeps. This is more likely to be achieved when antiparallel electrodes are brought, from a distance, into contact. In that case, one could perhaps stabilize a metastable magnetic state where magnetization in the contact rotates at an atomic scale. Nevertheless, we believe our measurements underline the importance of spin-orbit coupling in generating a large atomic AMR effect.

This work was supported by the European Union through the Contract MAGNOISE No. (IST-1999-10486). We also gratefully acknowledge fruitful discussions with R. Cron, C. Urbina, H. Jaffrès, A. Fert, C. Barreteau, and A. Levy Yeyati.

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