Magnetic and orbital blocking in Ni nanocontacts

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We address the fundamental question of whether magnetoresistance (MR) of atomic-sized contacts of nickel is very large because of the formation of a domain wall (DW) at the neck. Using ab initio transport calculations we find that, as in the case of nonmagnetic electrodes, transport in Ni nanocontacts depends very much on the orbital nature of the electrons. Our results are in agreement with several experiments in the average value of the conductance. On the other hand, contrary to existing claims, DW scattering does not account for large MR in Ni nanocontacts.

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The strong sensitivity of the current flow between two ferromagnetic metals (FM’s) separated by a nonmagnetic region to the relative orientation of their magnetization vectors is a fundamental physical phenomenon with a huge impact in the magnetoelectronics industry. The figure of merit is the ratio between the conductance for antiparallel (AP) and parallel (P) relative magnetic orientations of the FM’s, \( G = G_{AP}/G_{P} \), which can be selected with an external magnetic field. Two different conventions are used to characterize the so-called magnetoresistance (MR), \( MR_{1} = 100 \times (1 - x)/x \)% and \( MR_{2} = 100 \times (1/x - 1)/x \)%. Metallic multilayers with alternated ferromagnetic and nonmagnetic metallic layers display a large MR known as giant MR (GMR) (Ref. 2). MR in FM/insulator/FM systems is known as tunnel MR (TMR) (Ref. 3) and values of MR \( \approx 30 \)% have been reported. More recently, a number of groups have studied MR in Ni nanocontacts (produced by break junctions or electrodeposition), where two sections of a Ni wire are connected through single-5–10 or multiple11,12 nanometer-size contacts. In this arrangement the intermediate region connecting the two bulk FM’s has a different geometry but the same chemical composition, in contrast to GMR and TMR systems. Some groups have obtained values of MR going from \( 10^{2} \) to \( 10^{4} \) (\( x = 10^{-1} \) to \( x = 10^{-3} \)) (Refs. 5–7 and 12) while others obtain moderate or even negative values. In these systems the resistance predominantly comes from the region with the smallest section, where electron transport is coherent and conductance is dominated by the quantum-mechanical transmission of electrons at the Fermi level.13 Mainly, two different mechanisms have been proposed so far to account for the large values of MR when observed: Domain-wall (DW) scattering14 and magnetostriiction.8,11 For the former it has been argued that in the AP arrangement a DW is pinned at the nanocontact and is responsible for strong spin scattering which gives an extra contribution to the resistance as compared to the P configuration, resulting in a large “ballistic” MR (Ref. 14).

The fundamental question of whether MR is dramatically enhanced in atomic-sized ferromagnetic contacts due to the presence of a DW remains open and is the subject of this paper. Three ingredients are essential to answer this question: First, as in the case of nonmagnetic nanocontacts, is the electronic structure of the last atom(s) which determines the number of available transport channels.16 Second, is the presence of inhomogeneous magnetization profiles, e.g., a DW, which can induce spin scattering and affect current flow.14,17–19 Third, is the atomic structure (geometry) which affects both the electronic and magnetic structures, and thus, the transmission of these channels. Previous theoretical works present mutually conflicting results with methodologies that either used an oversimplified description of Ni electronic structure18,20,21 or idealized geometries.19,22 Here we present transport calculations across Ni nanocontacts describing the electronic, magnetic, and atomic structure with ab initio calculations.23–25 Our results lead us to conclude that intrinsic ballistic MR is certainly not large in atomic-size Ni nanocontacts.

Spin-dependent transport formalism. Transport through atomic-size metallic contacts is currently understood in terms of elastic transport of noninteracting quasiparticles through a one-body potential that describes their interaction with the constriction. In this approach, the conductance \( G \) is proportional to the quantum-mechanical transmission \( T \) associated with the potential. On the other hand, the spontaneous breaking of the spin degeneracy in transition-metal ferromagnets, which is due to electron-electron interactions, can also be properly understood in terms of a mean-field description, where quasiparticles interact with a spin-dependent self-consistent potential. Once the self-consistent field is determined for a given geometry, the quantum-mechanical spin-dependent and energy-dependent transmission probability \( T_{\sigma\sigma'}(E) \) can be obtained, and thereby the zero-bias conductance, using Landauer’s formula,13 is

\[
G = \frac{e^2}{h} \left[ T_{\uparrow\uparrow}(E_F) + T_{\downarrow\downarrow}(E_F) + T_{\uparrow\downarrow}(E_F) + T_{\downarrow\uparrow}(E_F) \right].
\]

In the above expression we only make explicit the dependence of \( T_{\sigma\sigma'} \) on the spin channels, which we assume is well-defined in the leads.

Ab initio cluster embedded calculations. It is an experimental fact that the chemical nature of the contact determines the conductance.16 As a rule of thumb,26 the conductance of single-atom metallic nanocontacts can be as large as the number of valence orbitals, but, in practice, is never larger than the number of valence electrons. A natural de-
scription of the problem is in terms of a localized atomic orbital basis, preferably starting from first principles. In previous publications we have presented a method to perform \textit{ab initio} calculations of quantum transport through atomic constrictions and molecules which is based on the code GAUSSIAN (Ref. 27). Our approach has been successful in explaining experimental results in paramagnetic nanocontacts. Here we take it a step further to study systems without spin degeneracy, like ferromagnetic nanocontacts. We solve the problem dividing the system in three different parts: left (L) and right (R) electrodes on one side and the contact region on the other. The spin-dependent one-body Hamiltonian is assumed fixed and homogeneous in the bulk electrodes, but it is determined self-consistently in the contact region subject to the appropriate magnetic boundary conditions.

The density-functional theory (DFT) calculations for the contact region are done with both local spin density approximation (LSDA) and the hybrid functional B3LYP (Ref. 27). The LSDA results are robust against different basis sets and so we rely here on a minimal basis set with a core pseudo-potential as described in previous works. On the other hand, the B3LYP functional is more sensitive to the basis set due to its nonlocal exchange contribution. Therefore we employ here an all-electron basis set. The electrodes are described by means of a semiempirical tight-binding Bethe lattice model. With the appropriate parameters, the Bethe lattice can provide a geometry-independent description of the contacts with a bulk density of states (DOS) which is smoother than the real DOS and mimics an average over both disorder realizations and the actual electrode crystal orientations. Spin-mixing solutions are not considered, i.e., \( S_z \) is a good quantum number. Thus, the last two terms in Eq. (1) do not give any contribution to the conductance. DW-like configurations are obtained for the adequate magnetic boundary conditions and the constraint \( S_z = 0 \).

\textbf{Results.} We restrict ourselves to the study of the last (first) plateau of conductance upon stretching (electrodeposition) as, e.g., in the experiment of Viret \textit{et al.} (Sullivan \textit{et al.}). A reference atomic structure of the contact region has been initially taken like that shown in the inset of Fig. 1. Following Viret \textit{et al.}, we consider the narrowest (and most important) region to consist of two pyramids facing each other, formed along the (001) direction, and with the two tip Ni atoms 2.6-Å apart forming a dimer. Bulk atomic distances and perfect crystalline order are assumed otherwise. \textit{Ab initio} simulations of the breaking process as the one shown in Fig. 2 support this choice. We stress that the section of the nanocontacts varies in the direction of the current flow. This is the situation in real nanocontacts and differs from perfect one-dimensional systems, studied in Refs. 22, and from bulk systems studied by van Hoof \textit{et al.} In this regard, the geometries proposed by Bagrets \textit{et al.} are closer to real nanocontacts, but are not backed up by experiments or simulations.

From the LSDA DOS projected on the tip atoms (not shown) we see that the \( s \) orbitals are spin split by less than 1 eV and that the minority (\( m \)) electrons are hybridized with the \( d \) levels, which are present at the Fermi energy. As a consequence, the LSDA DOS for the majority (\( M \)) electrons at the Fermi energy is significantly smaller than that for the \( m \) ones. These results are compatible with LSDA first-principles calculations for systems with translational invariance, and have been properly accounted for in Anderson-like model Hamiltonians, but are in marked contrast to the \( J_{sd} \) model, usually invoked to understand large values of the MR in nanocontacts. In these models the transmission of itinerant \( s \) electrons is perfect in the ferromagnetic case while the \( d \) electrons do not contribute to the current since they are localized. The MR depends thus dramatically on the ratio between the spin splitting of the conducting \( s \) electrons and the Fermi energy when a DW is present. A large spin splitting needed to give large MR is, however, at odds with the actual Ni band structure and this model must be ruled out from the outset to account for large MR in Ni nanocontacts.

Figure 1 shows the LSDA conductance as a function of energy for both up and down spin channels in two situations: (a) Parallel (P) and (b) antiparallel (AP) bulk magnetic ar-

\begin{figure*}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{(a) LSDA conductance per spin channel in the P configuration for the model nanocontact shown in the inset. (b) Same as in (a), but for the AP configuration.}
\end{figure*}

\begin{figure*}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{(Color online) Conductance of both spin channels for the P and AP configuration as a function of the stretching. The lines connecting the points are just to guide the eye. The insets show the relaxed contact geometry at different values of the displacement (0, 1.5, and 3 Å from left to right).}
\end{figure*}
arrangements. In both cases the self-consistent solution has been forced to respect the high symmetry of the nanocontact. In the AP case the self-consistent magnetization reverses abruptly between tip atoms. The resulting magnetic moment for the contact atoms is \( \approx 1.0 \mu_B \) in both situations. This value is significantly larger than that obtained for bulk or surface atoms (\( \approx 0.6 \mu_B \)) and reflects the low coordination of the tip atoms forming the contact. In the P case the \( M \) channel is, for the most part, composed of a single \( sp \) orbital channel and conducts perfectly around the Fermi energy (set to zero) while the \( d \) channel is composed of three orbital channels (one \( sp \) like and two \( d \) like, which conduct roughly the same), and exhibits a transmission strongly dependent on the scattering energy. In the AP case the system is invariant under the combined transformations that exchange the scattering energy. In the AP case now becomes slightly spin dependent. In Fig. 1 the conductance of both \( P \) and \( AP \) configurations are shown as a function of the stretching of the contact giving a very good description of the electronic structure and the AP configurations are shown as a function of the displacement between outer planes in the core cluster. To do so, we consider a cluster like that shown in Fig. 1. The inner atoms in the cluster (10 in total) are allowed to relax to local minimum energy configurations as we stretch. This results, logically, in lower energy solutions and in the loss of symmetry, so that the transmission in the AP case now becomes slightly spin dependent. In Fig. 2 the conductance at the Fermi energy per spin channel for the \( P \) and \( AP \) configurations are shown as a function of the stretching up to the breakup point, starting from a slightly compressed nanocontact. From this figure we see that the conductance of the \( m \) channel for the \( P \) configuration changes significantly upon small changes. The MR, on the contrary, barely changes as the nanocontact is stretched and is small, reaching vanishing values for the last points in Fig. 2. The conductance approaches a stable value around \( 2e^2/h \) for both \( P \) and \( AP \) configurations.

Discussion and conclusions. As mentioned earlier, the maximum number of conducting channels in atomic-size contacts is roughly determined by the number of valence electrons of the contact atom(s). However, as shown above, this hypothetical upper limit is never reached, particularly for the \( m \) electrons, remaining essentially only one \( M \) channel and one \( m \) channel transmitting in the \( P \) case for stretched contacts. This result is impossible to predict without a full atomistic self-consistent calculation. The \( M \) channel is \( sp \) type. Thus, this channel transmits almost perfectly and evolves smoothly with the stretching of the contact giving a stable contribution \( T_{1\downarrow} \approx 1 \) (see Fig. 2). The \( sp \) orbitals in the \( m \) channel are strongly hybridized with \( d \) orbitals and, therefore, are more sensitive to the contact geometry. The contribution to the conductance of the latter, which form narrower bands, disappears with the stretching and disorder, as expected (see Fig. 2). On the other hand, in the \( AP \) configuration mostly one \( sp \) orbital channel per spin contributes. The conductance per spin channel lies thus in the vicinity of
$e^2/h$, giving $=2e^2/h$ in total and is fairly stable during the last stage of the breaking of the nanocontact.

To conclude, the reason behind the very small MR values is the orbital (or geometric) blocking of most of the a priori available $m$ channels in the P configuration due to the non-ideal geometry of the nanocontacts. The number of bands at the Fermi energy in the case of a perfect monostrand infinite ferromagnetic chain is much larger than the number of non-zero eigenvalues of the transmission matrix in a nanocontact (for a given basis and functional). This phenomenon affects mainly the $d$ bands (no DW involved) and, therefore, we call it orbital blocking. On the other hand, $sp$ bands are less sensitive to geometry. In fact, there can only be a significant blocking of most of the bands in the P configuration due to the non-striction effects and the corresponding formation of wider section contacts or to the presence of adsorbates, which modify the local electronic structure.34

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