Quantum conductance of a single magnetic atom: An *ab initio* study

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Our *ab initio* study explains recent puzzling experiments on the conductance of a single Co adatom deposited either on Cu(111) or on ferromagnetic Co islands and contacted with both magnetic and nonmagnetic electrodes [N. Néel, J. Kröger, and R. Berndt, Phys. Rev. Lett. **102**, 086805 (2009)]. We provide clear evidence that the conductance of a single atomic junction in the contact regime is close to $G_0/2$ (G_0 is the quantum of conductance) for ferromagnetic electrodes and to G_0 for nonmagnetic ones. Spin-dependent calculations reveal that a conductance of $G_0/2$ originates from a combination of partially open majority and minority channels. The bonding between the Co adatom and the Co island reduces significantly the contribution of the minority *d* electrons to the conductance, leading to the observation of half-integer conductance.

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Understanding the electron transfer through atomic scale junctions is of great importance for the miniaturization of the future electronic devices. The conductance, G, of a spin-degenerate atomic-size contact is quantized in units of $G_0=2e^2/h$, according to the Landauer's formula $G=G_0\Sigma T_i$ (e is the electronic charge, h the Planck's constant, and T_i the transmission probability for the ith channel calculated at the Fermi level, E_F).[1] Quantized conductance arises from the ballistic electron transport in a narrow constriction as a consequence of the quantization of the electron motion perpendicular to the conductor. Pioneering theoretical studies of Sharvin showed that in constrictions much smaller than the electron mean-free path the ballistic transport is determined by the geometry only.[2] More than two decades ago this prediction was confirmed in an electrostatically confined two-dimensional electron gas.[3] Later the same was proved in metallic point contacts, where conductance steps of G_0 were observed as a function of the separation between the electrodes in mechanically controllable breaking junctions.[4]

In the case of magnetic systems the spin degeneracy is removed and each spin-polarized channel can contribute at most $G_0/2$ to the total conductance.[5] Several groups have reported the observation of half-integer conductance quantization for both magnetic and nonmagnetic nanocontacts fabricated by using the scanning tunneling microscope (STM) or breaking junctions.[6–13] The most exciting interpretation of such experiments is given by assuming the electron transport to occur via a single fully spin-polarized channel. However, the occurrence of half-integer conductance could be, as well, caused by the presence of contaminations, such as H_2 or CO, and could be completely unrelated to magnetism.[14] In general the proof of a direct relation between half-integer conductance and magnetism is to date still a matter of debate.[14]

A number of experiments have shown conductances quantized in multiples of G_0 for magnetic metallic junctions[15,16] and likewise several theoretical studies reported the absence of the half-integer-quantized conductance.[17–23] A serious obstacle to a complete understanding is the fact that the atomic details of a breaking junction are unknown so that one is never sure whether the observed conductance is associated to a single atom junction or to a more complex structure. In other words it is difficult to disentangle electronic and structural effects. A much more controlled way of carrying out the same measurements is offered by contacting single magnetic adatoms on a surface with STM.[24–30]

Very recent STM experiments from Néel *et al.*[31] have challenged our understanding of transport through single magnetic atoms. In the contact regime the conductance of a Co adatom was found to be G_0 for two nonmagnetic electrodes (substrate and tip) and $G_0/2$ for ferromagnetic ones. The results were explained by arguing that the $G_0/2$ conductance may be either due to transport through a combination of partially open spin channels or, most excitingly, through a single fully polarized channel. Unfortunately, experimentally it is difficult to distinguish between these two situations. In this paper, we present a state of the art *ab initio* study of the transport properties of a single adatom deposited on a metal surface. We follow the experimental setup of Néel *et al.*[31] and reveal that the conductance reduction from G_0 to $G_0/2$ when changing the electrodes from nonmagnetic to magnetic is caused by the strong bonding between the adatom and the electrodes, with both majority and minority spins contributing to the transport.

Our calculations are based on density-functional theory (DFT). The optimization of the atomic junctions for all tip-to-substrate distances is performed by using the VASP code[32] and the PW91 form of the generalized gradient approximation to the exchange and correlation functional. Projector augmented wave potentials[33] with an energy cutoff of 400 eV are used. All the parameters in the calculations are chosen to converge the total energy to within 10^{-5} eV.[34] Transport properties are calculated with SMEAGOL,[35] which combines the nonequilibrium Green's function formalism with DFT.[34] In SMEAGOL calculations, we use a standard double-ζ polarized basis for Cu and a triple-ζ basis plus two polarization orbitals for the *s* shell, for both Co and Ni. The cutoff radii of the first ζ's are determined via an equivalent energy shift of 0.02 Ry and higher ζ's are constructed with the split-norm scheme.[34] A 2×2 in-plane *k*-point grid and a real-space mesh cutoff of 350 Ry are used. All calculations are performed under zero bias.

The Cu(111) substrate is modeled by five layer thick slab containing 16 atoms in each atomic plane. A Co nanoisland is modeled by adding two Co atomic planes to the slab. The

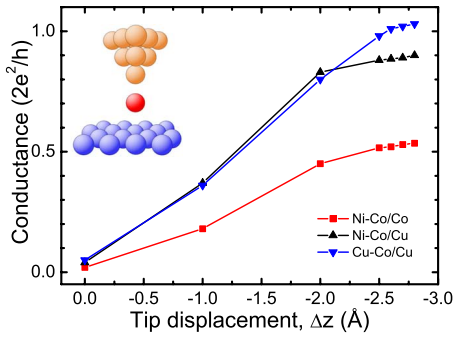

FIG. 1. (Color online) Conductance as a function of the tip displacement, Δz, for three junctions, Ni-Co/Co, Ni-Co/Cu, and Cu-Co/Cu. The inset is the geometrical setup for the calculation.

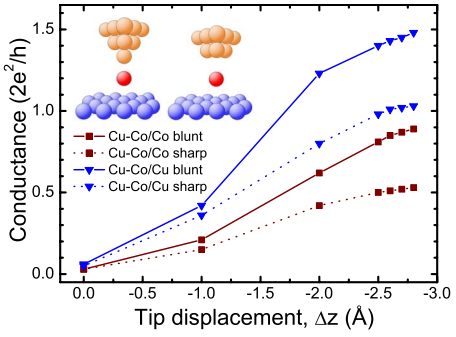

FIG. 2. (Color online) Conductance of Cu-Co/Co and Cu-Co/Cu junctions obtained by assuming either a sharp (dotted lines) or a blunt (solid lines) STM tip. The insets show the calculation setup in the case of sharp and blunt tip, respectively.

adatom is placed above the hollow site of the top layer. The tip is modeled as a pyramidal cluster,[36] constructed from ten Ni or Cu atoms and terminated with a single-atom apex. In transmission calculations, nine additional Cu(111) layers are included in the leads. All the atoms are fully relaxed with exception of the three bottom Cu layers of the substrate and the top layer of the tip. The geometries are converged until all the residual forces are smaller than 0.01 eV/Å.

First, we consider the following three junctions: Ni-Co/Co, Ni-Co/Cu, and Cu-Co/Cu (the junction is defined as tip adatom/substrate). These have all been investigated in the experiments of Néel *et al.*[31] The setup used in all calculations is a tip-adatom-substrate junction which mimics the experimental setup as closely as possible. Note, that in previous works a magnetic adatom was either put in a short nanochain or between two pyramids. The ground state of the system in all our calculations is the parallel alignment of the spins of the tip, the adatom, and the substrate. Figure 1 displays the conductance of a single Co adatom sandwiched between different combinations of electrodes as a function of the tip displacement Δz. This measure is used because the distance between the tip and the adatom is not directly accessible in most STM experiments. In experiments one usually fixes a certain tunneling current at a given voltage and, thus, sets a reference zero height ($\Delta z=0$). Note that due to the interaction between the tip and the adatom, the tip-adatom separations are different for different junctions even at the same tip displacement. For all junctions in the paper, $\Delta z=0$ corresponds to a tip-adatom separation of 5 Å, which is different in STM experiments. Therefore it should not be expected that the tip displacements in theoretical calculations are the same as those in experiments. For small tip displacement Δz (from 0 to −1 Å), the conductance rapidly increases, indicating a departure from the tunneling limit. Then, there is a further increase in the intermediate regime (−1 to −2 Å) and finally in the contact limit, where the chemical bond forms (−2 to −3 Å), the conductance exhibits only small variations with Δz. Importantly, our calculations reveal that at a large Δz (−2.5 to −3.0 Å) the conductance of the Ni-Co/Co junction is close to $G_0/2$ while that of Ni-Co/Cu and Cu-Co/Cu range between $0.88G_0$ and $0.98G_0$. These results are in excellent agreement with the experimental findings.[31]

To our surprise however, the calculated conductance of the Cu-Co/Co junction is also equal to $\sim G_0/2$ (see Fig. 2, square dotted line), which is significantly lower than what found in the experiments ($0.88G_0$).[31] We attribute such a disagreement to our idealized tip model, i.e., to the fact that our tip is atomically sharp while the apex of the tip used in experiments likely consists of several atoms. In order to prove that the tip structure significantly affects the transport through Co adatoms we have performed conductance calculations by using a blunt Cu tip. This consists of only nine atoms and it is obtained from the previous one by removing the most external Cu so that the apex consists of three atoms in the same plane. In experiments, however, the tip apex could be formed by a Cu cluster consists of several Cu atoms. The results in Fig. 2 show that for Cu-Co/Co the blunt Cu tip returns a conductance of $0.81G_0$, which is now close to the experimental value. Thus, our results suggest that measurements of Néel *et al.*[31] were most likely performed with blunt tips. As expected from Sharvin's argument,[2] a significant conductance enhancement is found also for the Cu-Co/Cu junction, which is larger than that of experimental results. The discrepancy between theoretical and experimental results may be due to the oversimplified blunt tip model. In order to obtain a more detailed insight into the effect of the atomic arrangement on the conductance, we now compare calculations performed either with fully relaxed and unrelaxed geometries. In particular, we concentrate the discussion on Ni-Co/Co and Ni-Co/Cu.

First we note that the vertical relaxation of a Co adatom on a Co monolayer is significantly larger than that on a Cu substrate. The adsorption height in fact goes from 1.57 Å for Co to 1.70 Å for Cu. The stronger interaction of the Co adatom with the Co substrate compared to that with the Cu one suggests that the influence of the tip on the adatom position could be more pronounced for the Cu substrate. Our calculations further show that at small Δz the position of the adatom is practically unaffected by the tip, hence that the junction conductance in the tunneling limit is little influenced by atomic relaxations (Fig. 3).

In contrast, in the transition and contact limits the attractive interaction between the tip and the adatom strongly changes the adatom adsorption height. An adatom upward vertical displacement of 0.1–0.2 Å leads to a reduction in the tip-adatom separation of 0.2–0.3 Å. This is at the origin of the enhancement of the conductance in a fully relaxed

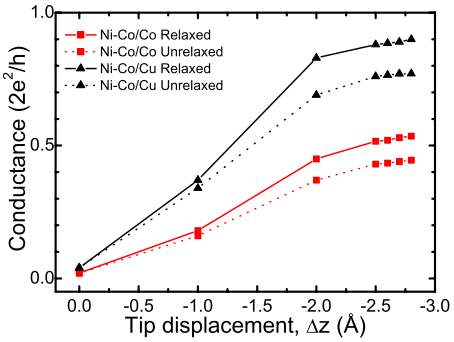

FIG. 3. (Color online) Conductance of Ni-Co/Co and Ni-Co/Cu junctions calculated with (solid lines) and without (dotted lines) geometrical relaxation.

geometry. Note that at $\Delta z < -2.8$ Å the adatom is pushed toward the substrate while the tip-apex atom is pushed upward due to the repulsive interaction.[28] Thus, despite the reduction in the tip-substrate separation, the distance between the apex tip and the adatom remains practically unchanged. Consequently, also the conductance exhibits only small changes. To summarize, the above results clearly demonstrate the interplay between atomic relaxation and junction conductance, namely, that by reducing the tip-substrate separation a local perturbation in the atomic arrangement of the junction is introduced leading to conductance enhancement.

Now we turn our attention to discussing the spin-dependent properties of the transport. Our DFT calculations show that the magnetic moment of the Co adatom reduces from 1.94 μ_B (1.79 μ_B) to 1.74 μ_B(1.63 μ_B) for Ni-Co/Co (Ni-Co/Cu), during the transition from tunneling to contact limit. Such large magnetic moment suggests that both spin directions might contribute to the transport. This is proved next in Fig. 4, where we present the transmission coefficient as a function of energy for the two junctions at a tip displacement of −2.5 Å. At a first glance, one notices that the main difference between Ni-Co/Co and Ni-Co/Cu is a strong suppression of the transmission in the minority channel for Ni-Co/Co while the majority one contributes approximately equally in the two junctions. These results unambiguously

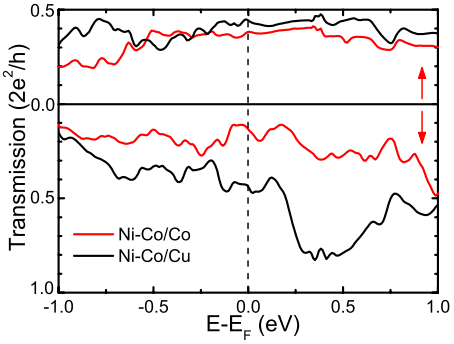

FIG. 4. (Color online) Zero-bias transmission coefficient for Ni-Co/Co and Ni-Co/Cu junctions at a tip displacement of −2.5 Å. The upper half of the figure is for majority spins (↑) and the lower part for minority (↓).

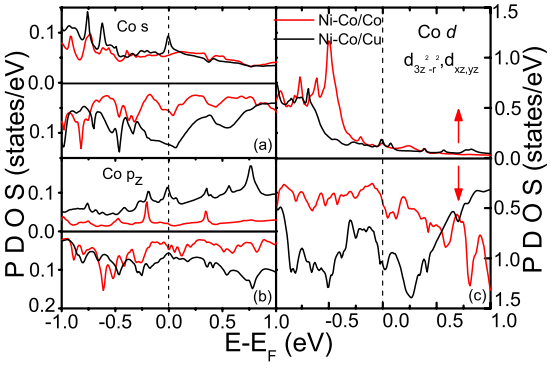

FIG. 5. (Color online) Projected density of states of the Co adatom in Ni-Co/Co and Ni-Co/Cu junctions at a tip displacement of −2.5 Å; (a) 4*s* states; (b) $4p_z$ states; and (c) *dz* (sum of $d_{3z^2-r^2}$ and $d_{xz,yz}$ orbitals) states.

prove that the $G_0/2$ conductance of Ni-Co/Co is caused by a strong suppression of the minority channel, although both electron spins contribute so that there is not a fully spin-polarized current.

If we define the spin polarization at E_F as $P=(T_\uparrow - T_\downarrow)/(T_\uparrow + T_\uparrow)$, we find $P=50\%$ for Ni-Co/Co and only $P=2.5\%$ for Ni-Co/Cu. This means that one can tune the spin-polarized current through a single magnetic adatom by choosing an appropriate substrate with the underlying physics related to the character of the bonding between the adatom and the substrate. Such a conclusion can be drawn from the analysis of the orbital projected density of states (PDOS) of the Co adatom for the two different junctions (Ni-Co/Co and Ni-Co/Cu). Figure 5 shows that the main contribution to the PDOS of the majority states of both junctions is from *s*, p_z, and *dz* (*dz* indicates the sum of $d_{3z^2-r^2}$ and $d_{xz,yz}$. The transport is along the *z* direction.) orbitals, which therefore participate the most to the transmission. Since *s*, p_z, and majority *dz* orbitals are rather insensitive to the magnetism it does not surprise that the contribution from the majority electrons to the transmission is similar in Ni-Co/Cu and Ni-Co/Co. In contrast spin-down *dz* electrons determine the minority conduction. Their contribution to the Co adatom PDOS is strongly suppressed in Ni-Co/Co as compared to Ni-Co/Cu around the Fermi level. This is mainly caused by the hybridization between the minority *d* states of the adatom and the minority *d* states of the Co substrate. When a Fe atom is adsorbed on the Co substrate, similar strong suppression of the Fe minority *d* states is observed in Ni-Fe/Co junction, which results in a conductance of $0.53G_0$.

In conclusion, by using state-of-the-art *ab initio* methods we have confirmed recent experimental findings on the half-integer conductance of single-atom magnetic junctions. We have given clear evidence that a significant reduction in the conductance when a Co adatom is placed on a Co surface is caused by the suppression of the minority channel, which in turn is due to the strong hybridization between the magnetic adatom and the substrate. The intensity of the hybridization is determined by the energy position of the minority *d* states of the adatom and the substrate. The interplay between the atomic structure and the conductance in the junction is also

explained. It is in fact true that spin-dependent electron transport through single magnetic adatoms can be engineered by an appropriate choice of the substrate, the atomic structure of the contact and the shape of the tip.

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