Conductance of single-atom magnetic junctions: A first-principles study

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We present a first-principles investigation to show that the contact conductance of a half conductance quantum ($G_0/2$) found previously does not generally hold for single-atom magnetic junctions composed of a tip and an adatom adsorbed on a surface. The contact conductance of the Ni-Co/Co(111) junction is approximately $G_0/2$, while for the Co-Co/Co(111), Ni-Ni/Ni(111), and Ni-Ni/Ni(001) junctions the contact conductances are $0.80 G_0$, $1.55 G_0$, and $1.77 G_0$, respectively. The deviation from $G_0/2$ is mainly caused by the variation of the spin-down conductance largely determined by the minority $d$ orbitals, as the spin-up one changes little for different junctions.

The conductance of atomic junctions has been extensively studied by experimental techniques such as the scanning tunneling microscope (STM) [2-6], mechanically controlled break junctions (MBCJ) technique [7-11], and electrochemical methods [12, 13]. Conductances of fractional and integer $G_0$ have been reported for both nonmagnetic and magnetic junctions. Of the particular interest is the observation of $G_0/2$ conductance, e.g., for the noble Pt, Pd, and magnetic Co single-atom chains [8], as well as Cu and Ni nanowires [12, 14], although the controversy still exists by arguing that the contamination of [12, 14], although the controversy still exists by arguing that the contamination of H$_2$ may cause the $G_0/2$ conductance [10]. Theoretically, the transport property of atomic junctions has been investigated intensively by using either tight-binding methods [15-17] or density functional theory (DFT) combined with quantum transport calculations based on nonequilibrium Green’s function (NEGF) formalism [18-27]. However, theoretical calculations have hardly obtained the $G_0/2$ conductance so far. This may be attributed to the difference between the simulation model and the real junction structure which is usually difficult to know experimentally.

Recently, a STM experiment found that in the contact regime the conductance is $G_0/2$ for a single-atom magnetic junction composed of a ferromagnetic Ni tip in contact with a single Co adatom on a ferromagnetic Co island [3]. This experiment provides a more detailed atomic geometry of the junction than those ever reported, which enables a precise theoretical modeling to explore the origin of the $G_0/2$ conductance. Calculations show that the conductance of $G_0/2$ is not due to a fully polarized single channel but a combination of the partially open majority channels and the suppressed minority channels [24]. However, it still remains unknown whether the $G_0/2$ conductance exists generally for other single-atom magnetic junctions, and how factors like junction structure and species affect the conclusion. In this work, we perform first-principles calculations to investigate this issue and find that the $G_0/2$ conductance is not a general behavior for several other single-atom magnetic junctions. This is because the minority-spin conductance is sensitive to the junction species.

We first study the magnetic junction fabricated by Néel et al. [3] The system is modeled by a tip-adatom/surface junction (denoted by Ni-Co/Co(111)) as displayed in Fig.1(a). The single Co adatom is adsorbed on the fcc site of the fcc Co(111) surface represented by a three-layer slab, with each layer containing 4 × 4 atoms. The Ni tip is modeled by a single Ni apex atom and a Ni(111) monolayer on a two-layer Co(111) slab (test calculations using three Ni(111) layers on a two-layer Co(111) slab show only minor effect on the result). The tip-apex atom is placed above the Co adatom in the $z$ direction. In transport calculations, these atoms construct the scattering region, and three additional Co(111) layers are added at the two ends of the scattering region, respectively, to mimic the left and right electrodes (leads). For the other three junctions considered in this work, i.e., Co-Co/Co(111), Ni-Ni/Ni(111), and Ni-Ni/Ni(001), the treatment is similar except that a four-layer slab is used for the Ni(001) surface and four additional atomic layers are added to represent the leads of the Ni(001) junction, as illustrated in Fig.1(b).

In this work, the ferromagnetic spin configuration is adopted for the whole junction. Our calculations show that if the junction (including the tip and the substrate) is considered as one system, the ferromagnetic configuration is the ground state, as was also found in a previous calculation for the Ni-Co/Co(111) junction [22]. If the junction is considered as two separated system (i.e., the tip and the substrate) the antiferromagnetic configuration between them may be also possible.
as considered in Ref. [4, 5]. However, the only consideration of the ferromagnetic configuration does not affect the purpose and conclusion of this work: The contact conductance of $G_0/2$ is not a general behavior for different single-atom magnetic junctions.

The scattering region is optimized using VASP code [27]. The bottom layer of the surface and the top layer of the tip are fixed during the structure optimization while the other atoms are fully relaxed until the maximum force is smaller than 0.01 eV/Å. Projector augmented-wave method [28] is used for the wave function expansion with an energy cutoff of 300 eV. The PW91 version [29] of the generalized gradient approximation (GGA) is adopted for the electron exchange and correlation.

For the quantum transport calculation, we adopt the NEGF-DFT approach [30, 31] which combines the NEGF formula for transport with ab initio DFT calculation for electronic structure. In practice, the infinitely large open system is divided into three parts: left lead, right lead, and scattering region, as mentioned above. The self-consistent Kohn-Sham Hamiltonian of the scattering region and the self-energies of two semi-infinite leads are used to construct a single-particle Green’s function from which the transmission coefficient ($T$) at any energy is calculated. The conductance $G$ then follows from a Landauer-type relation. For the DFT part, we use a numerical basis set to expand the wavefunction [32]: A double zeta plus polarization basis set (DZP) is adopted for all atomic species. The Perdew-Burke-Ernzerhof (PBE) version of GGA is used for the electron exchange and correlation and the optimized Troullier-Martins pseudopotentials [34] are used for the atomic cores. We define the spin-polarization ratio at Fermi energy as $P = (T_1 - T_0)/(T_1 + T_0)$, where $T_1$ and $T_0$ denote the transmission coefficient of the majority and minority spin, respectively.

The conductance of the magnetic Ni-Co/Co(111) junction is given in Fig 2(a) as a function of the tip height (i.e., the distance between the tip-apex atom and the surface before the relaxation). It shows that as the tip height decreases, the conductance increases and shows a faster change in the transition region around 5.4-5.2 Å, and then increases slowly in the contact region (below 5.2 Å). The conductance data in the transition and contact regions can be approximated by two straight lines, and their intersection point defines the contact conductance, as used in Refs. [3, 15]. According to this definition, we obtain a contact conductance of 0.48$G_0$ for the Ni-Co/Co(111) junction, which agrees very well with the experimental result [3]. The two spin components of the conductance give a spin-polarization ratio of 0.46 for the tip height of 5.2 Å, which is also in good agreement with the previous first-principles calculation [24].

Fig 2(b) gives the adatom’s adsorption height for different tip heights. On can see that in the contact region the adsorption height is obviously larger than that in the tunneling region, and so is the conductance. This is a result of the enhanced adatom-tip interaction in the contact region. An important thing to note is that the adsorption height experiences a large jump for the tip height around 5.3 Å, where the conductance also rapidly increases as shown in Fig 2(a). This indicates that the adsorption height of the adatom has a direct influence on the conductance because the resulting adatom-tip separation determines the coupling strength (the degree of orbital overlap). Physically, the adsorption height and the resulting adatom-tip separation for a specific tip height is determined by the interaction between the tip and the substrate, which may depend on the junction structure and species. Therefore, to investigate the influence of these two factors on the conductance is important to find out whether a $G_0/2$ conductance is a general result for other magnetic single-atom junctions.

Next, we investigate the conductance of the following three junctions, Co-Co/Co(111), Ni-Ni/Ni(111), and Ni-Ni/Ni(001). The first two have a similar structure as Ni-Co/Co(111) but are of different species. Thus by comparing their results, we can evaluate the role of species in de-
FIG. 3: (a) Conductance of the Co-Co/Co(111), Ni-Ni/Ni(111), and Ni-Ni/Ni(001) junctions, respectively. (b) and (c) show the spin-up and spin-down conductances of these three junctions and of the Ni-Co/Co(111) junction, respectively.

Determining the conductance of the magnetic junctions. The Ni-Ni/Ni(001) and Ni-Ni/Ni(111) junctions are of the same species but have different structures, which enables us to explore the influence of the atomic structure on the conductance.

Fig. 3(a) displays the conductance as a function of the tip height, and Figs. 3(b) and (c) give the two spin components of the conductance, for the Co-Co/Co(111), Ni-Ni/Ni(111) and Ni-Ni/Ni(001) junctions, respectively. We find that their contact conductances are $0.80G_0$, $1.55G_0$, and $1.77G_0$ with the corresponding spin-polarization ratios of 0.1, -0.45, and -0.33, respectively. Obviously, the contact conductances of the three junctions deviate largely from $G_0/2$ and also differ much from each other.

Specifically, the spin-up conductance of the Ni-Co/Co(111), Co-Co/Co(111), and Ni-Ni/Ni(111) junctions are very close (see Fig. 3(b)), while their spin-down conductances differ from each other significantly (see Fig. 3(c)). Since these three junctions are of the similar structure, this difference is due to the different junction species which give rise to different local electronic states and tip-adatom couplings.

We find that the spin-up conductance is mainly contributed by the majority $s$, $p_z$, and $d_z$ (sum of $d_{z^2}$, $r_z$, $d_{xz}$ and $d_{yz}$ with $z$ being the transport direction) orbitals and their contributions to the majority conductance are almost the same for the four different magnetic junctions [24]. Consequently, the spin-up conductance is little changed for the different junctions. As an example, we give in Fig. 4(a) the majority-orbital-projected density of states (PDOS) of the Ni adatom in the Ni-Ni/Ni(111) junction and that of the Co adatom in the Co-Co/Co(111) junction with a tip height of 4.4 Å. It can be seen that the majority PDOS at the Fermi energy are very close for the two adatoms, leading to the similar spin-up conductances for the two junctions (see Fig. 3(b)). In contrast, the spin-down conductance is basically determined by the minority $d_z$ orbitals. Their contribution to the minority PDOS of the adatom at the Fermi energy is much larger than the $s$ and $p_z$ orbitals, as shown in Fig. 4(c) for the Co-Co/Co(111) junction. Importantly, as displayed in Fig. 4(b), the minority $d_z$ PDOS of the adatom at the Fermi energy varies significantly between the junctions, leading to the very different spin-down conductances for these two single-atom magnetic junctions.

Now let us have a look at the effect of the junction structure. By comparing the results for the Ni-Ni/Ni(111) and Ni-Ni/Ni(001) junctions (see Fig. 3), we find that both the spin-down and spin-up conductances are different, causing a $0.2G_0$ difference in the total conductance as shown in Fig. 3(a). However, this difference is smaller than those between the Co-Co/Co(111), Ni-Co/Co(111), and Ni-Ni/Ni(111) junctions caused by the different junction species (see Fig. 3). Our results show that the $G_0/2$ conductance does not generally hold for the single-atom magnetic junctions investigated in this work. This is due to the large variation of the spin-down conductance which largely depends on the junction species as well as the junction structure.

Finally, we would like to comment on the major contribution from the majority or the minority to the total conductance. In Ref. [4], the authors investigated the conductance of single-atom magnetic junctions of Cr-Co/Fe(110) and Cr-Cr/Fe(110). It was found that the conductance is mainly determined by the majority channel. In this work, we investigate the contact conductance of single-atom magnetic junc-
tions with different chemical species and structures. Our calculations show that for the Ni-Co/Co(111) junction the contact conductance is also mainly determined by the majority channel: The majority and minority conductances are 0.38 and 0.13 $G_0$, respectively. However, for the other three junctions, the minority conductance is larger than the majority one. This indicates that the major contribution to the total conductance, from the majority or the minority, is also dependent on the junction species and structure.

In conclusion, by performing first-principles quantum transport calculations we have investigated the spin-dependent conductance of the four single-atom magnetic junctions. For the Ni-Co/Co(111) junction the conductance is very close to $G_0/2$, while for the Co-Co/Co(111), Ni-Ni/Ni(111) and Ni-Ni/Ni(001) junctions the conductances are far away from $G_0/2$ and differ significantly from each other. It was found that the spin-up conductance is little influenced by the junction species, and therefore, this difference is mainly due to the variation of the spin-down component determined by the minority $d_z$ orbitals which are sensitive to the junction species. On the other hand, the junction structure can affect both the spin-up and spin-down conductances but not as remarkably as does the junction species for the spin-down one. Our calculation shows that the previously found $G_0/2$ conductance does not generally exist for single-atom magnetic junctions since the conductance is largely dependent on the junction species and is also influenced considerably by the junction structure.

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