

Electronic structure and transport properties of atomic NiO spinvalves

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Abstract

Ab-initio quantum transport calculations show that short NiO chains suspended in Ni nanocontacts present a very strong spin-polarization of the conductance. The generalized gradient approximation we use here predicts a similar polarization of the conductance as the one previously computed with non-local exchange, confirming the robustness of the result. Their use as nanoscopic spinvalves is proposed.

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Introduction- While bulk nickel monoxide (NiO) is a common example of a correlated insulator with antiferromagnetic (AF) order (see e.g. [1,2]), short one-dimensional NiO chains can actually become half-metallic conductors, as we have shown recently with ab-initio quantum transport calculations [3]. There we employed the hybrid density functional B3LYP as it gives a reasonable description of the electronic and magnetic structure of bulk NiO [2] in contrary to the local density approximation (LDA) and the generalized gradient approximation (GGA) to density functional theory (DFT) [5]. B3LYP corrects the self-interaction error inherent in LDA and GGA by combining a (local) GGA exchange functional with the (non-local) Hartree-Fock (HF) exchange [4].

On the other hand, it is well known that metallic systems are better described with a pure GGA functional. Thus it is not clear a priori which kind of functional will give a better description of the composite system consisting of the atomic NiO chain and the metallic electrodes of a Ni nanocontact. It is therefore worthwhile to investigate the robustness of the obtained results [3] for the NiO chains when a pure GGA functional is used. Comparing the GGA results and B3LYP results further allows us to gain some insight on the effect and relevance of HF exchange in the B3LYP functional on the transport properties of the NiO chains.

Method- The electronic structure of the infinite chain is calculated with the CRYSTAL ab-initio program for crystalline structures [6] while the electronic structure and transport properties of the Ni-O-Ni nanobridge in the Ni nanocontact is calculated with our ALACANT ab-initio quantum transport program[3]. The basis sets employed here are those of our previous work[3]. The GGA functional employed in the calculations is the one by Perdew and Wang[7].

Results and discussion- Within the GGA the one-dimensional NiO chain is always conducting for FM order in contrast to the B3LYP results where the FM state can be either insulating or half-metallic[3]. In Fig. 1(a), we show the GGA band structure of an ideal one-dimensional NiO chain in the ferromagnetic (FM) phase. Compared to the B3LYP band structure for the half-metallic state at same lattice spacing [3] we see that the occupied bands have been raised considerably in energy. In particular, the doubly-degenerate flat minority-spin (m) band of type (d_2) composed of Ni $3d_{xy}$ and $3d_{x^2-y^2}$ orbitals (well below the Fermi level with B3LYP), now actually crosses the Fermi level. Also the doubly-degenerate and previously half-filled m band of type (d_1) composed of Ni $3d_{xz}$ and $3d_{yz}$ orbitals hybridized with O $2p_x$ and $2p_y$ orbital (the only conduction band with B3LYP) calculation are raised somewhat in energy. Consequently, the previously empty m band of type (d_0) composed of Ni $3d_{3z^2-r^2}$ orbitals is lowered in energy with respect to the other filled or partially filled $3d$ bands, and becomes a conduction band.

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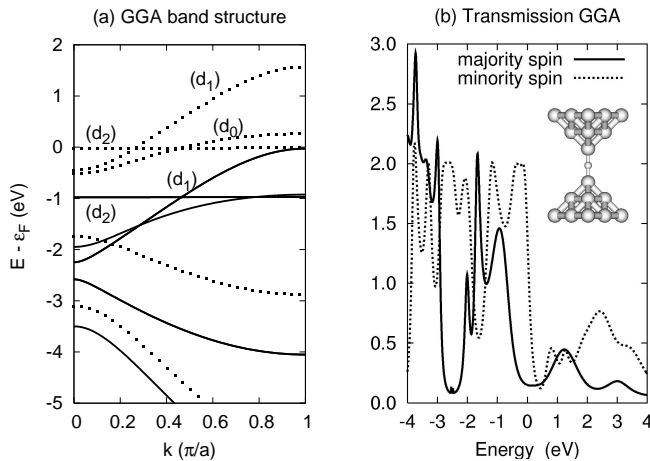


Fig. 1. (a) Spin-resolved band structure of one-dimensional Ni-O chain in the FM phase. (b) Transmission per spin channel for Ni-O-Ni bridge in a Ni nanocontact consisting of two pyramids in the 001 direction with 14 atoms each as shown in inset for parallel alignment of the electrodes' magnetizations. Solid lines indicate M bands/channels, dashed lines m bands/channels. Lattice parameter for the infinite chain and distance between Ni atoms in the Ni-O-Ni bridge are both 3.6\AA .

On the other hand also the majority spin (M) bands are raised in energy. The doubly-degenerate M band composed of Ni $3d_{xz}$ and $3d_{yz}$ orbitals hybridized with O $2p_x$ and $2p_y$ orbitals which was well below the Fermi level in the B3LYP calculation now also crosses the Fermi level near to its upper band edge. Thus in GGA the ideal case of the infinite NiO chain in the FM phase does not represent a half-metallic conductor, although the spin-polarization of the conduction bands is quite strong (5 m bands vs. 2 M bands).

The change in the electronic structure of the one-dimensional NiO chain on the GGA level with respect to the B3LYP results can be explained by the insufficient cancellation of the self-interaction by the GGA exchange functional, which causes the occupied Ni $3d$ orbitals to artificially rise in energy.

Finally, we have also calculated the electronic structure and transport properties of the single-oxygen atom bridge suspended between the tip atoms of a Ni nanocontact making up a Ni-O-Ni nanobridge. Fig. 1(b) shows the transmission per spin-channel calculated with GGA for the Ni-O-Ni nanobridge shown in the inset of Fig. We have checked that the results are stable with respect to the size of the pyramidal Ni tips intervening the Ni-O-Ni nanobridge and the semi-infinite Bethe lattice electrodes. Surprisingly, the transmission does not look very different from the transmission calculated with B3LYP for that case. Near the Fermi level the transmission is strongly spin-polarized: The transmission of the M channel is strongly suppressed while in the m channel essentially two perfectly transmitting channels contribute to the conductance.

This seems to be at odds with the band-structure calculated for the infinite one-dimensional chain, which suggests

that there should be 5 m- and 2 M channels contributing to the overall conductance. However, the geometry of the Ni nanocontact blocks the transmission of just these channels which due to the unphysical self-interaction of GGA have been raised to the Fermi level. Indeed, an orbital eigen-channel analysis [8] of the transmission reveals that only the doubly-degenerate band of type (d_1) contributes to the conductance of the m channel just as in the case of the B3LYP functional. The electrons in the flat m band of type (d_2) which actually crosses the Fermi level in the ideal case of the infinite chain are easily scattered as they present strongly localized electrons, and thus do not contribute to the overall conductance. The other m channel composed of Ni $3d_{3z^2-r^2}$ orbitals does not contribute either to the conductance since the symmetry of the orbital is not compatible with the geometry of the two Ni electrodes - a mechanism to which we have referred to in previous work as *orbital blocking* [9]. The small but finite conductance in the M channel relates to the doubly-degenerate M band of type (d_1) of the infinite NiO chain raised to the Fermi level due to the self-interaction error. The (d_1) band only crosses the Fermi energy near the upper band edge where the band becomes flat. Thus the electrons in this channel are quite susceptible to scattering near the Fermi level resulting in a low transmission.

Conclusions- In summary, we have performed GGA calculations of the electronic structure and transport properties of atomic NiO chains. Due to the insufficient cancellation of the self interaction by the GGA exchange functional the density of states at the Fermi level is considerably higher for GGA than for B3LYP and consequently the GGA conductance of the ideal infinite NiO chain is much higher than the B3LYP conductance. This points to the importance of correcting the self-interaction inherent in GGA for nanotransport calculations. However, in the more realistic case of a short chain suspended in a nanocontact just those channels which have been raised to the Fermi level by the artificial self-interaction are blocked by the scattering at the contacts. Due to this coincidence the GGA conductance is overall quite similar to that of B3LYP.

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