Spin-selective transport phenomena in helical molecular wires

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Background

Recently various spin-selective phenomena have been reported in transport through helical molecular wires attached to metallic leads[1], [2], [3], see Fig. 1. There is a general consensus that the spin selectivity results due to a combination of the spin-orbit coupling and the helicity. Theoretical understanding of the so called chirality-induced spin selectivity (CISS) is, however, not complete, because most theoretical reports grossly underestimate the magnitude of the effect [4].

We present two approaches to improve the quantitative description of the CISS.

1. Metal-molecular interfaces in the *GW* approximation

First-principles quantum transport simulations rely on an accurate description of electronic charged excitations. The latter are not well described in density functional theory; a natural framework to describe them is Hedin's *GW* approximation. Our calculations show that the *GW* method widens the HOMO-LUMO gap of metallic clusters. We rigorously quantify this observation by investigating an ensemble of disordered metallic clusters [6], see Fig. 2. Our observation has profound impact on the interpretation of standard calculations of molecular adsorbates. Namely, in such studies the infinite metallic surface is always replaced by a finite cluster (with periodic or vacuum boundary conditions). Our results imply that the density of states from the *GW* quasiparticles will be artificially depleted right at the Fermi level.

Therefore, the *GW* method can not be taken as an out-of-the-box improvement over the less accurate density-functional theory.

2. Spin currents in chiral molecular junctions

Considerations based on time-reversal invariance known in the field of spintronics allow for spin-currents in two-terminal coherent devices even in absence of external magnetic fields and magnetic impurities. Such spin-currents exist even in linear response if the device hosts more than one scattering channel. We exemplify this in a model of a molecular wire of a helical topology, see Fig. 3 for an example. We provide basic principles for designing molecular junctions that generate sizable spin currents [5]. These principles can be verified by ab-initio calculations and experimental setups.

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References

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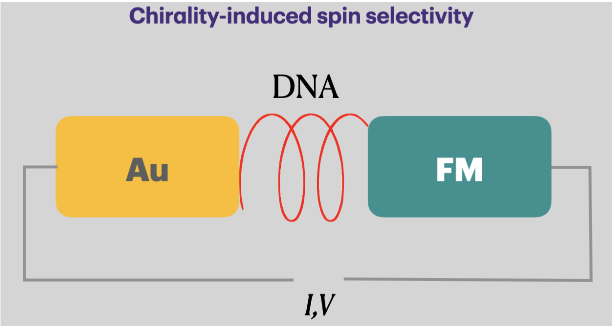

Fig. 1. Typical setup to measure the CISS in electronic current: a non-magnetic lead (Au) attaches to a helical (DNA-like) molecule. The spin selectivity manifests as the the magnetization of the right lead (a ferromagnet) reverses.

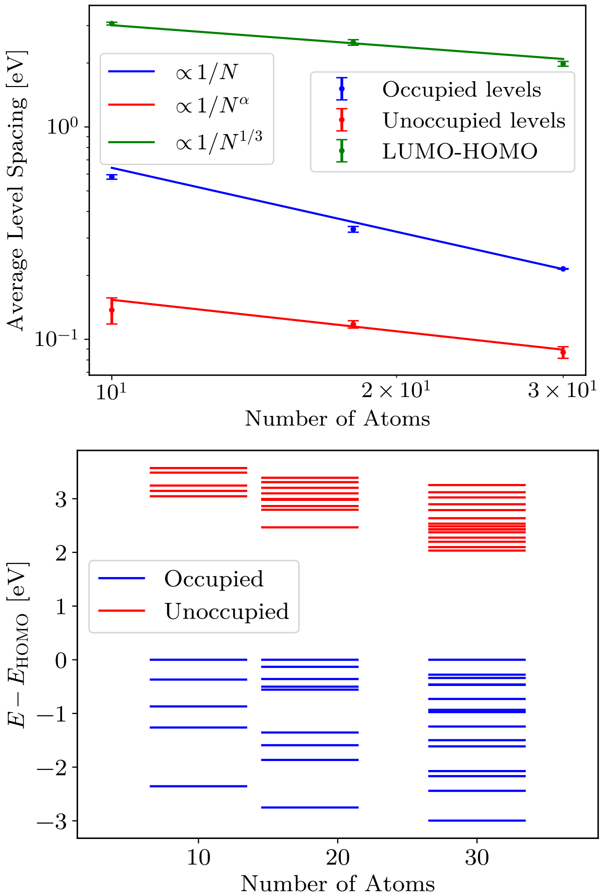

Fig. 2. Scaling of various charged excitations of metallic clusters in evGW. Top: average level spacings and gaps in an ensemble of metallic clusters as a function of the number of atoms. Lines are guides to the eye; they reveal that the HOMO-LUMO gaps scale with a slower power law, leading to HOMO-LUMO widening. Right, the energy levels of sample clusters is shown, with HOMO aligned to 0.

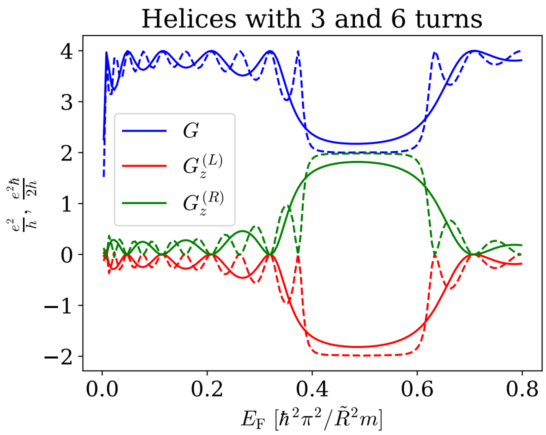

Fig. 3. Charge conductance G and spin left/right spin conductances $G_z^{(\mathcal{L},R)}$ of a model helical wire with 3 (6) turns are depicted by solid (dashed) lines as a function of the Fermi energy. The helix is attached to non-helical leads.