AN *AB INITIO*-STUDY OF ELECTRON TRANSPORT THROUGH NITRO-BENZENE: THE INFLUENCE OF LEADS AND CONTACTS

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The modification of chemical functions on conjugated aromatic molecules has been suggested as an enabling tool for implementing data storage [1] and information processing [2] in molecular electronics. Much attention has been devoted to nitro-groups as suitable ligands [2]-[4], since they generate states inside the HOMO-LUMO-gap of a delocalised aromatic π-system. In some cases this results in peaks in the current/voltage curves or so called negative differential resistance (NDR) behaviour. For an explanation of these peaks many mechanisms have been proposed involving inelastic or two-step tunneling processes through the induced states [4]. However, an accurate picture of the electronic structure of the electrode/molecule/electrode junction and its detailed relation to the conductance through the junction has so far not been provided for this type of molecules. Especially the influence, which the nature of the leads and the way they are connected to the molecules has on peaks in the conductance, which can also be found for calculations within a coherent elastic scattering approach [2], has not been studied so far.

We present *ab initio* conductance calculations within a formalism for coherent electron transport [5] for the simplest molecule in this class, nitro-benzene, connected to two leads in a para-configuration (Fig. 1). Our formalism uses partly occupied Wannier functions as a highly localized and efficient minimized basis set for the transport calculations [6], which still preserves the accuracy of our density functional theory (DFT) calculations. In our DFT-method [7],[8] we use ultrasoft pseudopotentials for the ionic cores and a plane wave basis set for the wavefunctions and electron density for the description of the electronic structure of the junction. By comparing the conductance, density of states and localization properties of molecular orbitals within the same rigorous theoretical framework, we analyze what effect a drastic variation of the leads has on the peak structure of the conductance, if the molecule in the center of the nano-junction is the same. Ín this comparison the molecule is connected by sulfur anchor groups to gold wires (Fig. 1a) and gold (111) surfaces (Fig. 1b), directly bonded to organic (poly-acetylene) leads (Fig. 1c) and to poly-acetylene-fragments anchored to gold (111) surfaces (Fig. 1d).

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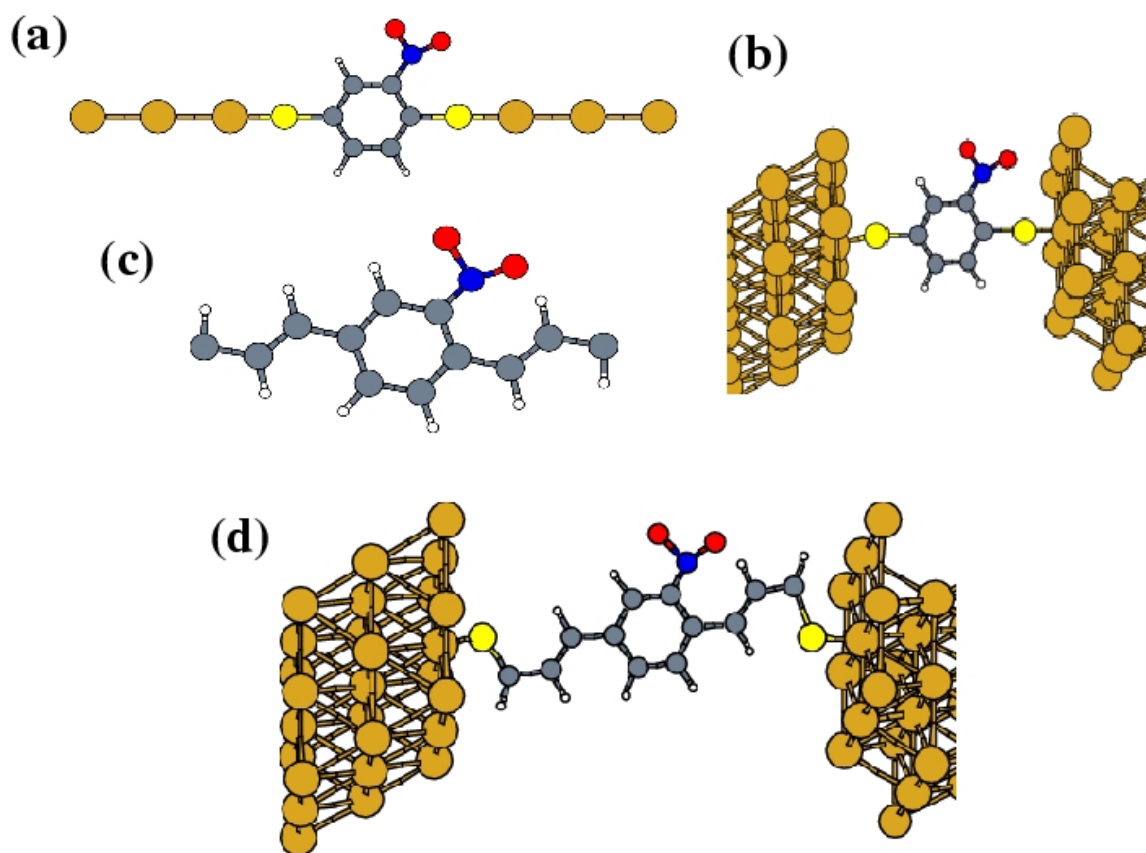
Fig. 1: A nitro-benzene molecule is connected to (a) gold wires by a sulphur anchor groups, (b) gold (111) surfaces by sulphur anchor groups, (c) poly-acetylene wires and (d) a combination of (b) and (c). The colour code for the atoms in this figure is: hydrogen-white, carbon-grey, nitrogen-blue, oxygen-red, sulphur-yellow, gold-gold (beige).