Molecular dynamics investigation of electron transport through single molecule junctions

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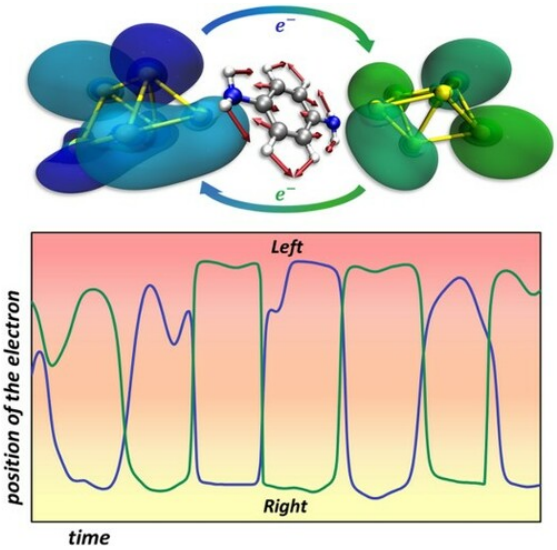

The investigation of molecular electron transport properties plays an important role from the design of molecular electronic devices to the understanding of biochemical processes. One of the main models of the examination of these properties is the so-called single molecule junction (SMJ) in which a molecule is trapped between two metallic electrodes. The theoretical studies of SMJs are mainly based on the Landauer theory in which the electronic structure of the system is usually treated with tight-binding models or density functional theory (DFT). The two main shortcomings of the Landauer model are the poor description of the electron-electron interaction and the lack of vibronic interactions.

In this work we interpret the electron transport through SMJs using *ab initio* quantum chemistry methods and investigate the role of molecular vibrations in the process. For this reason, *ab initio* quantum-classical molecular dynamics simulations are performed for the electron attached states which were calculated with CC2 and ADC(2) methods using the continuum orbital strategy. Two model systems, built from the benzene-1,4-diamine (BDA)[1] and the 1,4-diazabicyclo[2.2.2]octane (DABCO) molecules and gold clusters are studied, the first molecule being known as a good conductor, and the second one as an insulator. The different behavior of the two systems made it possible to connect the determined quantities to the quality of the conduction.

A periodic oscillation of the electron's position between the two electrodes is observed during the simulations in both systems, while the electron does not appear on the molecule significantly. The connection between the molecular vibrations and the periodic behavior of this quantity is investigated via normal mode analysis, allowing the identification of the vibrations which cause this effect. In both systems these vibrations have the same character, dominated by the two amine groups' opposite displacements towards the electrodes. The oscillation mechanism corresponds to the Landauer picture from which a tunneling mechanism was suggested previously for SMJs with small molecules. With the help of the parameters of the oscillation the quality of the molecular conductance can also be classified, therefore the model is able to distinguish the insulator and the conductor molecules. These are expected to be used for the quantitative prediction of molecular conductance with high-level *ab initio* wave function models.

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[1] Jelenfi, D. P.; Tajti, A.; Szalay P. G. First-principles interpretation of electron transport through single-molecule junctions using molecular dynamics of electron attached states *Mol. Phys.* **2021**, *119*, 21.