10:45 – 11:30

First-principles modelling of charge transport in molecular junctions: Redox chemistry and quantum interference

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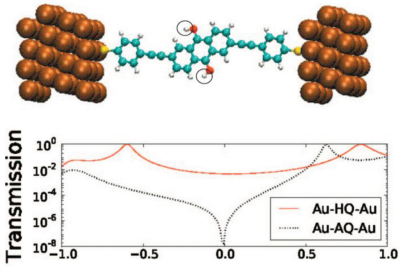

Controlling charge transport through a single molecule junction, i.e. a single molecule connected to source and drain electrodes, remains a long standing goal of molecular electronics. One way of tuning the transport properties of the molecule is to use an "electrochemical gate" thereby overcoming the technical challenges of incorporating gate electrodes for solid-state molecular devices. For redox active molecules, this approach can be used to switch between different redox states of a molecule. Of particular interest are then molecules exhibitng large differences in their transport properties between different redox states. I will discuss how the concept of quantum intereference [1], i.e. the complete or partial cancellation of the electron wave function due to phase difference between different paths through the molecule, can be exploited to realize molecular switches with on/off conductance ratios of several orders of magnitude. In particular, anthraquinone-based molecular junctions which are cross conjugated in the oxidized form and linearly conjugated in their reduced form are interesting candidates for such applications [2, 3]. Another way to tune, or at least influence, transport properties is by varying the electrode material. While most experiments to date have used Au electrodes due to their high stability, electrochemical control can be used to stabilize other types of metals which would otherwise oxidize. I will show how Ni electrodes can (i) yield better (and qualitatively different) contacts to bipyridine than Au and (ii) lead to highly spin polarized currents through the molecule, i.e. a spin filter. If time allows I will discuss the different but related problem of optimizing dyes for dye sensitized solar cells (DSSC). Our recent computational screening strategy has enabled us to build a library over the atomic structure, orbital shapes and

energies of more than 5000 functionalized porphyrins [4, 5]. Using the energy level alignment with the TiO_2 conduction band and the redox mediator, the optical absorption in the visible, and the energy barrier associated with the regeneration of the dye, we define a "quality factor" that allows us to score all the dyes in the configuration space. With this approach we identify known high efficiency dyes, suggest a number of new candidate dyes, and propose design strategies for improving the efficiency of DSSCs.

References

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