

Electronic and magnetic properties of single-molecule metallocene junctions

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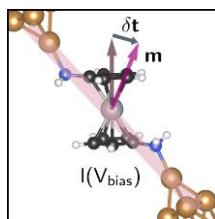
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Abstract:

Metallocenes, a well-known family of organo-metallic molecules in which a transition-metal atom is sandwiched between two organic ring-like ligands, have been recently attracting a renowned interest in view of their utility as robust functional building blocks in molecular-scale devices. In this talk, I will present several recent theoretical aspects associated to these molecules. First, I will show that the configurational degree of freedom of ferrocene (where the metal is iron - Fe) allows to mechanically-tune its conductance in single-molecule junctions. Using *ab initio* transport calculations and simple models, I will prove that ferrocene displays destructive quantum interference effects. By rotation of the ligands, the nature of the quantum interference can be mechanically controlled and the conductance modulated. Next, I will show that these devices also allow for direct interfacial contact between iron and a gold contact at room temperature in the absence of supporting coordinating ligands. This occurs only when the metal center is oxidized to a Fe^{3+} center, which can be induced *in-situ* by a photo-redox reaction, thus laying the foundation for light-switchable ferrocene-based single-molecule devices with potential mechanistic functionality. Finally, I will present a methodological study of how electric fields can be used to control the magnetic moments by non-magnetic means through spin-orbit torques. I will show first-principles calculations of this observable at a single-molecule level in a vanadocene (where the metal is vanadium) - copper single-molecule junction under bias, beyond the linear response regime. Invoking a self-consistency scheme between density functional theory and non-equilibrium Green's functions formalism, we compute the change of magnetization, as well as the associated spin-orbit torque. Our findings lay the foundational work to study spin dynamics at the single-molecule level in this and other promising molecular complexes.

The research presented here was performed in collaboration with M. Camarasa-Gómez (Universidad del País Vasco/CFM, Spain), F. Evers (Universität Regensburg, Germany), the group of L. Venkataraman (Columbia University NYC, USA), M. S. Inkpen (University of Southern California, USA).



Vanadocene-copper single-molecule junction, taken from Ref. [3]

References

- [1] M. Camarasa-Gómez*, D. Hernangómez-Pérez*, M. S. Inkpen, G. Lovat, E-Dean Fung, X. Roy, L. Venkataraman, and F. Evers, *Mechanically-tunable quantum interference in ferrocene-based molecular junctions*, Nano Lett. **20**, 6381 (2020). (* = equally contributing authors)
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[3] M. Camarasa-Gómez, D. Hernangómez-Pérez, and F. Evers, Spin-orbit torque in single-molecule junctions from ab initio, *J. Phys. Chem. Lett.* **15**, 5747 (2024).