Optoelectronic properties of single-molecule junction

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Electroluminescence of a single molecule can be induced by means of scanning tunneling microscopy (STM). However, due to electronic coupling between the molecule and the surface, the light emitted does not correspond to the intrinsic fluorescence of the molecule. Intrinsic fluorescence is only obtained when the molecule is decoupled from the electrodes, e.g. by an insulating layer and/or vacuum [1]. However, if we envision any kind of electronic circuit configuration, direct contact of the molecule with the electrodes (tip and substrate) is required, which leads to a broadening [2] or quenching [3] of the fluorescence peaks.

We propose here an alternative bottom-up approach to realize a π-conjugated molecular chain through on-surface co-polymerization, constituted by a chromophore embedded between decoupling molecular wires. The tip of a low temperature STM is then used to lift the wire and to decouple the chromophore from the electrodes while keeping a circuit-like configuration. Tunneling current, generated by applying a bias between the electrodes, allows to excite the chromophore molecule which exhibits a narrow emission line [4] (FWMH= 2 meV).

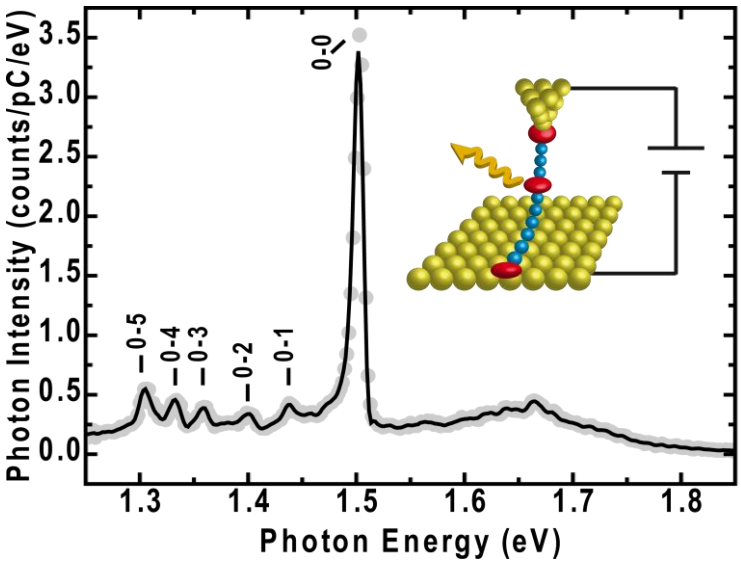

Figure 1 : Characteristic light emission spectra of a suspended molecular wire. In the inset: scheme of the experimental concept.

The plasmonic environment created by the golden tip and substrate plays a major role in the whole STM induced electroluminescence process. Here we report some insights on a mechanism, unreported at the single-molecule scale, implying excitation and de-excitation via interaction with the localized surface plasmons.

We show that this configuration allows to control the lifetime of the single molecule excited state (i.e. the coherence of the emitted light) over two orders of magnitude. Our system might open the way to the realization of controlled molecular transducers between electronic and plasmonic signals.

[1] X. H. Qiu, et al., *Vibrationally Resolved Fluorescence Excited with Submolecular Precision*, Science **299**, 542 (2003)
[2] G. Reecht, et al., *Electroluminescence of a Polythiophene Molecular Wire Suspended between a Metallic Surface and the Tip of a Scanning Tunneling Microscope*, Phys. Rev. Lett **112**, 047403 (2014)
[3] N. L. Schneider, et al., *Light Emission Probing Quantum Shot Noise and Charge Fluctuations at a Biased Molecular Junction*, Phys. Rev. Lett. **109**, 186601 (2012)
[4] M. Chong, et al. submitted for publication