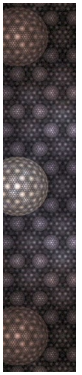


Optical properties at the single-molecule scale: Tunnel Electroluminescence with STM

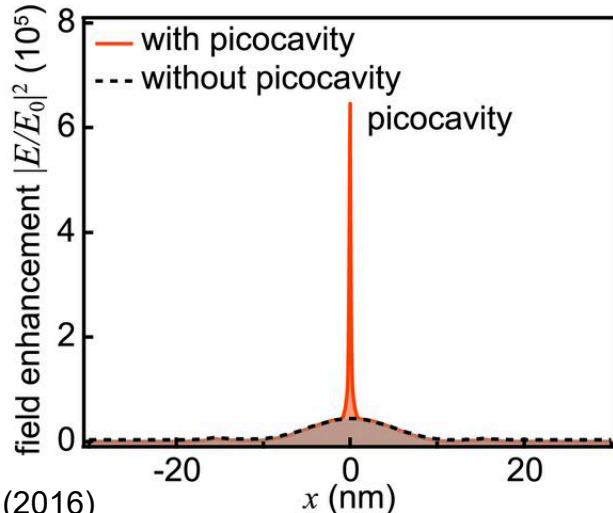
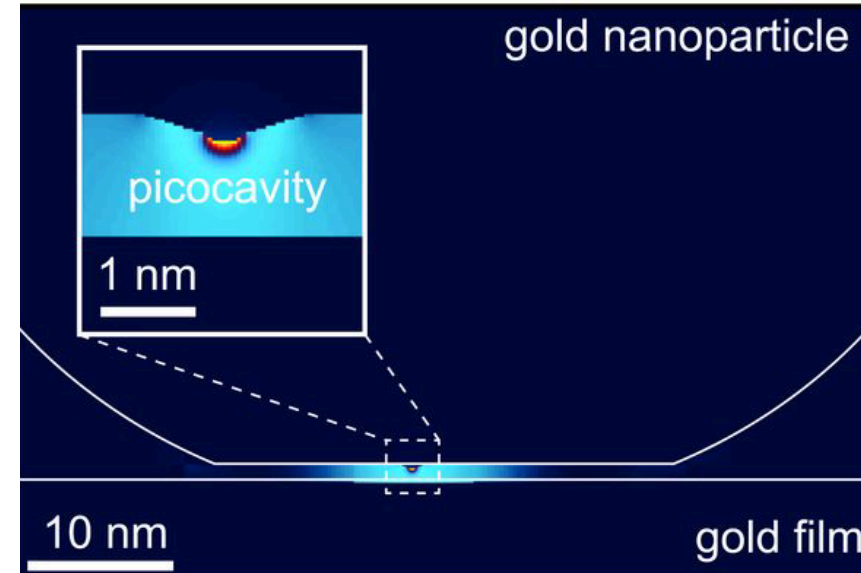
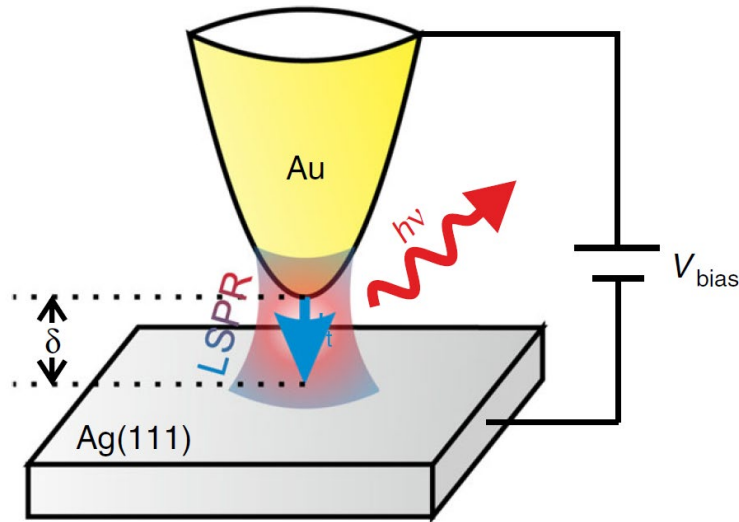
Roberto Otero

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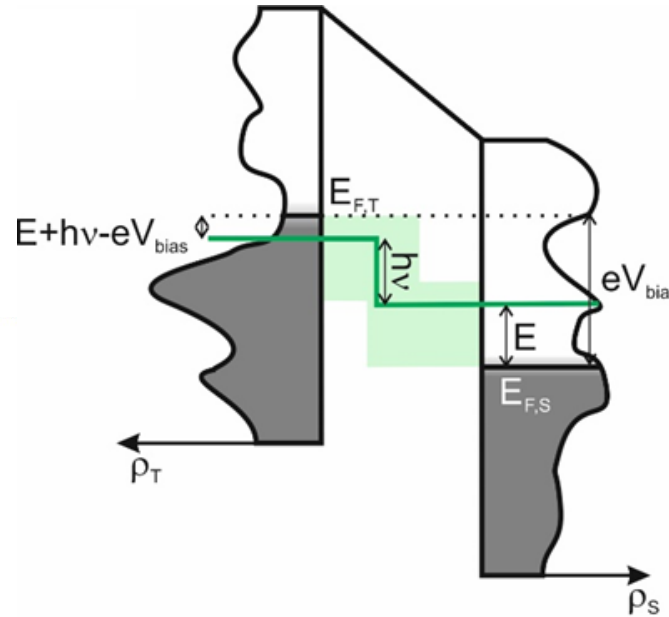
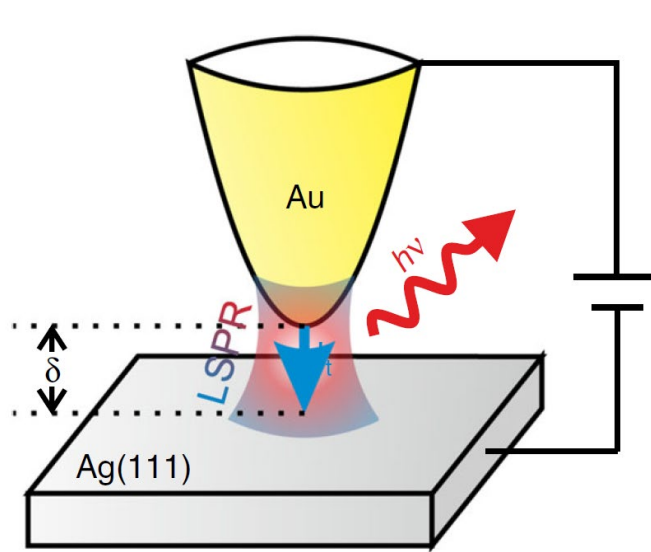
The STM junction as a Nanoparticle-on-Mirror plasmonic cavities



Tunneling junction = tunable, electrically-driven, NPoM plasmonic cavity.

Extreme field enhancement.

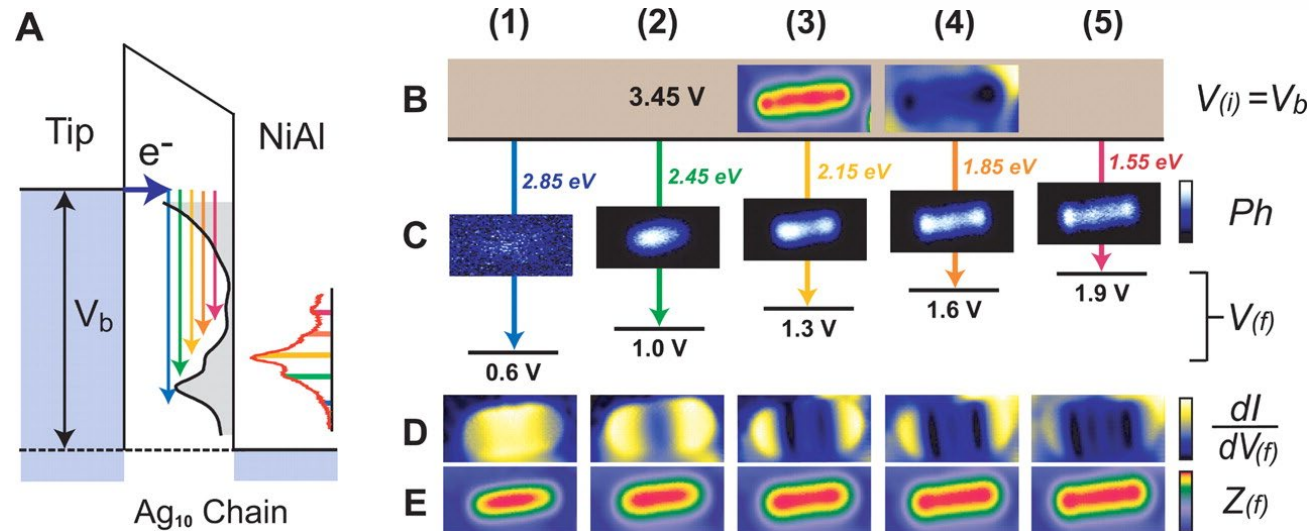
The light at the end of the tunnel



Tunnel cathodoluminescence:
Inelastic excitation of cavity
modes.

Tuneable plasmonic NPoM cavity

BUT influence of tip and surface electronic structure

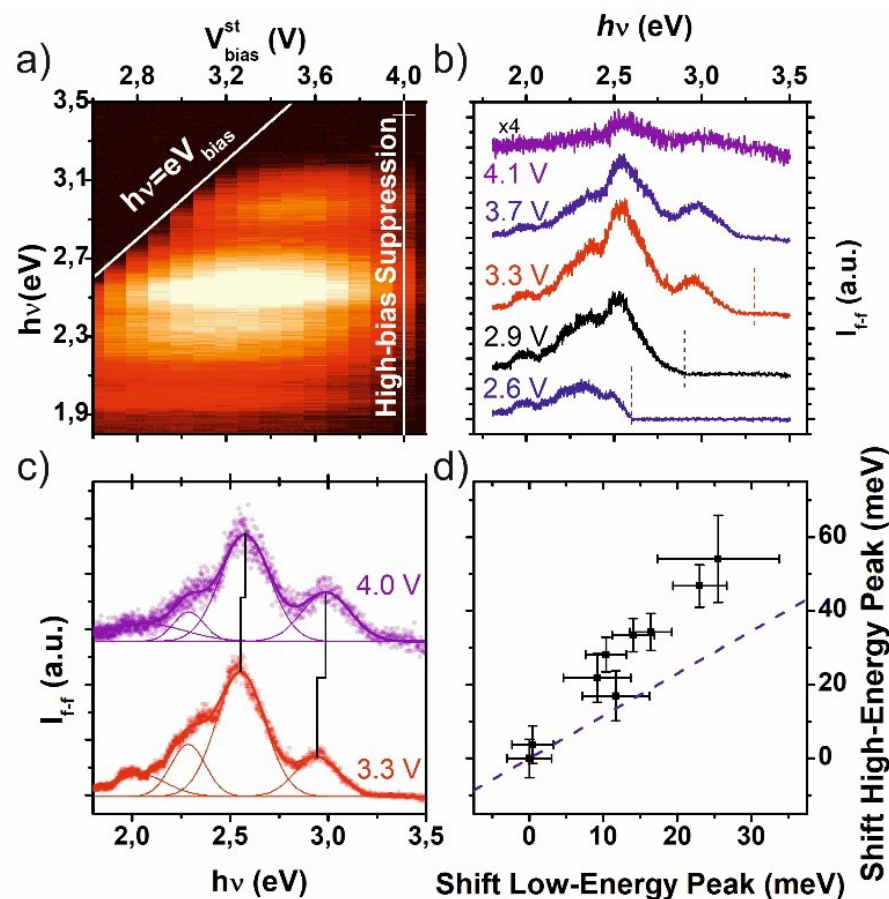


How to remove electronic structure factors?

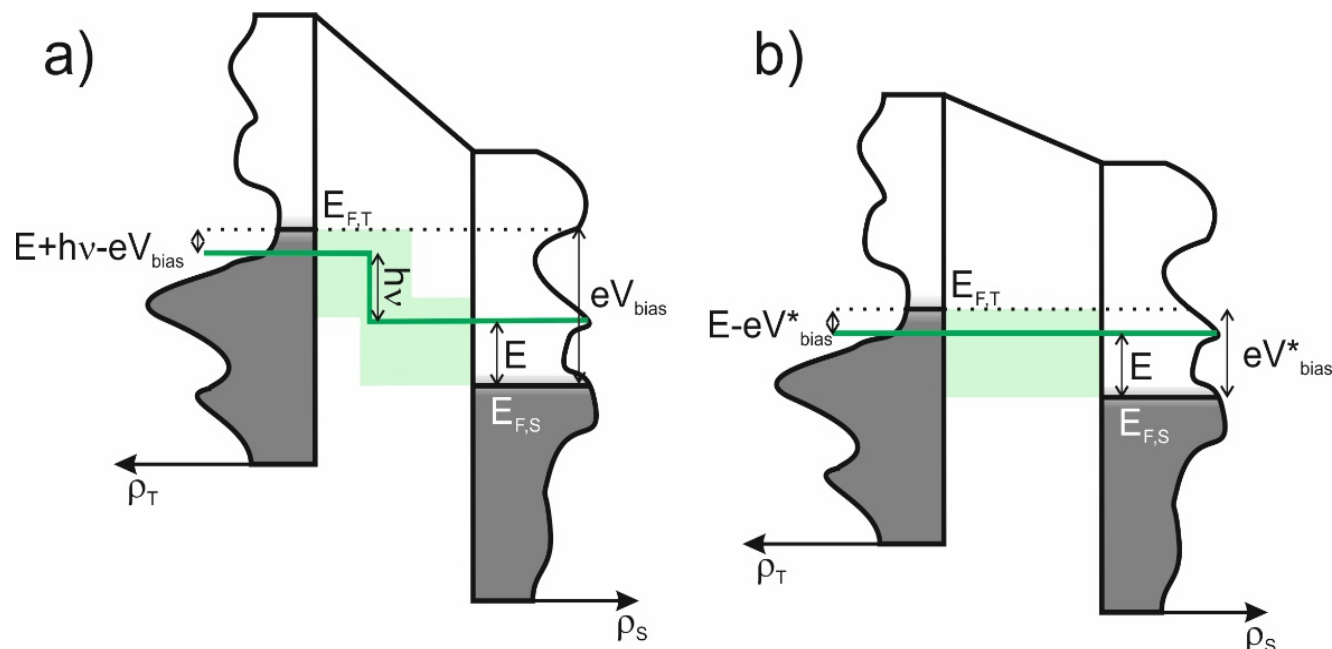
Science **325**, 981 (2009).



True optical properties from STML



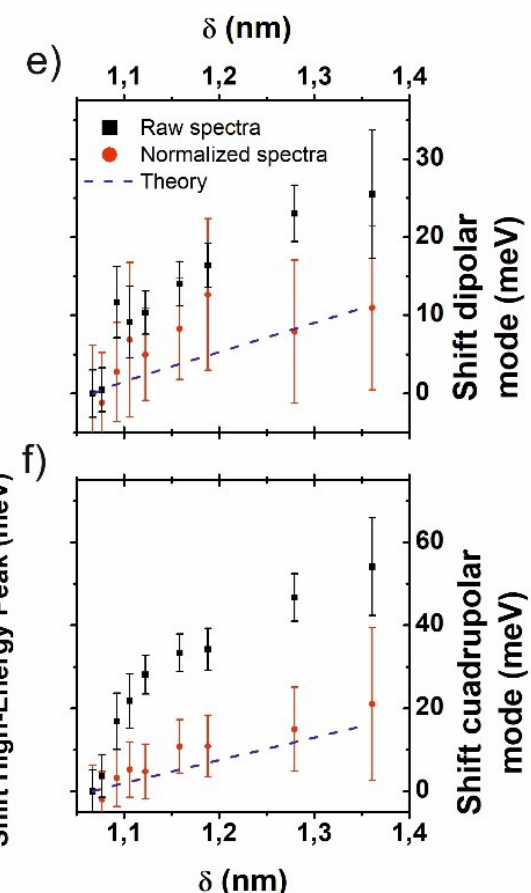
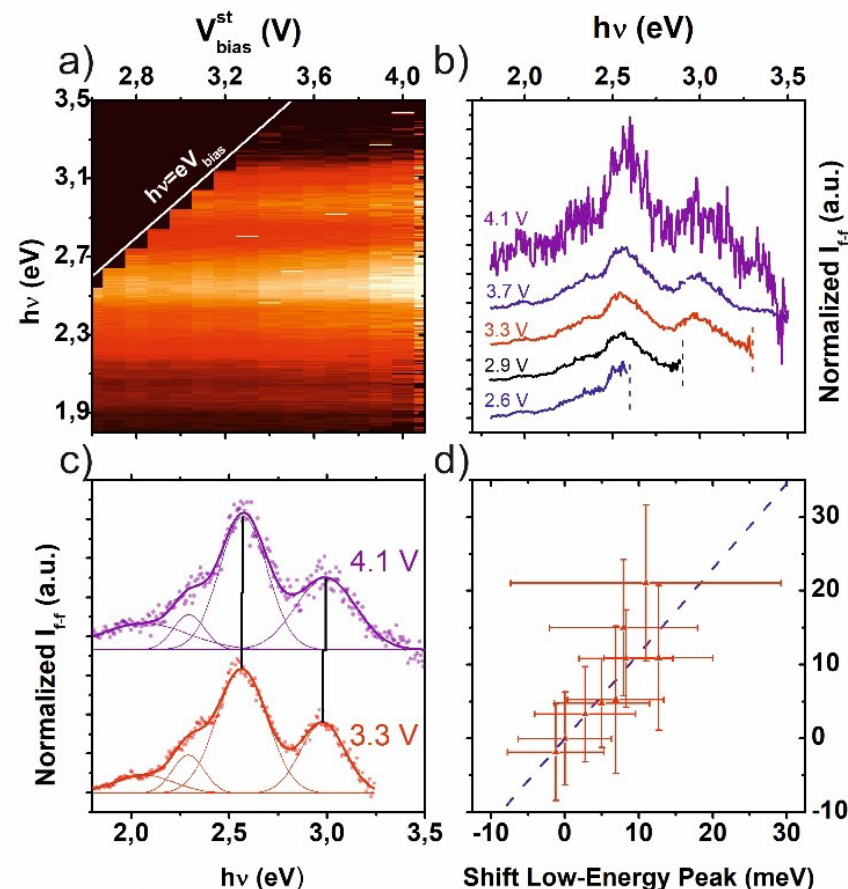
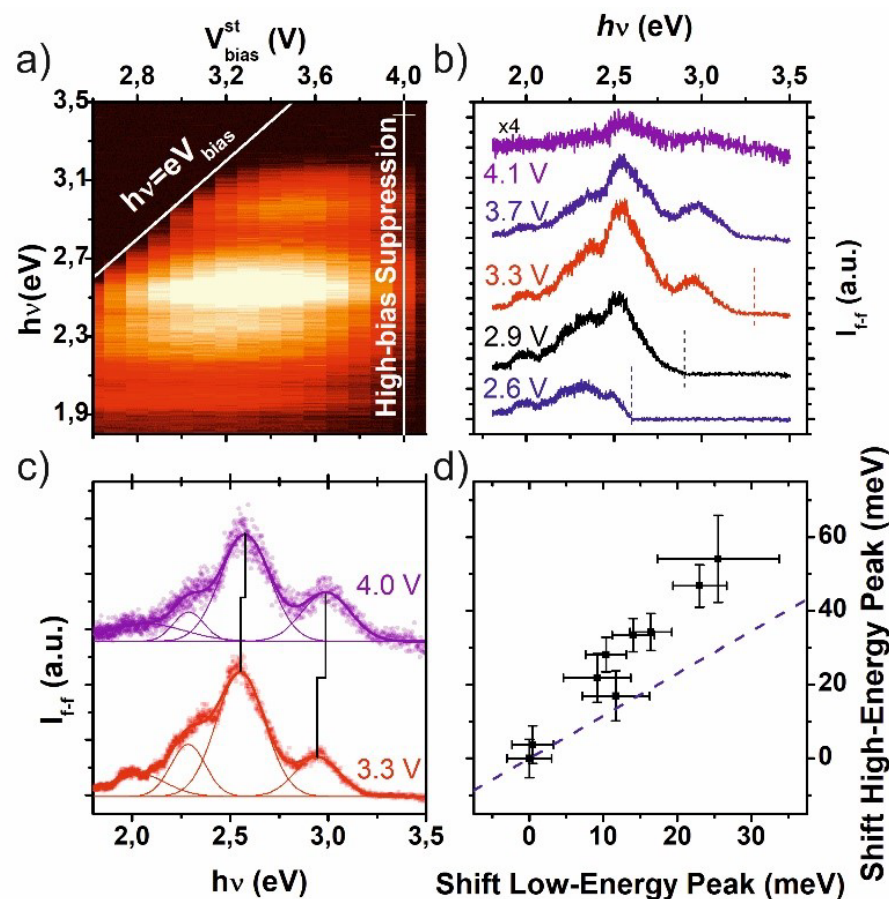
Raw data: Disagreement with EM calculations



“Unveiling the radiative local density of optical states of a plasmonic nanocavity by STM”, A. Martín-Jiménez, A. I. Fernández-Domínguez, K. Lauwaet, D. Granados, R. Miranda, F. J. García-Vidal* and **R. Otero***, *Nature Communications* **11**, 1021 (2020)



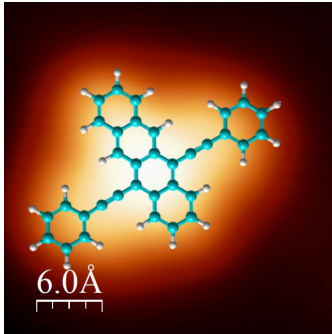
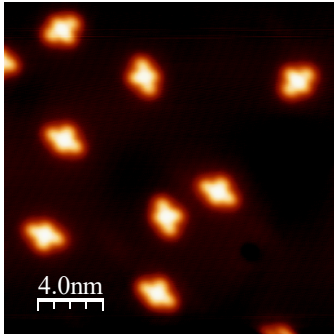
True optical properties from STML



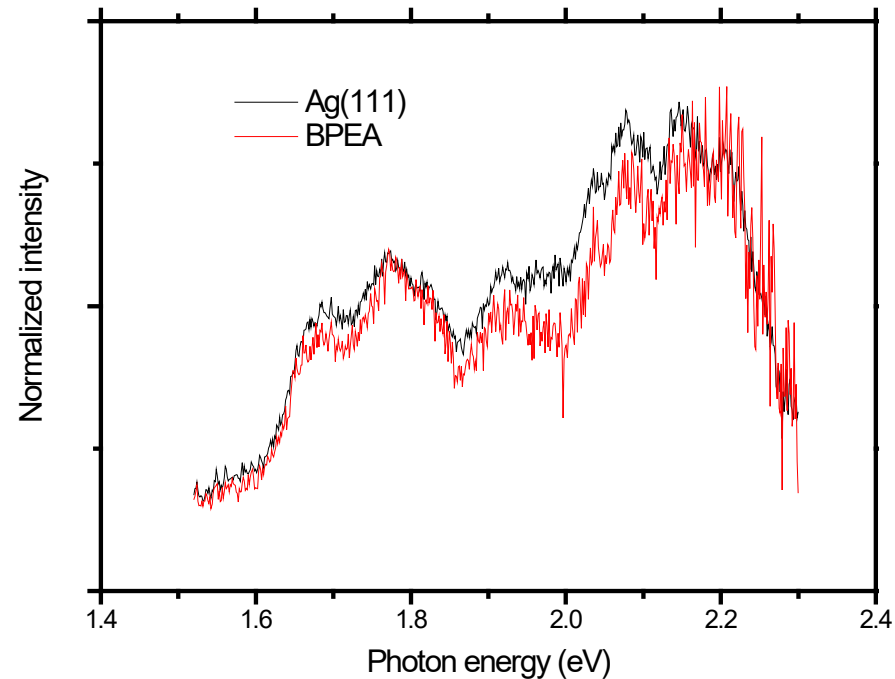
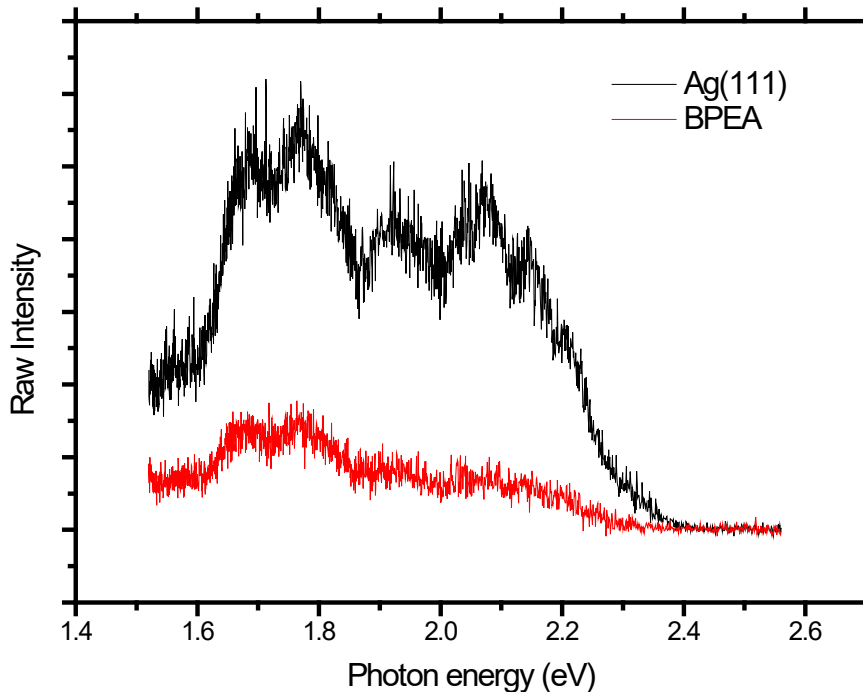
Normalized data: Agreement with EM calculations

“Unveiling the radiative local density of optical states of a plasmonic nanocavity by STM”, A. Martín-Jiménez, A. I. Fernández-Domínguez, K. Lauwaet, D. Granados, R. Miranda, F. J. García-Vidal* and **R. Otero***, *Nature Communications* **11**, 1021 (2020)

Optical properties of single molecules at metal surfaces

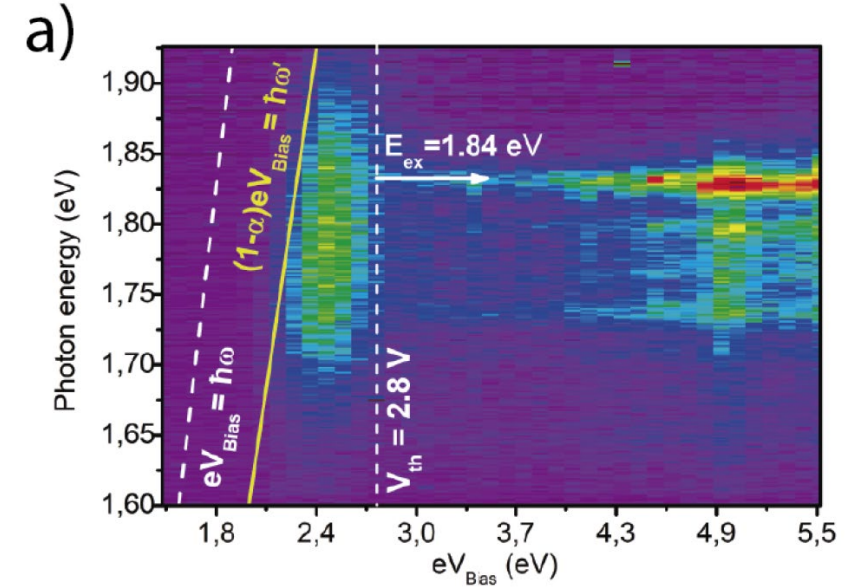
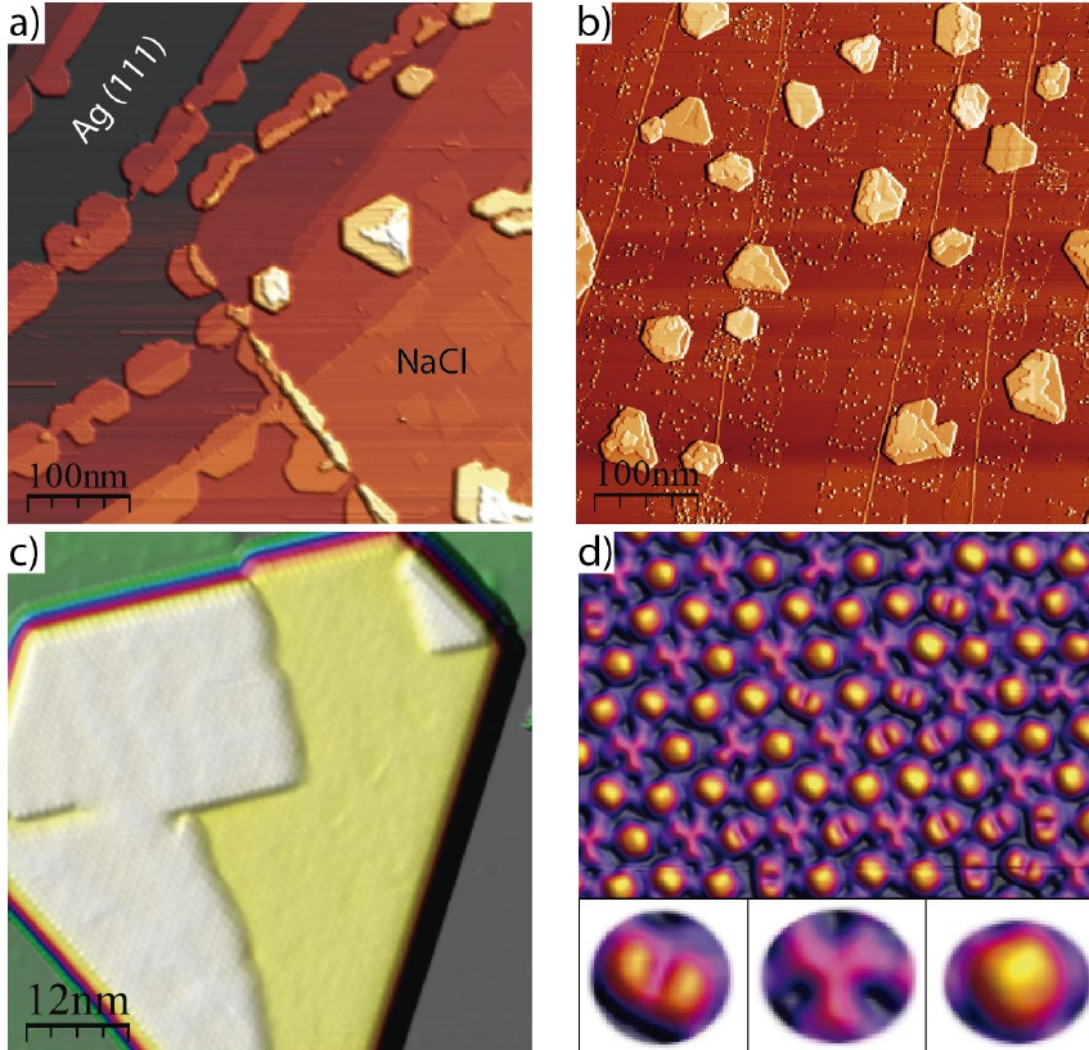


- Normalization is essential to compare optical spectra in inhomogeneous samples
- Transparency bands: access to the optical gap of individual molecules (and its renormalization)?



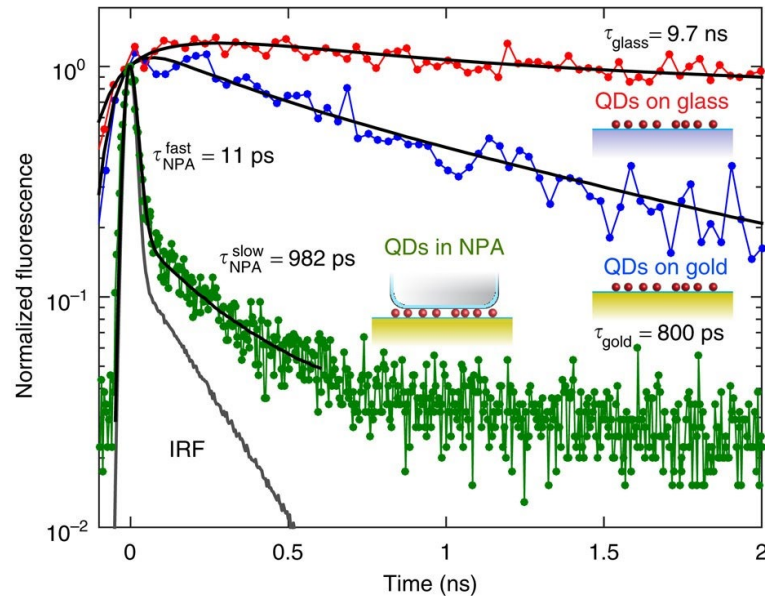


Plasmonic and excitonic emission



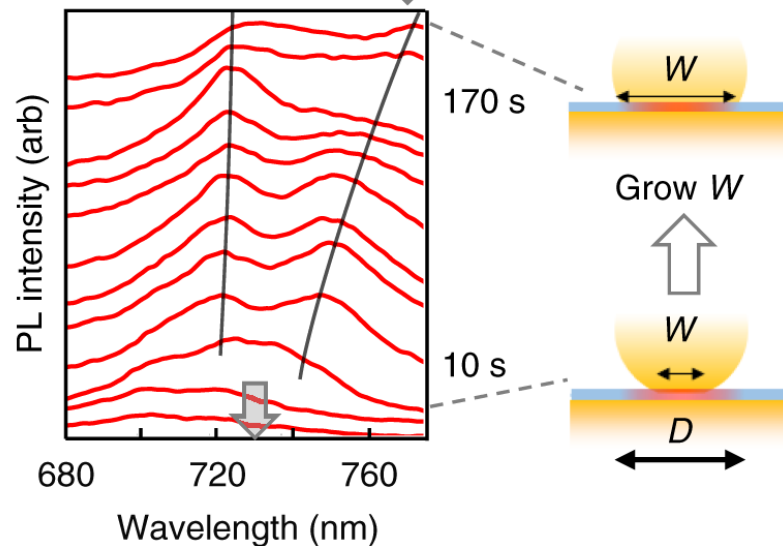
STML luminescence of C_{60} nanocrystallites on NaCl/Ag(111): Plasmonic at low bias, excitonic at high bias.

Light-matter interaction at nanocavities



Nat. Commun. **6**, 7788 (2015)

Polaritonic modes in the strong-coupling regime.
Both bright and dark, but only bright are observable.



Nat. Commun. **10**, 1049 (2019)

How to distinguish bright and dark modes?

Conclusions and outlook

- STM junction as a tunable NPoM plasmonic cavity.
- STM induced luminescence reproduces the radiative photonic density of states with a photon energy-dependent electronic structure factor.
- The electronic structure factor can be estimated (and thus removed) by measuring the $I(V)$ curves of the junction.
- Can we address the modifications in plasmonic properties due to individual atomic or molecular adsorbates
- TFM project: Optical properties of molecular nanostructures on metallic and dielectric substrates