Controlled Switching between Plasmonic and Molecular Luminescence at the Nanoscale

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The fact that light is emitted when a current is injected through a tunnel junction was first found in the 70's by Lambert and McCarthy. Currently it is exploited to analyze the light arising from the local junctions created between the tip and the sample in Scanning Tunneling Microscopes (STM). Different studies on STM induced luminescence of organic molecules directly adsorbed on top of noble metal substrates report a plasmonic origin of the emitted light which originates from the underlying substrate. In order to study the intrinsic light emission from organic molecules a decoupling layer between the molecules and the metal electrode is necessary to avoid radiationless energy transfer from a molecular energy level to the metallic substrate. In these studies, the most commonly proposed mechanism for light emission is that of radiative recombination of an electron and a hole located in two different molecular energy states.

In this contribution we study the light emitted from C60 nanocrystals grown on top of NaCl on Ag(111). We have observed that depending on the concrete tunneling parameters, when injecting electrons onto a C60 nanocrystal, we are able to reversibly switch from a broad plasmonic spectrum to a very sharp molecular resonance of 10meV width. While previous studies have shown a continuous transition from plasmonic to molecular emission as a function of the tip position, here we control the emission type and monochromaticity on a single spot with the tunneling parameters. Our results may be of great importance for applications such as single photon emitters or quantum computing since future devices may need to be able to switch monochromaticity in a controlled fashion.

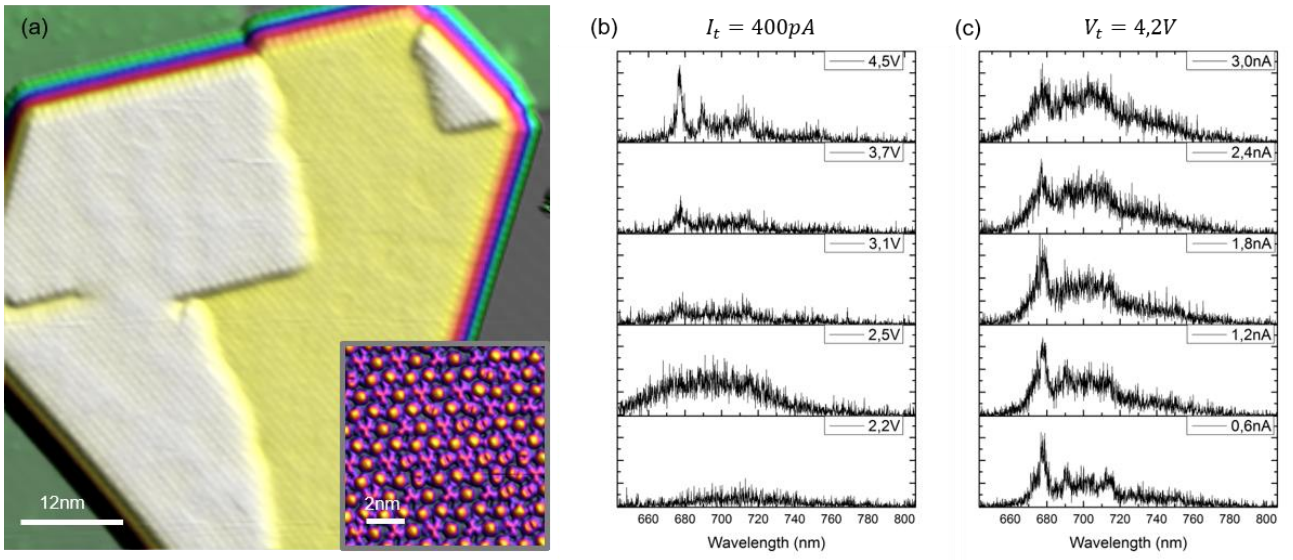

Fig 1. (a) STM topography image of a C60 nanocrystal on top of NaCl grown on Ag(111) (STM parameters: 60x60nm; +2.5V; 30pA). Inset: STM image with submolecular resolution where three orientations of the C60 molecules can be deduced (STM parameters: 10x10nm; +1.5V; 60pA). (b) Series of STM induced Luminescence spectra recorded at a fixed tunneling current at the indicated voltages. Upon increasing the bias, a transition between a broad featureless Plasmonic spectrum and a series of sharp emission lines can be observed (exposure time=100sec/spectrum). (c) Set of spectra acquired at a certain voltage at the indicated tunneling currents. As a result of the increase in current, the molecular sharp peaks become blurred due to plasmon excitation.