Ab-initio simulations of metal clusters

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Our optimal implementation of time-dependent density functional theory within linear response allows computing the optical properties of systems with several thousands of atoms [1,2]. We applied this method to study the dependence of the near-field enhancement and localization on the structural details of the plasmonic nano-gaps [3,4], the different size dispersion of the plasmon resonance of silver and sodium nanoparticles and how this behaviour correlates with the presence of 4d electrons in the Ag case [2], and more recently to describe valence EELS [5].

In this talk I will concentrate mostly in the correlation between transport properties across sub-nanometric metallic gaps and the optical response of the system. In Ref. [6] we presented a study of the simultaneous evolution of the structure and the optical response of a plasmonic junction as the particles forming the cavity approach and retract. Atomic reorganizations are responsible for a large hysteresis of the plasmonic response of the system, which shows a jump-to-contact instability during the approach process and the formation of an atom-sized neck across the junction during retraction. Our calculations show that, due to the conductance quantization in metal nanocontacts, small reconfigurations play a crucial role in determining the optical response. We observe abrupt changes in the intensity and spectral position of the plasmon resonances, and find a one-to-one correspondence between these jumps and those of the quantized transport as the neck cross-section diminishes. These results point out to a connection between transport and optics at the atomic scale at the frontier of current optoelectronics.

References

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[4] M. Urbieta, et al., ACS Nano 12, (2018) 585-595
[5] M. Barbry, P. Koval and D. Sánchez-Portal, in preparation (2018)
[6] F. Marchesin, et al., ACS Photonics 3, (2016) 269-277

Figures

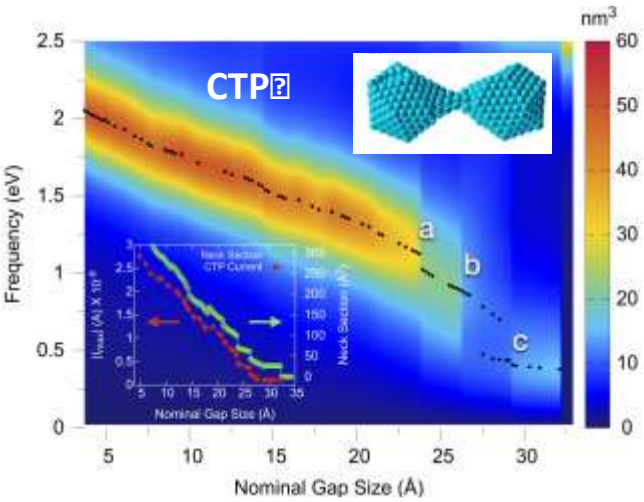

Figure 1: There is a one-to-one correspondence between the jumps in the current flowing through the metal neck, its cross-section and the far-field optical response at the frequency of the so-called charge transfer plasmon (CTP).