Addressing Plasmonic and Vibrational Excitations in Nanoscale Systems within Electron Energy Loss Spectroscopy

Javier Aizpurua

Center for Materials Physics (CSIC-UPV/EHU) and Donostia International Physics Center
Donostia / San Sebastián, Spain

Electron Energy Loss Spectroscopy (EELS) has turned into a very valuable tool to study low-energy excitations in matter. Both collective electronic excitations in metals, so-called plasmons, as well as vibrational fingerprints of solids are now routinely identified and characterized with the use of state-of-the-art microscopes [1], capable to resolve electron energy losses in the range of a few tens of eV. On top of the option to obtain atomically-resolved information of these excitations in bulk materials, a very interesting possibility for EELS emerges in the context of polaritonic excitations in nanostructures, typically probed by optical means. Electron energy losses in nanoscale structures are produced by the action of the structure-induced electromagnetic field acting back on the probing electron. The low-energy loss of such systems contains information on the polaritonic nature of their excitations, which extend along the nanostructure dimensions involving the mesoscopic scale of the system together with its bulk intrinsic properties.

In this contribution, a few cases will be presented where EELS of nanoscale systems is found to provide very relevant information on the nature and properties of polaritonic excitations, with an emphasis on the theoretical description that correctly interprets the spectral fingerprints. First, a system of nanoscale metallic clusters will be presented, where the atomistic details of the clusters are shown to govern the properties of their collective surface plasmon resonances [2]. A comparison between classical and fully quantum mechanical approaches serves to identify atomic-scale features in such nanoparticles and to understand optoelectronic properties within nanogaps. Secondly, we will turn our attention to vibrational excitations in films of Van der Waals materials made of hexagonal Boron Nitride (h-BN) [3], where a complete study of the phonon polaritons driven in nanofilms allow for interpreting the existence of special bulk modes, wave-guided modes, and edge modes driven in these novel nanomaterials. Finally, a study of the effect of temperature and the phonon population of the environment on the properties of the zero-loss peak in EELS is theoretically analyzed in order to interpret experimental data.

All together, Electron Energy Loss Spectroscopy at the nanoscale is shown to be a powerful tool to understand polaritonic excitations in novel materials and devices.

References:

[1] O Krivanek *et al*., Nature **514** (2014), 209.
[2] M Barbry *et al.*, Nano Letters **15** (2015) 3410; M Urbieta *et al*., (submitted).
[3] A Govyadinov *et al.*, Nature Communications **8** (2017), 95.