Vibrational surface EELS probes confined phonon modes

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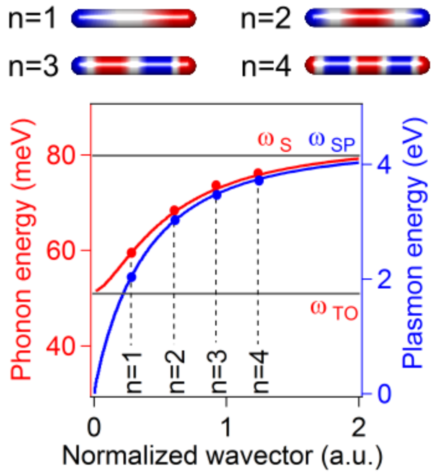
In a pioneering work, Fuchs and Kliewer demonstrated the efficiency of the local continuum dielectric model (LCDM) to describe electromagnetic excitations in a finite system such as slabs [1] and infinite cylinders. Already in these simple systems, the electromagnetic coupling between surfaces induces surface phonon splitting in so-called Fuchs-Kliewer (FK) modes with different charge distribution symmetries. The Fuchs-Kliewer work has been extended with an impressive success to the description of SP in simple systems such as slabs and cylinders. Indeed, given similar electromagnetic boundary conditions, it is no surprise that similar physics is involved; in particular, surface waves, either SP or SPh can be regarded as surface charge densities waves (see figure 1). However, beyond the LDCM, which will not be evoked hereafter, the microscopic origin of the surface charge density waves is rather different at the atomic scale between SPs (free electron charges) and SPhs (ions vibrations).

Recently, two reports [2,3] have demonstrated the amazing possibility to probe vibrational excitations from nanoparticles with a spatial resolution much smaller than the corresponding free-space phonon wavelength using EELS. While Lagos et al. [3] evidenced a strong spatial and spectral modulation of the EELS signal over a nanoparticle, Krivanek et al. [2] did not. In a recent paper [4], we show that discrepancies among different EELS experiments as well as their relation to optical near- and far-field optical experiments can be understood by introducing the concept of confined bright and dark surface phonon modes, whose density of states is probed by EELS. Such a concise formalism is the vibrational counterpart of the broadly used formalism for localized surface plasmons; it makes it straightforward to predict or interpret phenomena already known for localized surface plasmons such as environment-related energy shifts or the possibility of 3D mapping of the related surface charge densities.



[1] Fuchs and Kliewer, Physical Review 140, 1965

[2] Krivanek et al., Nature 514, 2014

[3] Lagos et al., Nature 543, 2017

[4] Lourenco-Martins and Kociak, Physical Review X 7, 2017

Figure 1. (Top) Surface charge densities associated with the four first surface phonon (or equivalently plasmon) modes. (Bottom) Comparison between the dispersion relation of surface phonons (red) and surface plasmon (blue).