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Novel insights in optical properties of nanomaterials allowed by high resolution EELS and cathodoluminescence

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I will present recent works that show how increased spectral resolution in EELS and cathodoluminescence, as well as novel experimental designs, allows for the electron microscope to catch up with optical techniques – and maybe even surpass them.

Indeed, several years have passed since the new generation of monochromators have demonstrated sub-10 meV zero-loss peak FWHM [1]. As a reduction of the zero-loss peak FWHM conducts to both an increased energy resolution and an access to the low energy (far infra-red) part of the optical range, these monochromators have triggered an intense effort to reach the atomic scale vibrational mapping, only very recently achieved [2]. Beyond these remarkable achievements, less attention has been devoted to the interest of those monochromators to get new information on the optical properties of nanomaterials.

I will therefore first present results on the optical response of surface phonon polaritons in the mid to far-infrared range. Indeed, we have successfully performed the reconstruction of their vectorial electromagnetic local density of states in three dimensions [3] (see Figure 1). This reconstruction was based on a novel eigenmode-based analysis of tomographic spectral imaging series of MgO cubes [4]. I'll then turn to the combined measurement of EELS and cathodoluminescence of WS_2 monolayers in the visible range. Although combined EELS and cathodoluminescence have proved to be possible and full of insights for plasmons [5], the required spectral resolution to resolve optically relevant features in novel materials such as TMDs monolayers could not be reached. I will show how close we are now to optical experiments, and what new knowledge the tens of nanometer spatial resolution brings us [6].

Finally, I will introduce new theoretical developments taking fully into account the quantum nature of the incoming and outgoing waves as used in an EELS experiment. In particular, by drawing a formal analogy with optical polarization (see Figure 2), I will show how the quantum nature of the electron can be used to tackle the measurement of optical activity at the nanometer scale [7].

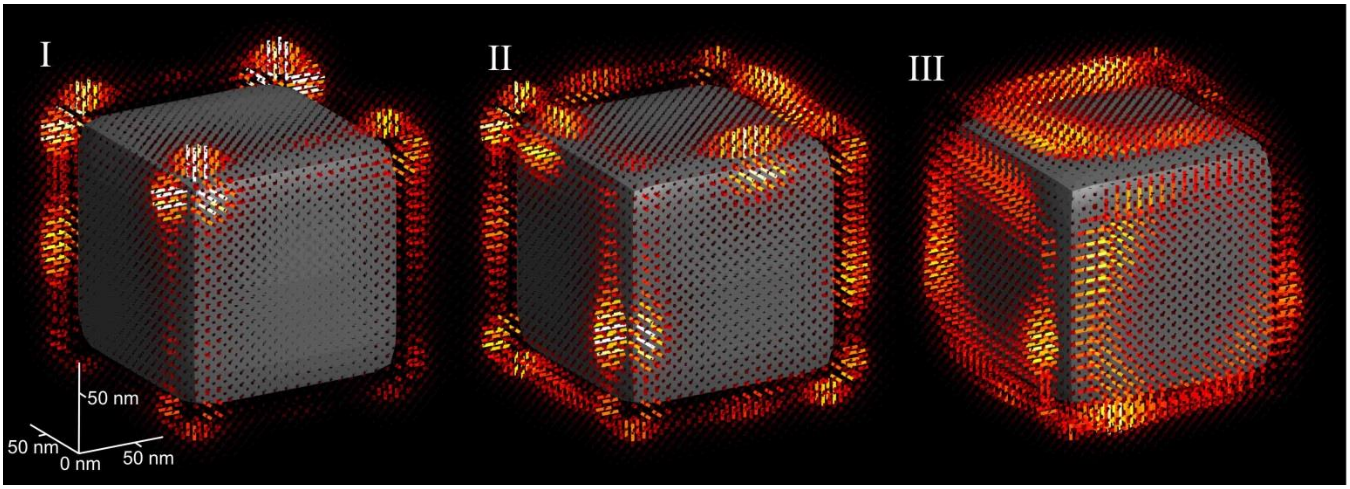

Figure 1. Three-dimensional reconstruction of the surface phonon EMLDOS of a MgO cube for the three main modes (adapted from [4])

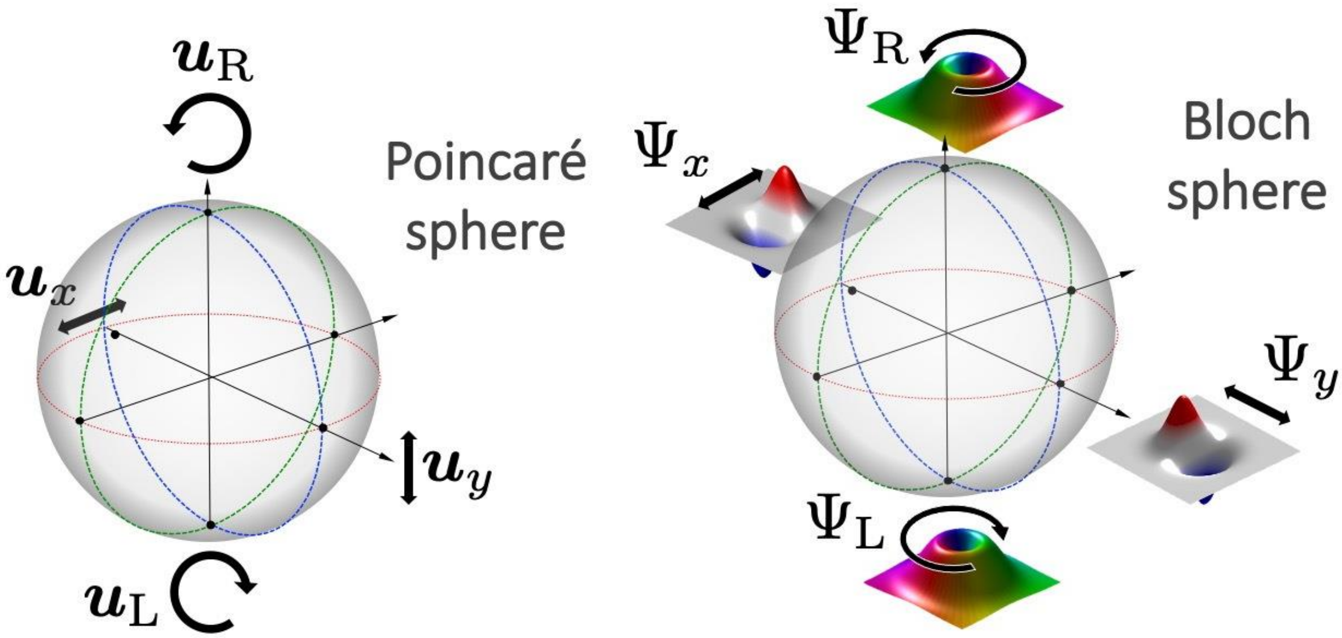

Figure 2. Comparison between the Poincaré sphere (optical polarization) and the Bloch sphere (Hermitte- and Laguerre-Gauss functions, adapted from [7])

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