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Modelling Spatially-Resolved Electron Energy-Loss Spectra in the Low-Loss Region

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High-resolution mapping of low-energy excitations such as plasmons, excitons, or phonons and their interaction is required for understanding and engineering optical, electrical, or thermal properties of nanostructured materials. An unparalleled technique for analyzing the low-energy excitations down to the atomic scale is electron energy-loss spectroscopy (EELS) in a scanning transmission electron microscope (STEM), which enables the acquisition of spectra with a few-meV/Å spectral/spatial resolution [1,2].

Recent STEM-EELS experiments have been complemented or even guided by theoretical modelling and predictions. This talk will review two main approaches for simulating low-loss EEL spectra, using either classical electrodynamics or ab-initio methods. Macroscopic electrodynamics calculations are suitable for describing polaritonic excitations, such as plasmon or phonon polaritons that typically emerge in metals or ionic crystals, respectively. We will discuss examples of spatially-resolved EELS of electron-beam excitation of phonon polaritons in hexagonal boron nitride [3] and an engineered system sustaining strongly-coupled plasmon and phonon polaritons (see Fig. 1) [4].

In addition, STEM-EELS of molecular systems or momentum-resolved EELS requires modelling at truly microscopic level. As an example, we will theoretically describe vibrations in a hBN-like molecule as probed by a tightly focused electron beam. The results obtained with assistance of ab-initio calculations demonstrate that a single isotope impurity in this molecule would significantly affect the EELS probability (Fig. 2) [5]. Finally, we will discuss possible future directions in STEM-EELS, considering both theoretical and experimental efforts [6].

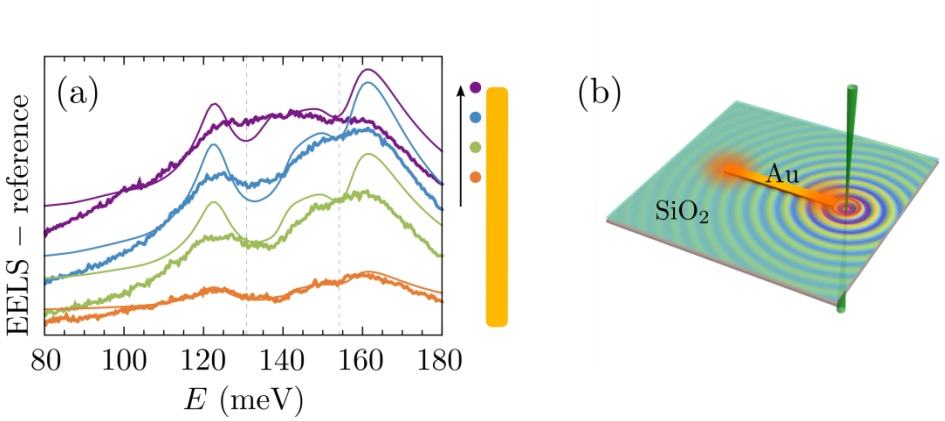

Figure 1. (a) Comparison of post-processed experimental and simulated electron energy-loss spectra (thick *vs* thin lines) for different electron-beam positions along a 3-μm-long gold antenna as marked by the color-matched dots in the schematics. The antenna is fabricated on top of a silicon dioxide film of 40-nm thickness. As the 60-keV electron beam is scanned along the axis of the antenna, we observe spectral signatures of coupling between its dipolar plasmon and phonon polaritons excited in the substrate, schematically shown in (b).

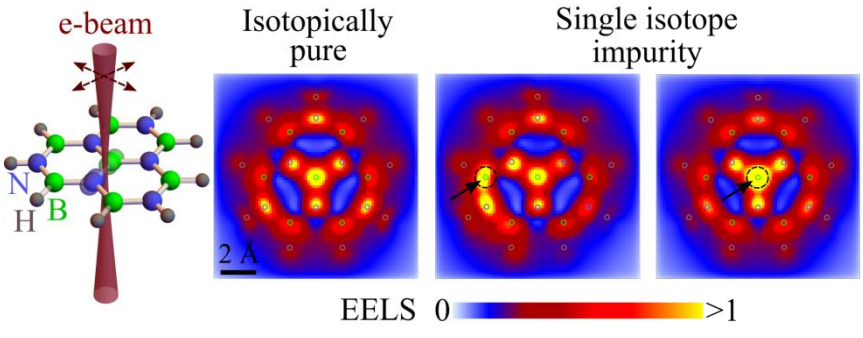

Figure 2. By scanning a tightly focused electron beam across a hBN-like molecule as shown in the schematics, we can obtain spatially-resolved maps corresponding to excitation of different vibrational modes. The maps filtered around ~180 meV energy are associated with an optically active vibrational mode that is highly sensitive to the isotopic purity of boron atoms present in the molecule (impurities marked by black arrows).

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