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Vibrational STEM-EELS Simulations with the FRFPMS Method: Applications and Advances

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Modern monochromated STEM-EELS has enabled the study of low energy excitations at energies of few tens to hundreds of meV by drastically reducing the width of the zero-loss peak and thereby the intensity in its tails [1]. This enables spectroscopic experiments of vibrational excitations, such as phonons, but also polaritons [2], and possibly magnons [3]. The technique allows for high spatial resolution [4] as well as high momentum resolution [5,6,7] and is thus an ideal complement to other existing tools for studies of low energy excitations at the nano- and atomic-scale.

The theoretical description of inelastic electron scattering in the (S)TEM has the unique challenge, that the elastic interaction between electrons and the sample is very strong, leading to a strong thickness dependence of the intensity in individual Bragg reflections. It is therefore imperative to include the effect of this elastic interaction into accurate simulations of inelastic scattering. Furthermore, since waves scattered inelastically on a specific excitation are in theory incoherent with waves scattered on every other excitation, one separate calculation needs to be repeated for every considered transition. In the case of phonons and defect systems, such calculations become quickly unmanageably expensive due to the sheer number of transitions. For such systems, we introduced the Frequency-Resolved Frozen Phonon Multislice (FRFPMS) method, which sidesteps the problem by requiring no explicit knowledge of the phonon modes [8,9]. The method boils down to choosing a grid of frequencies, for which one wishes to calculate the inelastic spectrum, and then to perform a set of Molecular Dynamics (MD) simulations, in which a specific type of thermostat predominantly excites each of these vibrational frequencies. For each frequency a full frozen phonon multislice method calculation is run using snapshots sampled from the MD trajectories and the inelastic contribution is then extracted by suitably averaging the beam exit wave functions akin to Ref. [10].

In this contribution, we follow up on our previous year's contribution [11] and consider simulations for a planar defect [12], which corroborate experimental findings of nano-scale variations in vibrational STEM-EELS near defects [13], both with atomic-size beams, but also with a nano-size beam. Furthermore, we consider some methodological advances: the improvements in spectra and momentum-resolved EELS when switching the interatomic potential to a Machine Learned GAP potential [14] and the influence of tuning the parameters of the Generalized Langevin Equation thermostat in the Molecular Dynamics simulation used to generate the frequency-dependent sets of snapshots [15].

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