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Spectroscopy at Ultra-Low Energy Losses at Atomic Resolution

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Recent hardware developments in monochromation of the electron beam [1] in scanning transmission electron microscopy (STEM) have opened new domains of physics to the spectroscopic studies at down to atomic resolution. Most notably, excitation of *phonons*, happening in the tens to few hundreds of meV range, has been observed at unprecedented spatial resolutions [2,3,4]. Compared to other spectroscopic methods, high spatial resolution is the primary advantage of STEM. It gives access to local changes in the electron energy loss spectra (EELS) near interfaces or defects, also within this ultra-low energy-loss range [3,4]. The primary objects of interest for STEM-EELS are thus local changes of the low energy excitations.

To predict and properly interpret the experimental data, reliable simulation methods are necessary. Such methods also need to have a favorable scaling with respect to system sizes (number of atoms, N_{at}). For instance, a structure model with an interface or a localized defect needs to be sufficiently large to avoid self-interaction of the defect with its periodic images. As an example, an fcc silicon structure model of 1000 atoms would be a cube of dimensions 2.7^3 nm^3. Realistic sample thicknesses are often an order of manitude larger and lateral dimensions may often need to be larger to contain the spreading electron beam wavefunction in multislice calculations. Thus system sizes of tens of thousands of atoms are often needed.

Transition potential multislice-based approaches for calculation of inelastic scattering [5,6] provide a transparent and accurate way of simulating STEM-EELS of phonons, and similarly with Bloch-waves based approaches [7] they allow to both include electron challeling effects as well as spectroscopic information. Nevertheless, these approaches require explicit knowledge of phonon modes, which requires diagonalization of dynamical matrix scaling with the third power of N_{at}. In the domain of tens of thousands of atoms such approaches quickly become computationally too expensive.

We have recently presented a method named frequency resolved frozen phonon multislice method (FRFPMS; [8]), which has computing costs scaling linearly with N_{at}. We will discuss its capabilities and recent developments. This method is also a starting point for a description of another type of excitations in solid state, in particular, the excitations of a magnetic subsystem called *magnons*. Magnons occupy approximately the same energy range as phonons, however compared to phonons their interaction with electron beam is significantly weaker. Diffuse scattering due to magnons [9] and a semi-classical theory of scattering of electron beams on magnons [10] have been presented recently. We will discuss our recent developments in the theory of magnon scattering in STEM-EELS and routes to potential detection of this type of quasiparticle excitations by monochromated STEM [11].

References:

[1] O. Krivanek et al., Nature 514, 209 (2014).
[2] F. Hage et al., Phys. Rev. Lett. 122, 016103 (2019).
[3] F. Hage et al., Science 367, 1124 (2020).
[4] X. Yan et al., Nature 589, 65 (2021).
[5] B. D. Forbes and L. Allen, Phys. Rev. B 94, 014110 (2016).
[6] C. Dwyer, Phys. Rev. B 96, 224102 (2017).
[7] P. Rez and A. Singh, Ultramic. 220, 113162 (2021).
[8] P. M. Zeiger and J. Rusz, Phys. Rev. Lett. 124, 025501 (2020).
[9] K. Lyon et al., Phys. Rev. B 104, 214418 (2021).
[10] B. Mendis, Ultramic. 230, 113390 (2021).
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