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Paul M Zeiger, Juri Barthel, Leslie J Allen, Ján Rusz

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Simulations of Phonon Spectroscopy in the Impact Scattering Regime – Advances and Applications

Paul M. Zeiger[1,*], Juri Barthel[2,3], Leslie J. Allen[4], and Ján Rusz[1]

[1]*Uppsala University, Department of Physics and Astronomy, Uppsala, Sweden*
[2]*RWTH Aachen University, Central Facility for Electron Microscopy, Aachen, Germany*
[3]*Forschungszentrum Jülich GmbH, Ernst Ruska-Centre (ER-C 2), Jülich, Germany*
[4]*University of Melbourne, School of Physics, Victoria, Australia*

*Corresponding author: paul.zeiger@physics.uu.se

Phonon spectroscopy in the Scanning Transmission Electron Microscope (STEM) has attracted a lot of attention since the introduction of monochromators, which reduced the width of the zero-loss peak to around 10 meV [1], and thereby spawned the field of Ultra-High Energy Resolution Electron Energy Loss Spectroscopy (UHER-EELS). The main excitement in this field is due to an unprecedented combination of excellent spatial resolution in the STEM and decent energy resolution for studying vibrational excitations with typical energies of tens to a few hundreds of meV. Prime examples of exploiting these capabilities are, among others, Refs. [2–4], in which modifications of the vibrational signatures of atoms near different types of defects are investigated.

However, the interaction of the electron beam with the specimen is complex and the interpretation of EELS experiments can be complicated. Often simulations are required to account for dynamical diffraction effects and to understand details of the electron scattering process. Since calculations usually invoke periodic boundary conditions, defect systems are notorious from a theoretical point of view, because of the large super cell sizes, which are required to isolate the defect from its periodic images. This problem becomes very prominent in simulations of phonons around defects, since the computational complexity of phonon calculations scales with approximately the third power of the number of atoms.

We have introduced a method, which sidesteps this unfavorable scaling and scales linearly with the number of atoms, dubbed the Frequency-Resolved Frozen Phonon Multislice (FRFPMS) method [5,6]. While the method has some overhead, this scaling behavior makes it ideal for studying (large) defect systems and we have readily applied the method to a system of hBN, which contains an extended planar defect [7]. The core idea of the method is to divide the range of vibrational frequencies of the material of interest into a number of so-called frequency bins. For each of these bins, one runs a full Frozen Phonon Multislice (FPMS) calculation for sets of atomic structure snapshots, which are sampled from a MD trajectory, in which predominantly the range of frequencies of the considered frequency bin are excited.

In this contribution, we focus on the inelastic phonon scattering process and how it is represented in the FRFPMS method. We first review the conceptual underpinnings of the method briefly and turn our attention then to some of the results in a forthcoming publication [8], in which we consider the FRFPMS method analytically for a toy model in the form of a single Quantum Harmonic Oscillator (QHO). We compare the results to inelastic scattering theory in the form of the Transition Potential (TP) formalism [9] and based on a first-order Born approximation [10], since the FRFPMS method can only be accurate for single phonon scattering. We find, that the original prescription of the FRFPMS method overestimates the amount of inelastically scattered intensity at higher momentum transfers (scattering angles), since it does not take the smearing of the potential by those modes into account, which are not within the range of frequencies of the considered frequency bin. Furthermore the amount of elastic scattering is also overestimated towards large angles. This finding is illustrated in Figure 1. The Figure also shows how this discrepancy in the inelastic and elastic scattering intensities can be remedied for reduced displacements by smearing the potential explicitly with the full Debye-Waller factor, which takes the smearing due to all modes into account. The FRFPMS method revised in such a way results in a much improved description of the single inelastic scattering process and the elastic scattering process.

Finally we apply the revised FRFPMS method to the calculation of the inelastic intensity along certain high symmetry paths in reciprocal space for hBN. Figure 2 displays the results of this calculation and the main visible changes due to the inclusion of the DWF in the revised FRFPMS method are visible at large momentum transfers above 5 $Å^{-1}$ or below -5 $Å^{-1}$. There the inelastic intensity is slightly reduced for the visible bands, but the most prominent feature is a reduction in the background intensity, where no bands appear.

Concluding this brief summary of our contribution, we have developed a better understanding of the phonon scattering process and how it is represented in the FRFPMS method. We have proposed a revision of the method, which should model single inelastic scattering and elastic scattering even more accurately, than the original FRFPMS method does, and also demonstrated these improvements numerically [11].

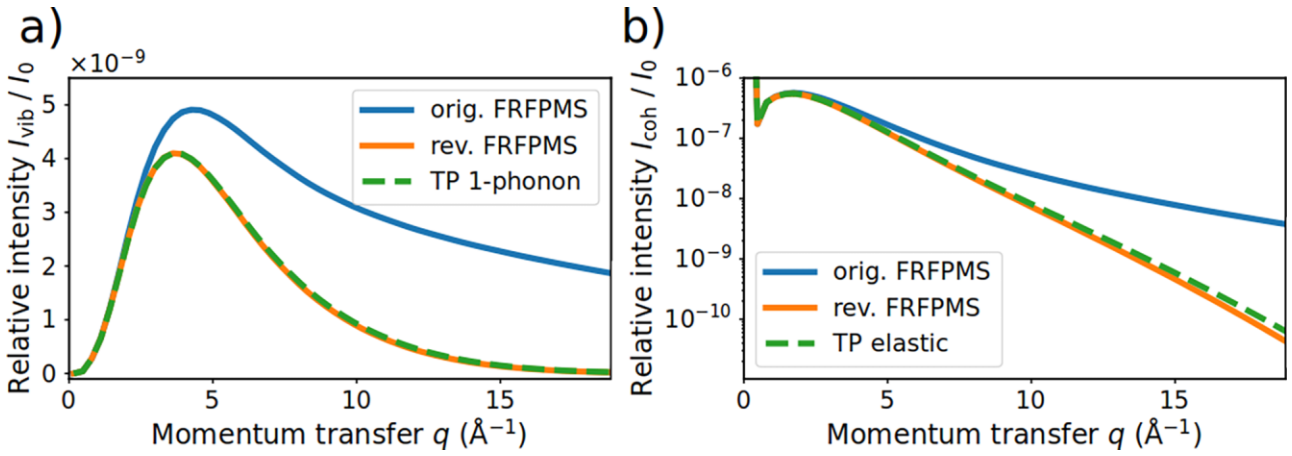

Fig. 1. Scattered electron intensity for a target made of a single carbon atom, whose motion is modeled as a QHO: comparison of the shape of the inelastically scattered intensity in a) and the coherently averaged (unscattered+elastically scattered) intensity in b). Starting with the labels in a) and skipping duplicates, the labels refer to the original FRFPMS method, the revised FRFPMS method, the single inelastically scattered intensity in the TP formalism and the elastically scattered intensity in the TP formalism, respectively.

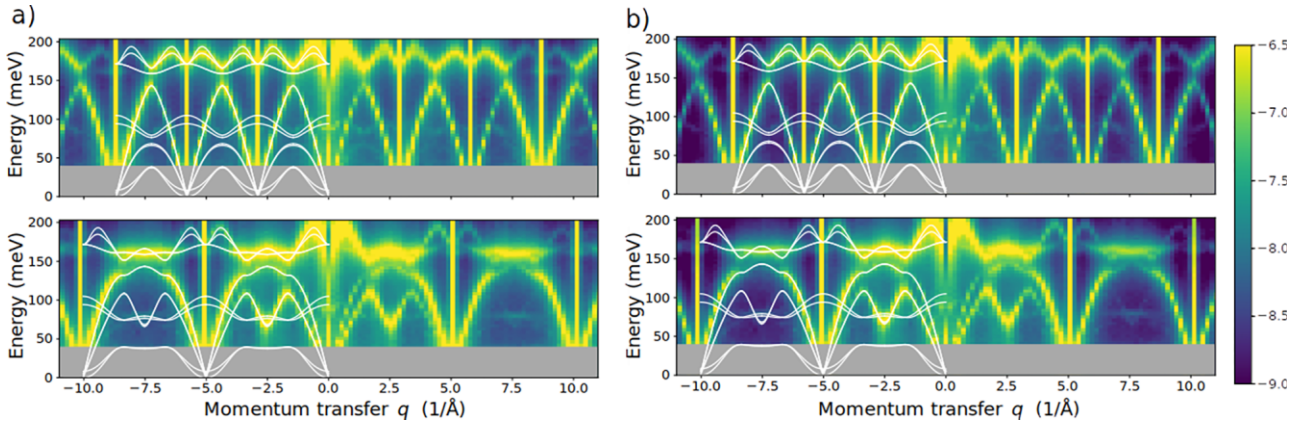

Fig. 2. Plot of the inelastically scattered intensity for the original FRFPMS method (a) and revised FRFPMS method (b) along the Γ-M-Γ direction (top panels) and the Γ-K-M-K-Γ direction (bottom panels) in hBN. The color scale corresponds to the decadic logarithm of the relative intensity.

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