Phonon Scattering Mapping and Temperature Measurement in Nanostructures

M. J. Lagos[1,2], G. A. Botton[1,2], A. Trügler[3], U. Hohenester[3] and P. E. Batson[4]

[1.] Department of Materials Science and Engineering, McMaster University, Hamilton, Canada.
[2.] Canadian Centre for Electron Microscopy, McMaster University, Hamilton, Canada.
[3.] Department of Physics, University of Graz, Graz, Austria.
[4.] Department of Physics, Rutgers University, New Jersey, USA.

Recently, a new technology of aberration-corrected monochromated electron microscopes opened the doors for phonon spectroscopy studies in materials using atom-wide electron probes [1]. The experimental data, however, can present enormous interpretation challenges, requiring a much deeper understanding of the inelastic electron scattering by elementary excitations (phonons, plasmons, etc.) sustained in finite-size objects, probe geometry and collection conditions, probe screening effects, scattering *f*-sum rules, symmetry selection rules, etc. In this work, we present recent progress towards the understanding of phonon excitations in a nanosized volume of matter probed by fast electrons. Our work explores: (i) the excitation of surface and bulk phonon modes within a single nanoparticle and at complex interfaces, and (ii) the measurement of local temperature in single nanostructures using electron energy-loss and energy-gain scattering. We performed spatially-resolved EELS using a ~ 0.15 nm probe and 10 meV energy resolution.

We studied the phonon response of isolated MgO nanocubes and mapped the spatial distribution of both surface and bulk phonon scattering [2]. We detected strong surface phonon polaritons (SPhP) localized near the nanocube surface, extending both within the cube and outside into the vacuum. We also detected bulk optical and acoustic modes, containing lattice contributions spanning the whole Brilloiun zone, and located mainly within the inner regions of the nanocube. Our theoretical results showed that electron scattering by excitation of short-wavelength acoustic phonon modes is localized down to the atom level (*impact scattering*) [3], in contrast to the SPhP excitations, which can be driven by scattering at large impact parameters (*dipole scattering*). The effect of thin substrates on the SPhP response of single particles is also studied [4]. We found that strong image-charge effects could lead to generation of hybrid modes resulting in the suppression of inelastic electron scattering by certain modes of the particles. We also probed SiC/SiO_2 interfaces and found a complex response of optical interface modes in the Reststrahlend bands of those materials, affected by the size and shape of the interface.

In addition, we studied the thermal effects on the inelastic electron scattering by lattice vibrations. We detected energy loss and gain processes whose scattering amplitudes are linked by the Boltzmann factor, as established by the *Principle of Detailed Balancing* (PDB). We measured the nanostructure temperature by plotting the logarithm of the ratio of the scattering intensities on the gain and loss sides of zero energy [5]. The obtained curves are linear for several materials, supporting the use of PDB on the nanoscale, and pointing out a linear universal behavior of the vibrational scattering at certain thermal conditions. Also, this method yields measurements without reference to the nanomaterial morphology, or scattering conditions. Our experimental results obtained at room temperature indicate that the probed nanosystem indeed is close to 290 K with a precision of about 7-10K with ~ 99% of confidence level and agreed very well with the accepted values for room temperature conditions. Similar results were obtained for measurements at high temperatures (up to 1000 K).

Our results provide progress in understanding the excitation of SPhP's and bulk phonons sustained in nanostructures using fast electrons.

References:

[1] O. Krivanek, *et al*, Nature **514** (2014), 209.
[2] M. J. Lagos, *et al*, Nature **543** (2017), 529.
[3] U. Hohenester, *et al,* Physical Review B **97** (2018), 165418.
[4] M. J. Lagos, *et al*, Microscopy **67** (2018), i3.
[5] M. J. Lagos and P. E. Batson, NanoLetters **18** (2018), 4556.
[6] We acknowledge V. Amarasinghe and L. Feldman for providing samples. We also thank financial support of Canada NSERC (Award #RGPIN-2019-07143) and US Department of Energy (Award #DE-SC0005132).