Highlights

Innovative materials with strong interplay of spin orbital charge and topological degrees of freedom - 2019

Position and momentum mapping of vibrations in graphene nanostructures in the electron microscope

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Even though the knowledge of vibrational properties is a key part of our understanding and optimization of a material's behaviour, phonon dispersions of freestanding 2D materials have long remained elusive because of the experimental limitations of inelastic X-ray and neutron scattering. Here we show that electron energy loss (EEL) spectroscopy in a transmission electron microscope with improved momentum and energy resolution can successfully probe the local vibrational charge responses in nanosized materials. We developed an ab-initio theory that allows to express the EEL intensity in terms of a newly introduced momentum-dependent effective charge which fully takes into account the effect of valence-electron screening beyond the spherical rigid-ion approximation. Measured scattering intensities are accurately reproduced by our theory, thus enabling a detailed interpretation of experimental data. Remarkably, we show that sizeable EEL intensities can be expected at large momenta in metals and insulators alike, irrespectively of their infrared polarizability. Additionally, a nanometre-scale mapping of selected momentum-resolved vibrational modes using graphene nanoribbon structures has enabled us to spatially disentangle bulk, edge and surface vibrations, proving the feasibility of studying local vibrational modes in truly 2D freestanding monolayers at the nanometre scale.

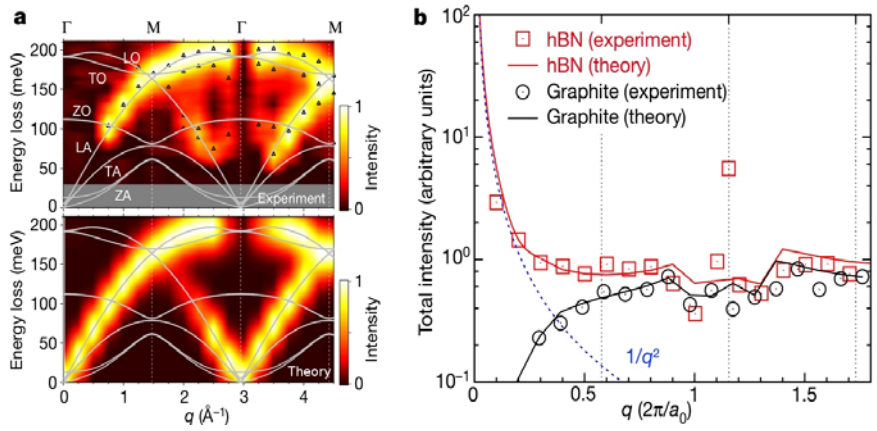

Fig. 1: a) Comparison of experimental (top) and theoretical (bottom) EEL spectra for few-layer graphite; the grey area show experimentally inaccessible region dominated by quasi-elastic peak. b) Total phonon intensity of metallic graphite and semiconducting boron-nitride from theory and experiments, displaying comparable strength at large momenta