Correlating Bosons to Heterogeneities in Two-dimensional Materials with Vibrational EELS and 4D-STEM

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Heterogeneities are inevitable in two-dimensional materials existing directly from fabrication processes, per the 2nd law of thermodynamics. These heterogeneities exist as intrinsic structural defects, [1,2] impurities in lattice sites, [3] topological disorder, [4] and strain-driven interfaces. [5] So, how can we study these defects in detail to correlate their impact on materials properties? In this talk, aberration-corrected scanning transmission electron microscopy (STEM) is combined with monochromatic electron energy loss spectroscopy (EELS) to combine unprecedented spatial resolution with world-class energy resolution from an electron probe to decouple the nature of bosons arising from subtle heterogeneities. On-axis EELS, where our bright field disc is perfectly aligned with the EELS entrance aperture, leaves our signal being dominated by dipole scattering, so we get a delocalized signal. In order to get a localized signal from adatoms and vacancies in our material, we must go off-axis. Hereby, off-axis EELS is employed, where a bright-field disc is electrically shifted outside of the EELS entrance aperture by the projector lenses such that a portion of the dark field disc now enters the spectrometer window to give information on the impact scattering in the off-axis regime. Last, we use position-averaged converged beam electron diffraction (PACBED) to get precise calculations on the structural disorder in the system at the site of heterogeneities to correlate structural disorder to vibrational spectra. Ultimately, this work not only pushes boundaries in electron microscopy, but provides avenues to the entire scientific landscape on decoupling the defect-property relationship in solids for better design of next-generation nanoelectronics.

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