Probing Nanoscale and Atomic-Level Exotic Vibrational Modes and Phonon Dynamics using Monochromated Electron Energy-Loss Spectroscopy

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Imperfections such as stacking faults and interfaces are recognized as controlling factors in modifying thermal properties and heat transport by scattering phonons and changing vibrational spectra. They play a more pivotal role when the material's dimension is reduced to the nanoscale. However, it is traditionally difficult to obtain experimental evidence of defect-induced phonon modes and phonon dynamics due to the lack of effective tools with sufficient spatial resolution. Meanwhile, in a perfect crystal, the vibration of individual atoms, known as thermal ellipsoids, could exhibit strong anisotropies and fundamentally affect the orientation dependency of dielectric, thermal and optical properties. The averaged thermal ellipsoids were conventionally estimated by diffraction methods, which encounter critical drawbacks of lacking spatial and energy resolutions. Monochromated electron energy-loss spectroscopy (EELS) in the advanced electron microscope reached an unprecedented energy resolution of a few millielectronvolts (meV), enabling the detection of phonon modes in materials with nanoscale and even atomic resolution. In this talk, I will present several examples that utilize a series of space- and angle-resolved vibrational EELS methods to investigate exotic vibrational states and phonon dynamics in diverse scenarios. (1) At a single stacking fault in SiC, the acoustic vibration modes at the X point undergo a red shift of 3.8 meV with increased spectral intensity and such defect phonons are confined to within a few nanometers of the defect. (2) Localized interfacial phonon modes were revealed in both Si-Ge heterojunctions and monolayer MoS_2-WSe_2 heterointerfaces and could promote the thermal transport across solid-solid interfaces. (3) A differential phonon momentum mapping method was developed and utilized to observe a strong specular reflection of Si optical phonons at compositionally abrupt interfaces between Si and SiGe quantum dots. (4) We recently developed a novel momentum-selective dark-field EELS method with a desired momentum exchange and probed unexpectedly frequency-linked thermal ellipsoids of individual oxygen atoms in $SrTiO_3$. Our work charts a definitive course for investigating phonon propagation in perfect crystals and near defects and provides guidance for the thermal nanoengineering of nanotransistors, power electronics, and thermoelectric devices.

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