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High Spatial Resolution Low-Voltage Electron Imaging and Spectroscopy of Two-Dimensional Materials and Semiconductor Nanostructures

Xiaoqing Pan[1,2,3], Xingxu Yan[1], Chaitanya A. Gadre[2] and Toshihiro Aoki[3]

[1.] Department of Materials Science and Engineering, University of California - Irvine, Irvine, CA, USA.
[2.] Department of Physics and Astronomy, University of California - Irvine, Irvine, CA, USA.
[3.] Irvine Materials Research Institute (IMRI), University of California - Irvine, Irvine, CA, USA.

Low-voltage aberration-corrected scanning transmission electron microscopy (AC-STEM) imaging is a powerful approach for resolving atomic structures and probing local properties of two-dimensional (2D) materials such as electronic properties and phonon characteristics. A 30-60 keV electron beam can significantly reduce knock-on damage on most 2D materials while maintaining a respectable 1 Å spatial resolution. Another significant advantage of low-voltage electron microscopy is the improvement of the energy resolution. With a newly-designed monochromator and spectrometer, electron energy-loss spectroscopy (EELS) in a STEM can probe vibrational properties of materials with sub-10 meV energy resolution [1-4], surveying local defect-induced perturbations of phonon modes. Here, we utilize high spatially-resolved vibrational spectroscopy and imaging to study optical acoustic, and polaritonic phonon modes in 2D materials (hexagonal boron nitride, *h*-BN) as well as semiconductor nanostructures.

2D *h*-BN nanosheets possess a hyperbolic phonon polariton (HPhP) response and have potential applications in hyperlensing, negative refraction and sub-diffractional resonators [3]. Thickness is the dominant factor in tuning the polariton dispersion and propagation direction inside the material. Low voltage STEM imaging is applied to distinguish the atomic structure of 1L (Fig. 1a) and 2L (Fig. 1b) *h*-BN as well as multilayered ones. Using high energy resolution vibrational spectroscopy, the HPhP are observed in 2D *h*-BN nanosheets with different layer number (Fig. 1c). Note that the detection of HPhP in 1L and 2L samples is still very challenging for optical methods. Energy position of the HPhP blueshifts with increasing layer number (Fig. 1d), which is consistent with the simulated results based on the polariton dispersion curve [3]. The results demonstrate the reliability of vibrational spectroscopy in investigating the polariton response of atomically thin 2D materials. Furthermore, combining low-voltage atomic resolution imaging and diffraction information, variation in stacking order and twisting angle additionally tune the HPhP energy position and the associated propagation of electromagnetic waves inside the material. This research benefits the understanding of electromagnetic properties of 2D material and enriches the development of tunable hyperbolic communication devices.

Silicon-Germanium quantum dots (QD) are promising systems for thermoelectric applications [5]. Since Si and Ge are nonpolar materials, only bulk phonon modes are detected in vibrational spectra, allowing for the study of local strain effects with high spatial resolution. Fig. 2a shows an atomic resolution high angle annular dark field (HAADF) image of a Si/Ge QD consisting of pure Si matrix surrounding a $Ge_{0.5}Si_{0.5}$ QD. Individual phonon signals from the Si and QD, shown in Fig. 2b, highlight the differences in the two signals as well as the redshift of the Si TO/LO peak, which is also demonstrated by a nanometer step line scan in Fig. 2c. The redshift is associated with the tensile strain on the Si atoms due to the Si-Ge intermixing inside the QD.

In summary, high resolution low-voltage imaging and vibrational spectroscopy is a powerful tool capable of revealing polariton modulation in 2D materials and localized phonon behavior in complex nanostructures and can be extended as a general method to advance the investigation of local phonon and polariton modes associated with grain boundaries, interfaces and defects [6].

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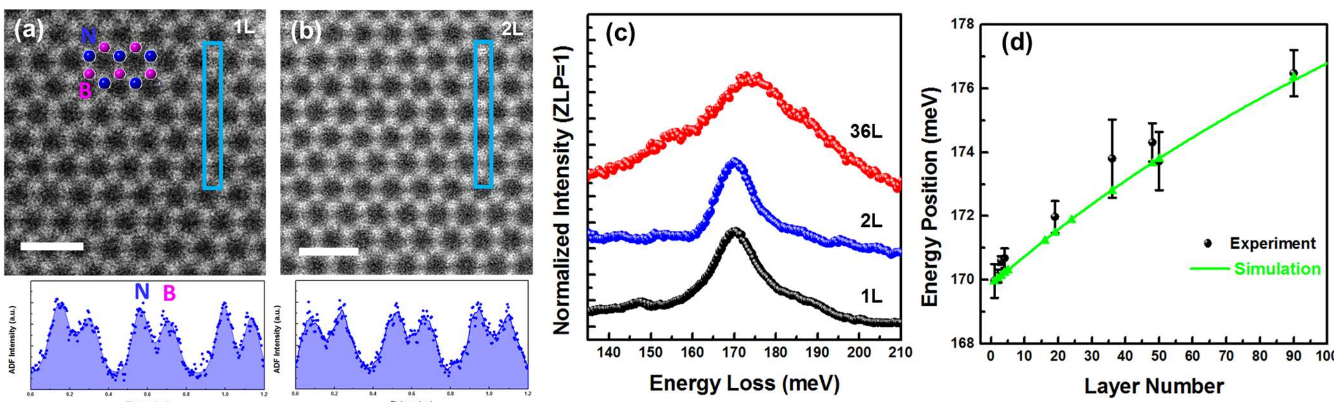

Figure 1. Imaging and vibrational spectra of *h*-BN with different layer numbers. (a) STEM image of monolayer (1L) *h*-BN overlaid by atomic structure and line profile of ADF intensity of the cyan box. The pink and blue balls represent B and N atoms, respectively. (b) STEM image of 2L *h*-BN and line profile of ADF intensity of the cyan box. Each atomic column has equal intensity, which is consistent with atomic structure of AA'-stacked 2L *h*-BN. (c) The background-subtracted EEL spectra of 1L, 2L and 36L *h*-BN with HPhP signal. (d) Peak position of HPhP as a function of layer number and simulated results.

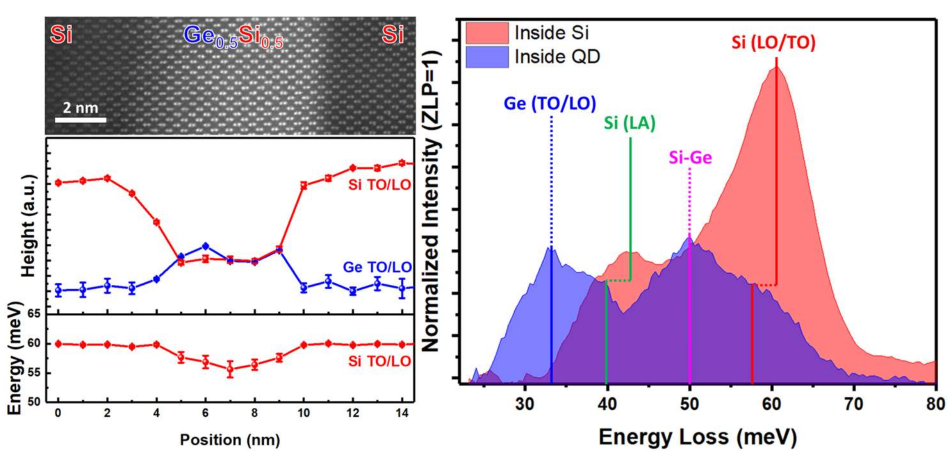

Figure 2. High Spatially-resolved optical and acoustic phonons of nanostructures. (a) Atomic resolution STEM image of the Si/Ge QD. (b) Two background subtracted spectra acquired from the Si and Si/Ge QD regions. (c) Line profile of peak height of Si TO/LO and Ge TO/LO modes across Si/Ge QD, along with the energy position of Si TO/LO mode.