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# Emergent Phonon Phenomena at Interfaces Probed by Vibrational EELS

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Uncovering phonon transport mechanisms is crucial in determining thermal conductivity in semiconductors. Phonon transport can be greatly modulated by nanostructures, interfaces, and defects. Of these scattering mechanisms, interfaces between two differing materials can scatter phonons efficiently. Additionally, interface modes have been known to increase thermal conductivity by providing an intermediate mode to facilitate transport of phonons between different materials [1]. However, this requires detailed investigation of the nanoscale physics and has hardly been explored by experimental means. The phonon resistance provided by these structures are often a result of a mismatch between the local phonon density of states (LDOS) [1], studies of which have eluded optical measurement techniques due to their insufficient spatial resolution and cannot aid in the physical understanding of nanoscale thermal transport. However, recent developments in electron microscopy have enable the acquisition of vibrational spectra with few-meV energy resolution and down to sub-ångström spatial resolution [2,3]. Armed with this technical capability, we demonstrate a mapping of phonons revealing an interface mode at the Si-Ge interface and phonon dynamics of SiGe quantum dots (QDs).

The Si-Ge is a model system that has been commonly investigated due to its low-lattice mismatch and abundant electronic applications. Fig. 1A shows an atomic resolution HAADF image of an atomically sharp Si-Ge interface acquired using a 33-mrad convergence semi-angle probe. Representative electron energy-loss spectra (EELS) in Fig. 1B show distinct spectral features of Ge, Si-Ge interface, and Si vibrational modes including the optical phonon modes of Si and Ge at 60 meV and 34 meV, respectively, as well as low-energy acoustic phonon modes. The contour plot in Fig. 1C reveals an abrupt transition between the vibrational structures of Ge and Si and reveals an interfacial mode at the interface at around 48 meV which cannot be ascribed to the bulk phonon modes of either Si or Ge or be fitted by their linear combination [4]. This is more clearly demonstrated by the line profile in Fig. 1D and reveals that the 48-meV interface mode is confined to within a 1.3 nm of the interface.

The abruptness of the interface between Si-Ge correlates to the spatial extent of the interface modes as well as affect phonon transmission and reflection properties. The growth mechanism of SiGe quantum dots (QDs) produces two types of interfaces that form the boundary of the QDs: a gradual interface at the top and an abrupt one at the bottom (Fig. 2A). In order to see the effect of various interfaces on phonon transport, we employ a 3 mrad convergence semi-angle probe to enable the investigation of phonon modes at specific points in momentum space within the first Brillouin zone (FBZ). Additionally, post-specimen lenses are utilized to collect the scattered, off-axis beam at locations denoted by $\Delta_+$ and $\Delta_-$ in momentum space denoted in Fig. 2B. Conservation of energy and momentum dictates that these points in momentum space within FBZ correspond to backward and forward propagating phonon modes, respectively. By taking the difference of forward and backward propagating modes, net momentum information i.e., direction of propagation, is obtained. Utilizing this novel technique, the differential momentum mapping in Fig. 2C reveals that there is indeed a strong reflection from the abrupt interface while the gradual interface provides a weaker directionality. The abrupt change in LDOS creates an environment where propagating Si optical modes experience a shortage of corresponding modes on the other side of the interface, leading to a reflection. By utilizing averaged and resolved momentum conditions, phonon momenta can be imaged to obtain information about phonon propagation at the nanometer scale [5, 6].

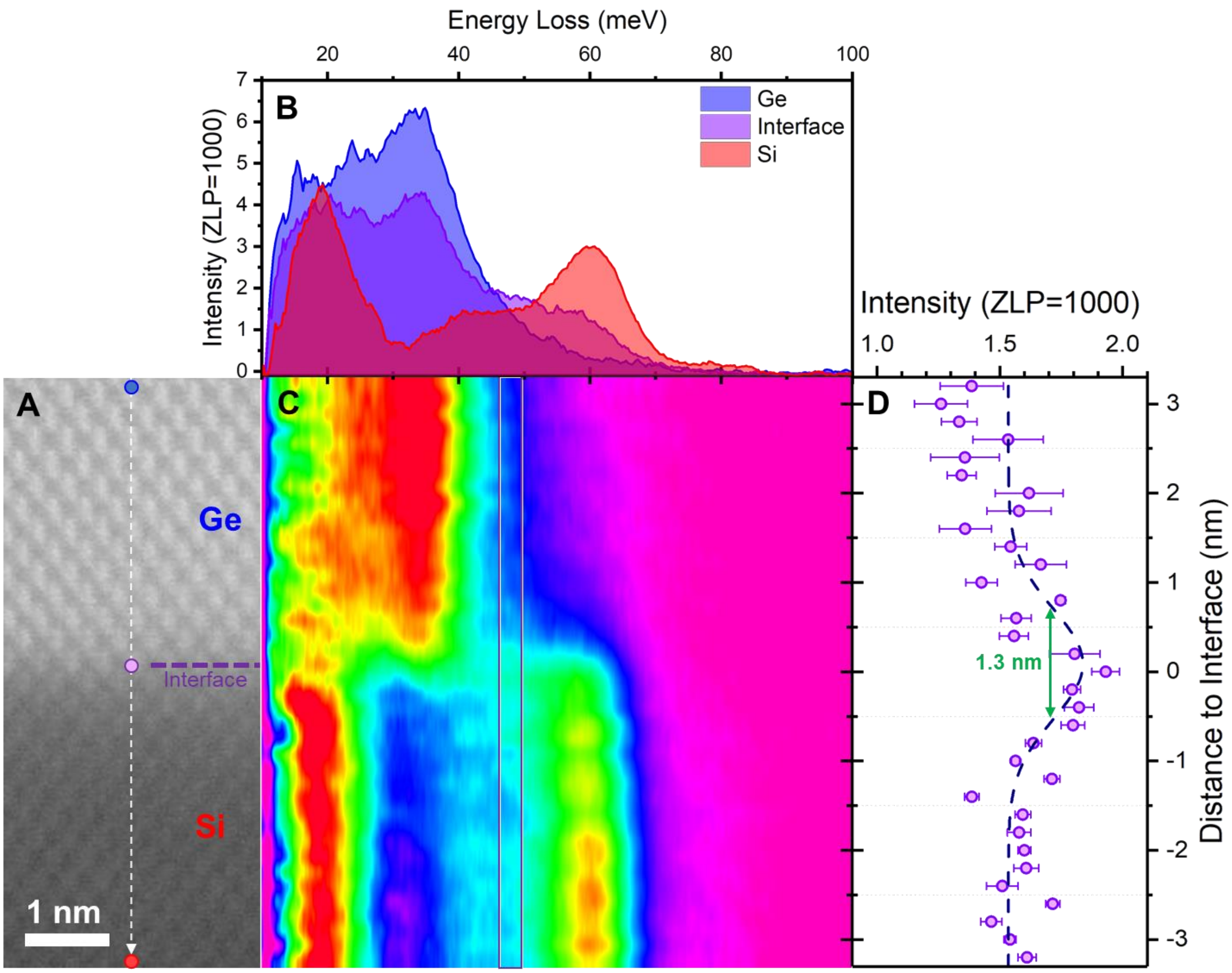


**Figure 1.** Vibrational spectra across a Si-Ge interface under a 33mrad, on-axis beam-detector geometry. **A**. [110]-zone-axis HAADF STEM image of the Si-Ge interface. Scale bar: 1 nm. **B**. Representative vibrational spectra of Ge, Si-Ge interface, and Si at locations marked in **A** by the blue, purple, and red circles, respectively. **C**. Spectral contour plot of spatially resolved vibrational spectra along the direction denoted by the white arrow in **A**. **D**. Line profile of spectral intensity at 48 meV in **C**, overlaid with a Gaussian fit.

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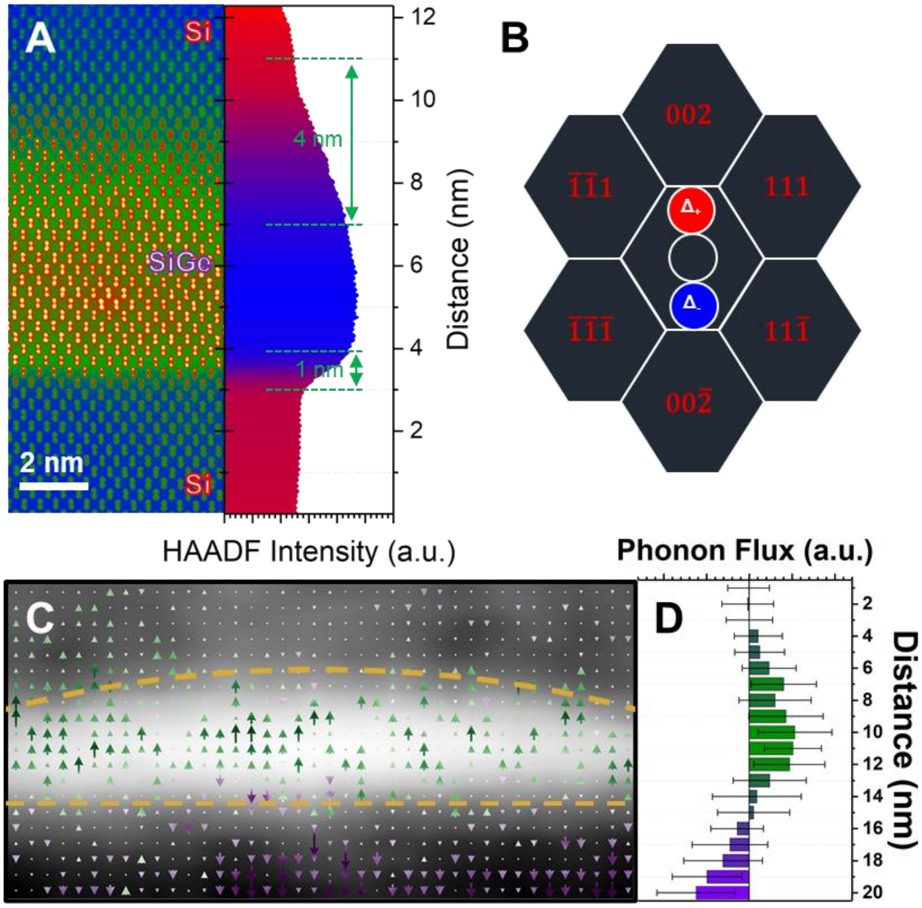


**Figure 2:** Momentum resolved differential mapping of phonon reflection. **A**. HAADF image containing both top and bottom QD interfaces. Line profile to the right shows a gradual, 4-nm-wide interface and an abrupt, 1-nm-wide one at the top and bottom QD interfaces, respectively. **B.** Reciprocal space schematic of Si. White borders denote individual Brillion zones (BZs). Red and blue regions in the FBZ correspond to the areas where momentum resolved differential data was obtained. The three regions indicated by circles are 3 mrad in diameter. **C.** Phonon flux vector map obtained from the product of group velocity and the vibrational EELS intensity with given momentum direction. Map is overlaid on a HAADF image obtained using a 3-mrad convergence semi-angle probe. **D**. Horizontally average line profile of **C**.

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