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Nanoscale Phonon Mapping of Single SiGe Quantum Dots by Vibrational EELS

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Thermoelectrics are a class of materials possessing low thermal conductivity and high electron mobility that serve to convert heat to electricity [1]. Through the engineering of complex structures such as alloys, nanostructures, and superlattice interfaces, the propagation of phonons can be manipulated to suppress material thermal conductivity while maintaining reasonable electrical conductivity [2]. Due to the lack of spatial resolution from conventional optical methods, the experimental study of phonon behavior at nanostructure interfaces has been impossible until now. Recent developments in electron energy loss spectroscopy (EELS) have made it possible to study phonons at nanometer resolution [3] Here we demonstrate the two-dimensional nanoscale vibrational mapping of a single SiGe quantum dot (QD) using an atom-sized probe in the electron microscope combed with angle resolved capability. For the first time, we observe the nanoscale composition-driven redshift of the Si optical mode as a function of Ge composition within a single QD. Our data also reveal differences in local vibrational structure around the QD suggesting a thermal diode-like properties. Our work demonstrates high spatial resolution vibrational characterization of nanostructures that can be extended to other nanostructures and superlattice systems.

Fig 1 contains elemental and vibrational information of a single SiGe QD showing high spatial correlation with Ge content, compositional strain, and phonon mode excitation probability. EELS core-loss mapping reveals a composition non-uniformity formed by Si-Ge diffusion during the CVD growth process that leaves a sharp interface at the base and a gradual interface at the top of the QD. The alloying of Si and Ge produces a composition-driven tensile strain on the Si atoms in the QD and causes the Si optical mode (OM) to be redshifted. The redshift line profile in Fig. 1e reaches a minimum close to the bottom interface and shows the same trend as the composition line profile. As a result of this composition inhomogeneity, the local vibrational structure is modulated, and phonon density of states (PDOS), which is proportional to EELS vibrational intensity, is changed. In the Si OM peak intensity mapping in Fig. 1d, the modes are enhanced in the Si region below the QD relative to the region at the top. Owing to the sharp change in density of states above and below the sharp interface, vibrations are reflected downwards away from the QD more intensely than those approaching the top interface where there is a more gradual change in PDOS.

Reflection of OM phonon momentum was observed by employing a momentum resolved STEM-EELS geometry. Spectral maps were acquired from both the red and blue circles in Fig. 2a and their vibrational signals subtracted from one another to obtain a differential momentum vector map of a single QD. The result in Fig. 2b confirms a reflection of OM's from the sharp interface. The mapping of momentum resolved phonons offers us valuable insight into interface dynamics. This nanoscale spatial correlation between composition and phonon softening was not possible before the advancements in energy monochromation in the TEM but now gives us deeper insight into the physical functions of thermoelectric nanostructures. [4]

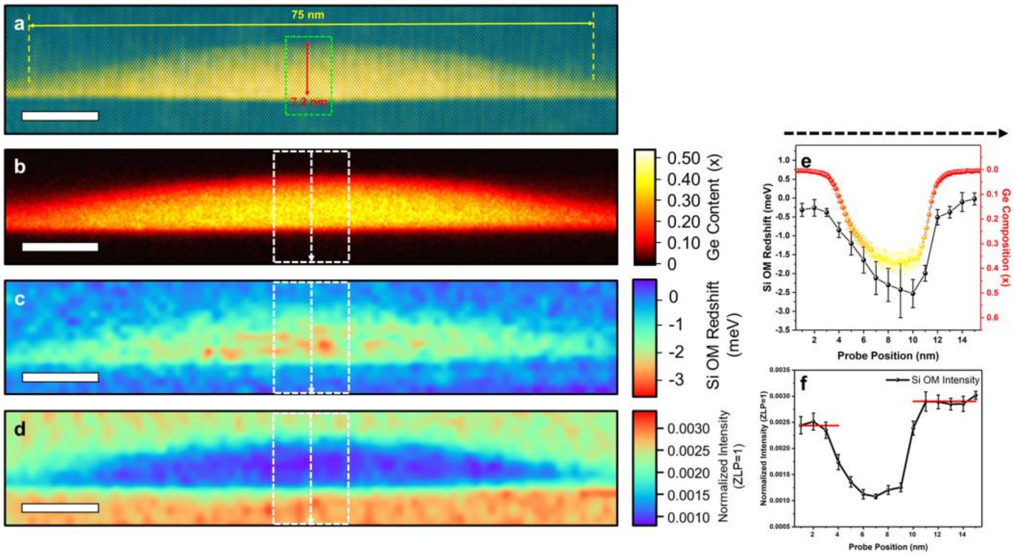

Figure 1. Nanoscale elemental and vibrational spectroscopy of a SiGe QD. (a) Atomic resolution high angle annular dark field image of a SiGe QD. (b) Core loss elemental mapping of a SiGe QD. (c-d) Mapping of the Si optical mode redshift and peak intensity respectively (e) Line profiles of composition (b) and redshift (c) mapping. (f) Line profile of Si optical mode intensity. scale bars are 10nm.

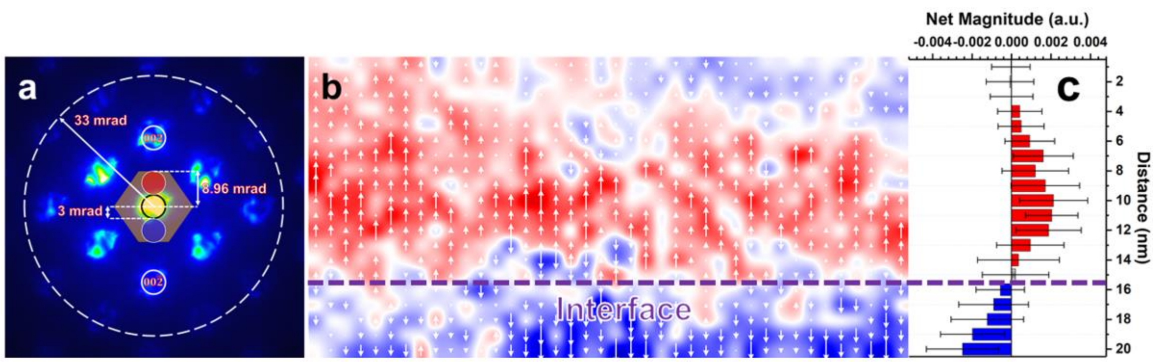

Figure 2. Momentum resolved differential mapping of phonon vertical momentum. (a) 3mrad convergence semi-angle diffraction pattern where the 002 and 00-2 spots are labeled. First Brillion zone of Si is marked in orange and distance to the X point is labeled. The red and blue circles near the center denote the EELS entrance aperture positions where off axis data was acquired is illustrated. (b) Differential momentum mapping from spectra acquired at the blue and red circled locations in (a). (c) Horizontally averaged line profile of (b).

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