Lattice Vibrational Modes in Si/Ge Core-shell NWs

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Introduction

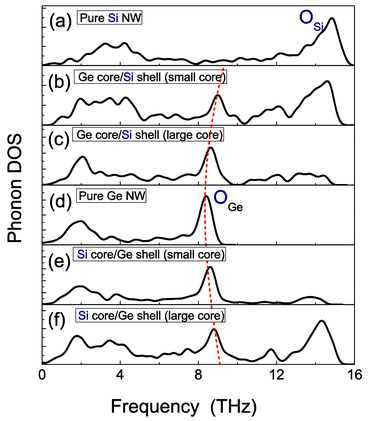
Si/Ge core-shell nanowires (NWs) has been demonstrated to have promising electrical and optical properties associated with broad applications. Recently, they are shown to own higher efficiency than Si NWs as thermoelectric devices. How to design core and shell geometries are of crucial interest for their applications. Moreover, lattice vibrational modes and corresponding phonons are the foundation of thermal properties of solids. Therefore, we study lattice vibrational modes of Si/Ge core-shell NWs and their Raman scattering spectrum to extract useful structural information and hope to understand their relevant thermal properties.

Computational Methods

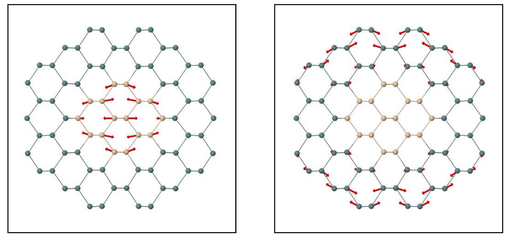
1. The calculation is based on the density functional theory (DFT) within the local density approximation (LDA) and normal-conserving pseudopotential. We employ the linear response approach to obtain lattice vibrational modes and their frequencies.
2. We calculate the Raman scattering spectrum by considering the non-resonant first-order process. We have spatially averaged the Raman activity for NWs aligned randomly.

High-frequency Optical Modes

We first focus on the high-frequency optical modes. In the figure of density of vibrational modes (DVM), we observe two characteristic peaks for most core-shell NWs, representing high-frequency optical Ge-Ge modes and Si-Si mode respectively. The two peaks exhibit substantial frequency shifts as the core-shell size varies. For example, as Ge core size increases in the case of Ge-core/Si-shell NWs, both peaks have a red shift.

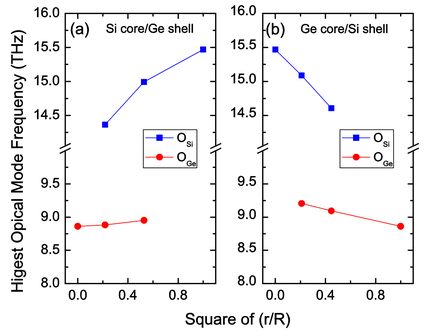


Vibrational mode density of states of Si/Ge core-shell NWs and homogenous NWs with wire diameter of about 2.3 nm.



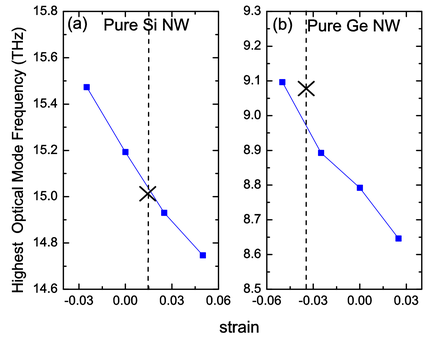
The typical vibrations of high-frequency optical modes in Si/Ge core-shell NWs.

The physical origin of the above shifts of prominent peaks in the DVM can be attributed to two factors, quantum confinement and strain condition. The strain in core-shell NWs is roughly proportional to the core/wire ratio according to the Vegard's law. This is confirmed by our simulation in the following plots.

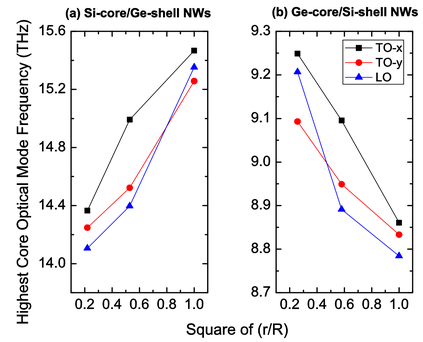


The frequency of the optical mode picked from two peaks in the DVM.

To further verify the strain effect, we study the highest transverse optical mode frequency of pure SiNW and pure GeNW under external axial strain. The highest frequency of core modes of two typical core-shell NWs is marked by at the position of the corresponding strain that the core endures by a cross symbol.



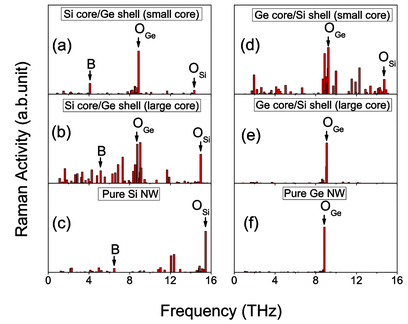
The highest longitudinal and transverse optical modes splitting is observed in Si/Ge core-shell NWs. The splitting varies as the core shell structure changes. TO along [001] direction always has the highest frequency. The change of splitting is a result of competition of internal strain and quantum confinement.



Highest LO-TO mode splitting in Si/Ge core-shell NWs

Raman Scattering Spectrum

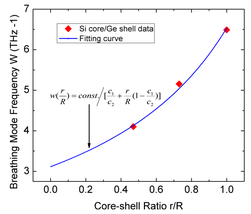
Raman scattering is a widely used approach to detect lattice vibrational modes of solids. Our calculated Raman spectrum shows the following features.

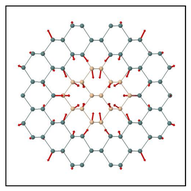


1. Those high-frequency optical modes consisting of peaks O_{Si} and O_{Ge} are strongly active.
2. Those Ge modes have much stronger Raman signals than those Si modes.
3. A Raman scattering peak is usually enhanced if the corresponding mode within the core region while it is depressed if the mode is within the shell region.
4. While the pure GeNW which has only one dominant Raman peak at the highest optical frequency, the pure SiNW displays bright Raman activities for many phonon modes within the low-frequency regime.

Radial Breathing Modes (RBMs)

Because of the unique geometry of quasi-one-dimension nanostructures, RBMs in core-shell NWs are of great interests and importance. The RBM is identified in the Si-core/Ge-shell NWs studied in this work and it is Raman active and can be detected by the Raman scattering experiments. On the other hand, for our studied Ge-core/Si-shell NWs, we cannot identify the RBM by the simple eye guidance. In this sense, the RBM may not a good option to detect the structure of Si-core/Ge-shell NWs.





By solving the classic wave equation with a cylindrical boundary condition and regarding core and shell regions as two different homogenous elastic media with different sound velocities C_1 and C_2 respectively, the frequency of RBM ω is given by:

$$\omega(\frac{R_1}{c_1} + \frac{R_2 - R_1}{c_2}) \approx const$$

Conclusion

We present a first-principles study on lattice vibrational modes of Si/Ge core-shell nanowires (NWs). The internal strain induced by the lattice mismatch between core and hell contributes to significant frequency shifts and energy-level splitting of high-frequency optical modes. In particular, these frequency shifts can be detected by Raman scattering experiments, providing convenient and nondestructive ways to obtain structural information of core-shell materials. Radial breathing modes (RBM) are identified in Si-core/Ge-shell NWs and their frequency relation is derived. Our obtained vibrational modes and their frequencies could be useful for thermoelectric applications as well.

References and Acknowledgement

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