Phononics 2019: 5th International Conference on Phononic Crystals, Metamaterials, Phonon Transport, Topological Phononics

SHINES
Spins and Heat In Nanoscale Electronic Systems

Engineering the Phonons:

Probing Phonon Dispersion with Brillouin Spectroscopy

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Plenary Talk - 2019 Phononics, University of Arizona

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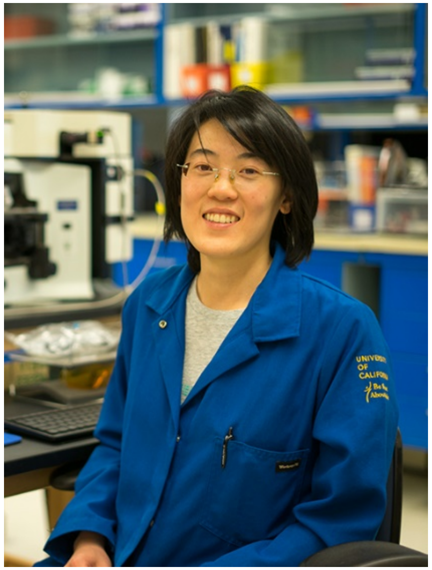
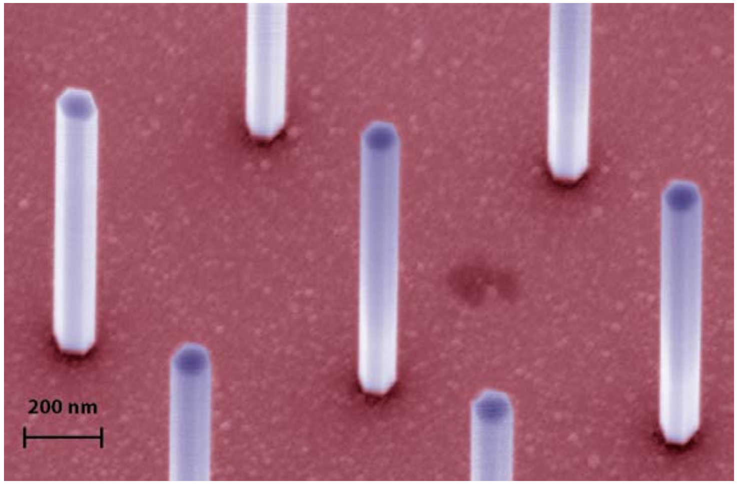
Acknowledgements

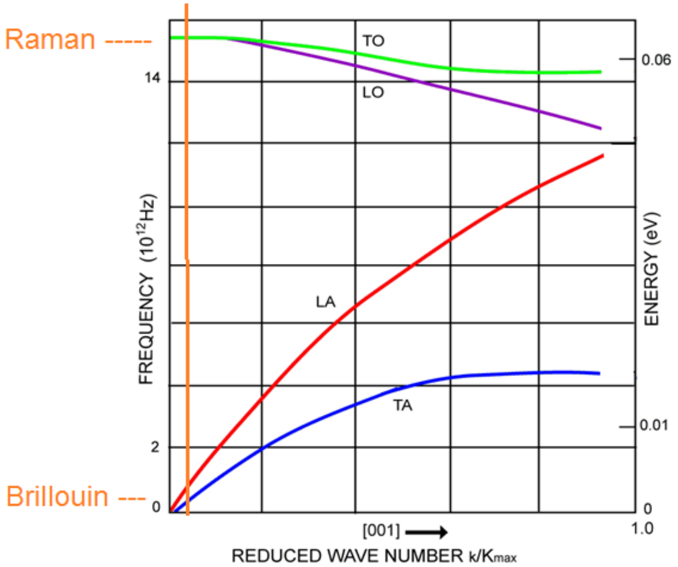


- Dr. Fariborz Kargar – Project Scientist: Brillouin and Raman spectroscopy
- Chun-Yu Tammy Huang – PhD Student: nanofabrication and Brillouin spectroscopy
- Adane Geremew – PhD Student: nanofabrication and Raman spectroscopy
- Ece Aytan – PhD Student: Raman spectroscopy; characterization (current job: Intel)

Outline

- → Phonon confinement concept
- → Implications for electron and thermal transport
- → Confinement in individual nanostructures
- → Brillouin spectroscopy of acoustic phonons
- → Interplay between confinement in individual nanostructures and periodic superlattices
- → Phonon dispersion change in bulk materials

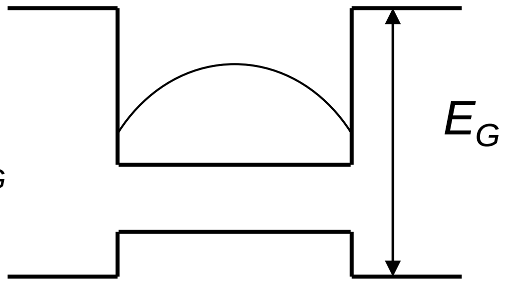




Nanoscale Phonon Engineering

Definition: phonon engineering is an approach for modifying the thermal, electrical and optical properties of materials via tuning the phonon characteristics at nanometer scale through the spatial confinement-induced changes in the phonon spectrum.

Band-structure engineering: mismatch of E_G

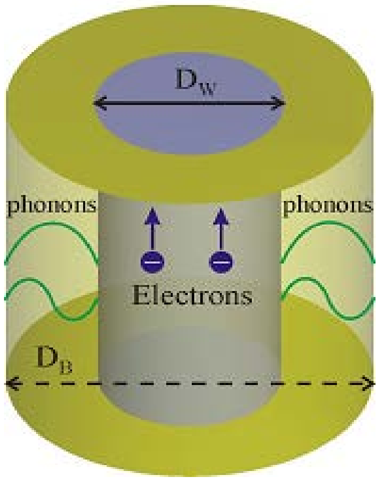


Goals:

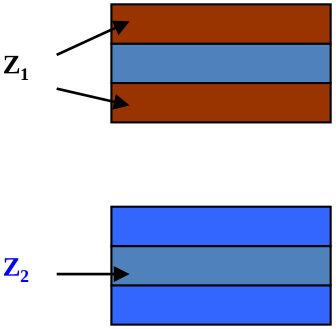
- Change in electron – phonon scattering → mobility and spin
- Change in phonon group velocity and DOS → thermal conductivity
- Control of the phonon energies → optical response

Tuning Parameters:

- Crystalline structure
- Dimensions
- Sound velocity
- Mass density
- Acoustic impedance
- Interface
- Phonon frequencies



Phonon engineering: mismatch of $Z=\rho V_{sound}$

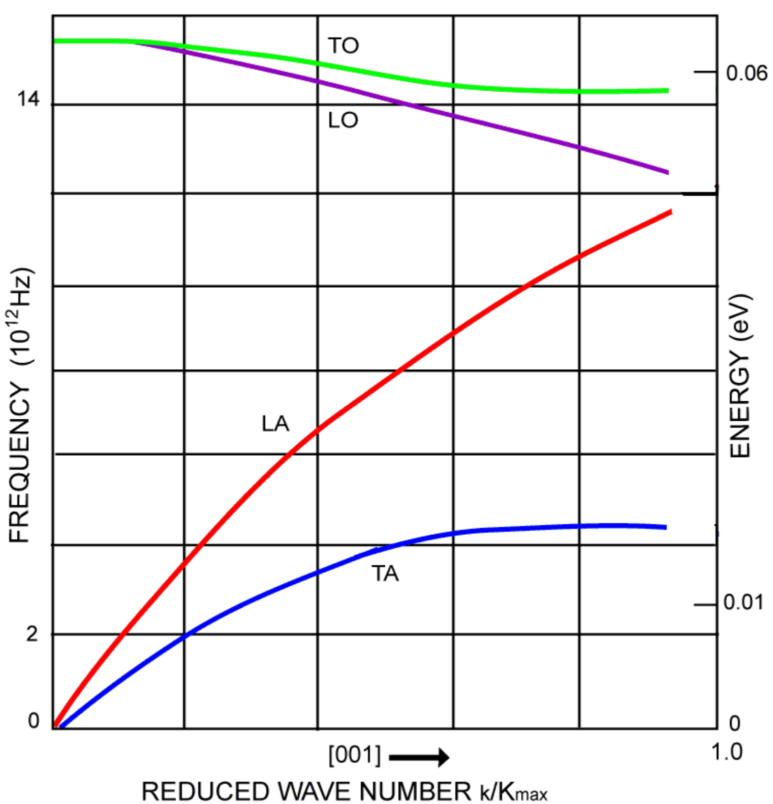


Acoustic Impedance: $Z=\rho V_s$ [kg/m²s]

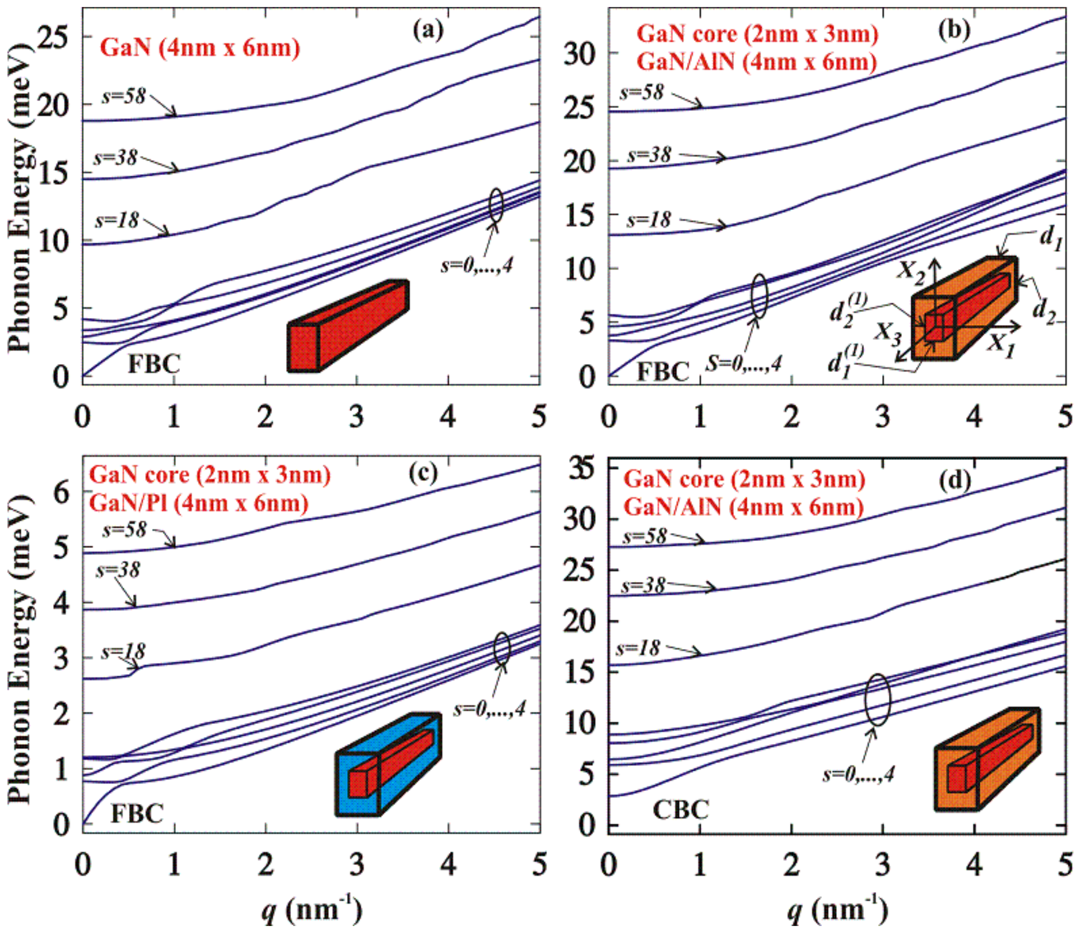
A.A. Balandin, "Nanophononics: Phonon engineering in nanostructures and nanodevices," *J. Nanoscience and Nanotechnology*, **5**, 7 (2005).

Bulk vs. Confined Acoustic Phonons

Bulk Semiconductor



E.P. Pokatilov, D.L. Nika and A.A. Balandin, "Acoustic-phonon propagation in semiconductor nanowires with elastically dissimilar barriers," *Physical Review B*, **72**, 113311 (2005)



D.L. Nika, E.P. Pokatilov and A.A. Balandin, *Appl. Phys. Lett.*, **93**, 173111 (2008).

Electron Mobility Calculation in Nanowires

Theoretical Formalism - Electron Mobility in Semiconductors

Bulk Silicon

$$\mu_l \sim (m^*)^{-5/2} T^{-3/2}$$ ← acoustic phonons

$$\mu_i \sim (m^*)^{-1/2} N_I^{-1} T^{3/2}$$ ← impurities

Momentum relaxation rate due to phonons:

$$\tau_{\mathrm{ph}}^{-1}(k_z) = \frac{2\pi}{\hbar} E_a^2 \sum_{\mathbf{q}} \left| \langle \nabla \cdot \mathbf{u}_{\mathbf{q}} \rangle \right|^2 \frac{q_z}{k_z} \left[\left(N_{\mathbf{q}} + 1 \right) \delta(\varepsilon_{k_z - q_z} + \hbar\omega_{\mathbf{q}} - \varepsilon_{k_z}) + N_{-\mathbf{q}} \delta(\varepsilon_{k_z - q_z} - \hbar\omega_{-\mathbf{q}} - \varepsilon_{k_z}) \right]$$

Momentum relaxation rate due to ionized impurities:

$$\tau_{\mathrm{imp}}^{-1}(k_z) = \frac{2\pi m N_I R_1^2}{\hbar^3 k_z} \left(\frac{Ze^2}{2\pi\varepsilon_0 \varepsilon} \right)^2 \left[\ln(k_z R_1) \right]^2$$

The low-field electron mobility in the nanowire:

$$\mu = -2\frac{e}{m} \int_0^\infty \varepsilon^{1/2} \frac{\partial f_0}{\partial \varepsilon} \tau(\varepsilon) d\varepsilon \Big/ \int_0^\infty \varepsilon^{-1/2} f_0(\varepsilon) d\varepsilon$$

$f_o(\varepsilon)$ is the electron occupation number given by the Fermi-Dirac distribution. In the non-degenerate case, it is given by a Maxwellian distribution.

Phonons in a cylindrical nanowire:

$$\mathbf{u}_{\omega,q_z} = \left[\left(\frac{dG}{dr_\perp} + q_z \frac{dF}{dr_\perp} \right) \mathbf{e}_\rho + i\left(-q_z G(r_\perp) + q_t^2 F(r_\perp) \right) \mathbf{e}_z \right] e^{-iq_z z}$$

$$G^{(n)}(r_\perp) = C_1^{(n)} J_0(q_\ell r_\perp) + C_2^{(n)} N_0(q_\ell r_\perp)$$

$$F^{(n)}(r_\perp) = C_3^{(n)} J_0(q_t r_\perp) + C_4^{(n)} N_0(q_t r_\perp)$$

$$q_\ell^2 = (\omega / c_\ell)^2 - q_z^2$$

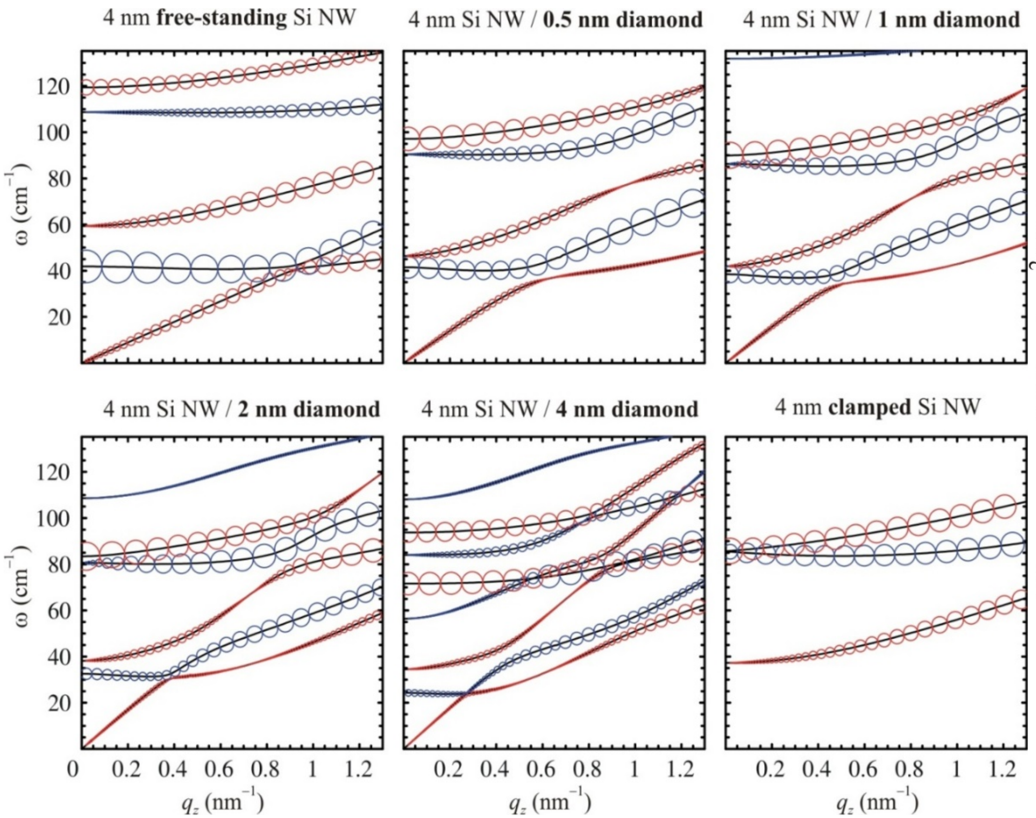
$$q_t^2 = (\omega / c_t)^2 - q_z^2$$

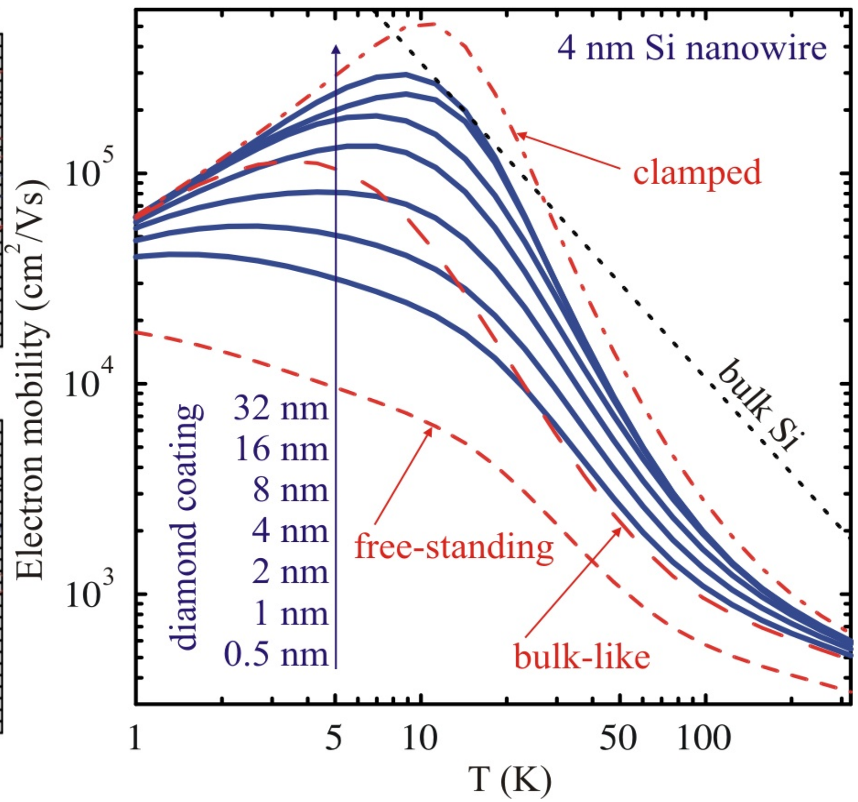
Bulk-like phonons:

$$\omega_{\mathbf{q}} = c_\ell q$$

$$\mathbf{u}_{\mathbf{q}} = \sqrt{\frac{\hbar}{2c_\ell \rho V}} \frac{\mathbf{q}}{q^{3/2}} e^{-i\mathbf{q}\cdot\mathbf{r}}$$

Nanowires with “Acoustically Hard” Barriers

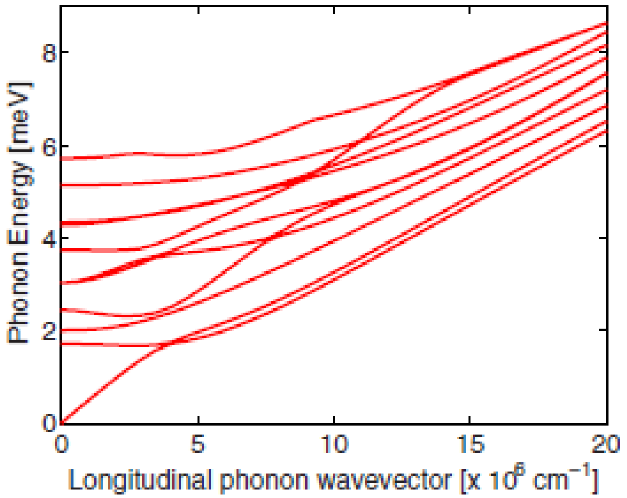


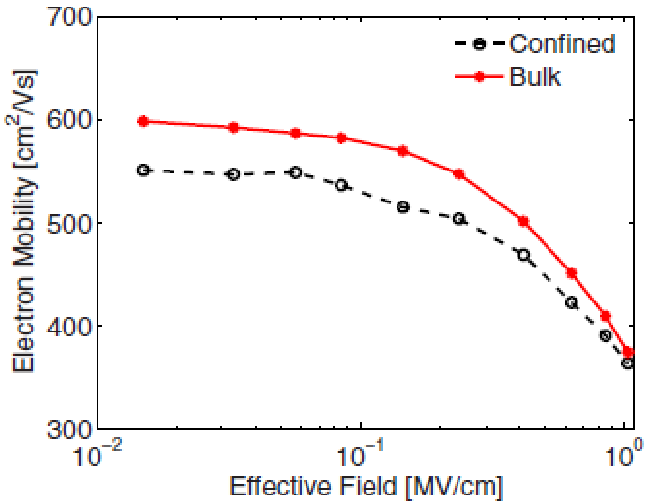


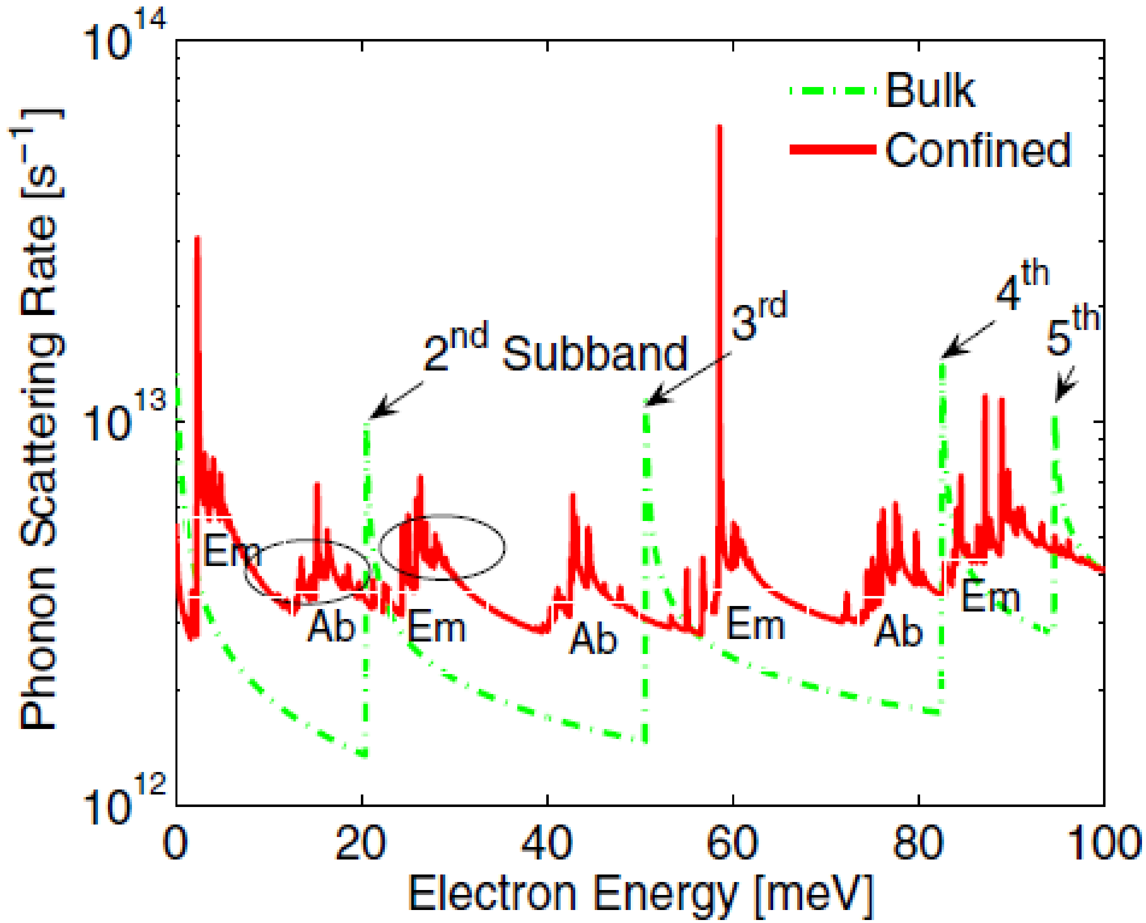
→ The size of the circles is proportional to the average divergence of displacement
→ Contribution of higher energy modes is negligible compared to the shown modes
→ “True” acoustic mode changes velocity from that in Si to the diamond
→ Contribution of the “true” acoustic mode to scattering decreases with increasing coating thickness

V.A. Fonoberov and A.A. Balandin, “*Nano Letters*, **6**, 2442 (2006).

Phonon Confinement Effects on Electron Transport







E. B. Ramayya, D. Vasileska, S. M. Goodnick, and I. Knezevic, J. APPLIED PHYSICS, 104, 063711 (2008).

Phonon Confinement Effects on Thermal Transport

- Acoustic phonons are the main heat carriers in semiconductors and insulators:

$$K_P = \frac{1}{3} C_P \upsilon \Lambda = \frac{1}{3} C_P \upsilon^2 \tau$$

$$(1/\tau) = (1/\tau)_B + (1/\tau)_P + (1/\tau)_{DC} + \ \ldots$$

- Introduction of impurities affects thermal conductivity via increased scattering making terms Γ and N_D larger:

$$(1/\tau)_P = \frac{V_0 \Gamma \omega^4}{4 \pi \upsilon^3} \qquad (1/\tau)_{DC} = \eta N_D \frac{V_0^{4/3} \omega^3}{\upsilon^2} \qquad \Gamma = \sum_i f_i \left(1 - \frac{M_i}{M}\right)^2$$

- Changes in the phonon group velocity affects scattering on defects and in Umklapp phonon processes

A. Balandin and K. L. Wang, "Significant decrease of the lattice thermal conductivity due to phonon confinement in a free-standing semiconductor quantum well," Phys. Rev. B, 58, 1544 (1998).

Can We Control Phonon Dispersion in Nanostructures?

→ Modifying the acoustic phonon spectrum in individual nanostructures via spatial confinement would bring benefits for controlling phonon-electron interaction and phonon thermal conduction at the nanoscale.
→ Recent studies suggested that phonons with extra large MFP contribute substantially more to thermal conductivity than previously believed (40% for Si near RT with MFP above 1 μm).
→ Importance of long wavelength phonons: $\tau \propto \omega^{-s}$

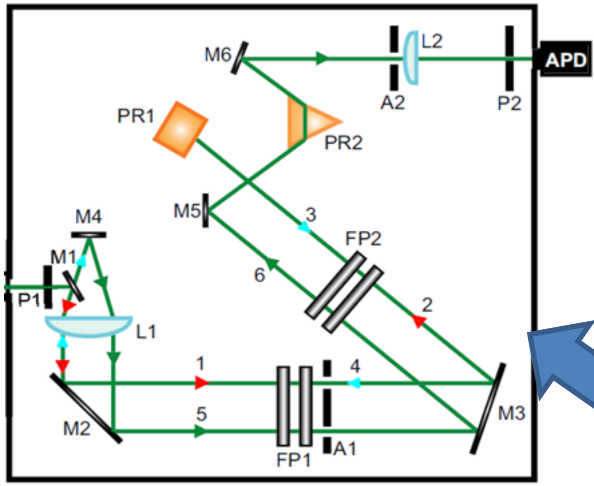
→ Conclusive experimental evidence of acoustic phonon confinement in individual free-standing nanostructures was missing.
→ The length scale, at which phonon dispersion undergoes changes was debated.

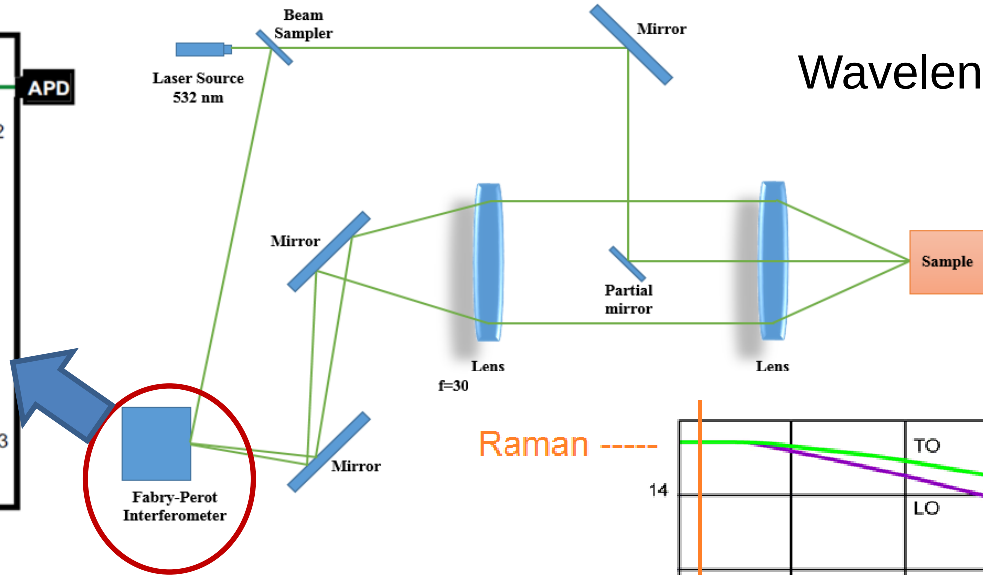
Thermal phonon wavelength: $\lambda_T \approx (\upsilon_s h)/(k_B T)$

"Grey" phonon MFP: $\Lambda_G = 3K/(C_V \upsilon_s)$

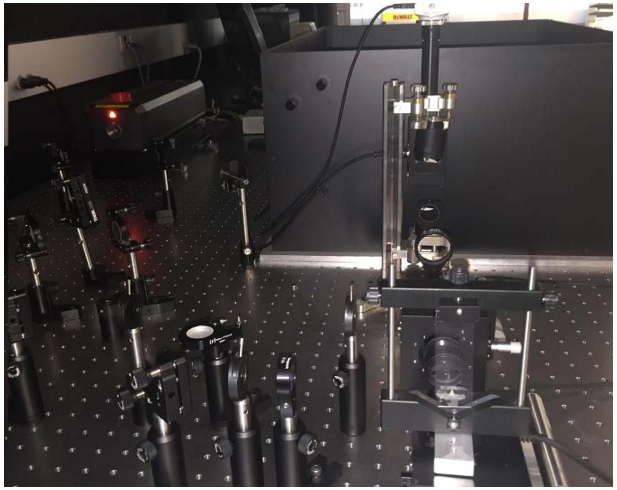
→ The natural roughness of nanostructures and crystal inharmonicity would destroy the phonon wave coherency – no phonon confinement effects in real structures even if theory suggests they should be present.

Brillouin – Mandelstam Spectroscopy

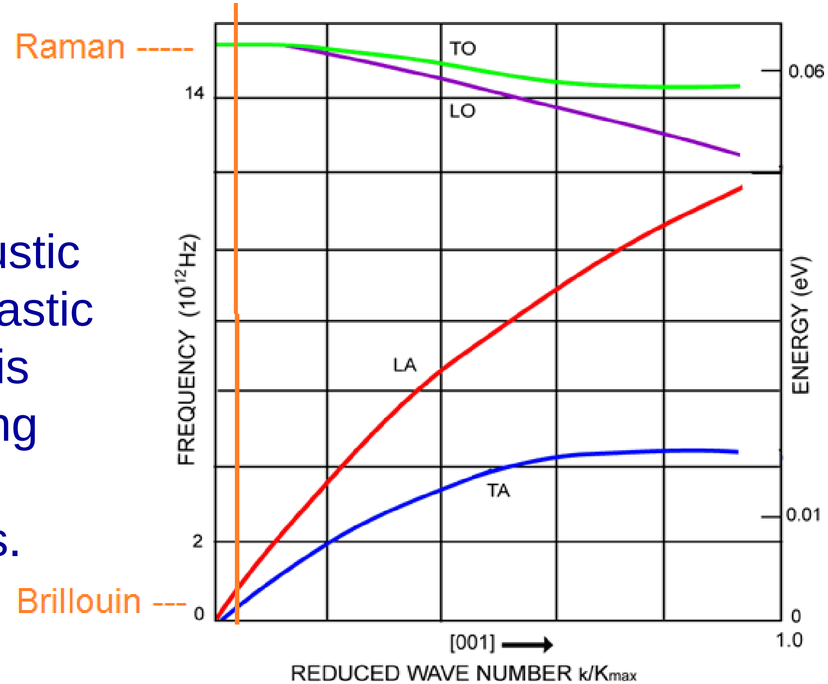




Wavelength: 532 nm



Observing acoustic phonons in inelastic light scattering is more challenging than observing optical phonons.

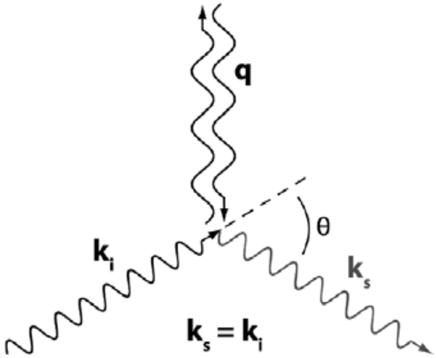


Basics of Brillouin-Mandelstam Technique

Momentum and Energy Conservations:

$$\boldsymbol{k_s} - \boldsymbol{k_i} = \pm \boldsymbol{q} \qquad \text{(Momentum)}$$

$$\omega_s - \omega_i = \pm \omega \qquad \text{(Energy)}$$



(-) sign: phonon creation; Stokes
(+) sign: phonon annihilation; anti-Stokes

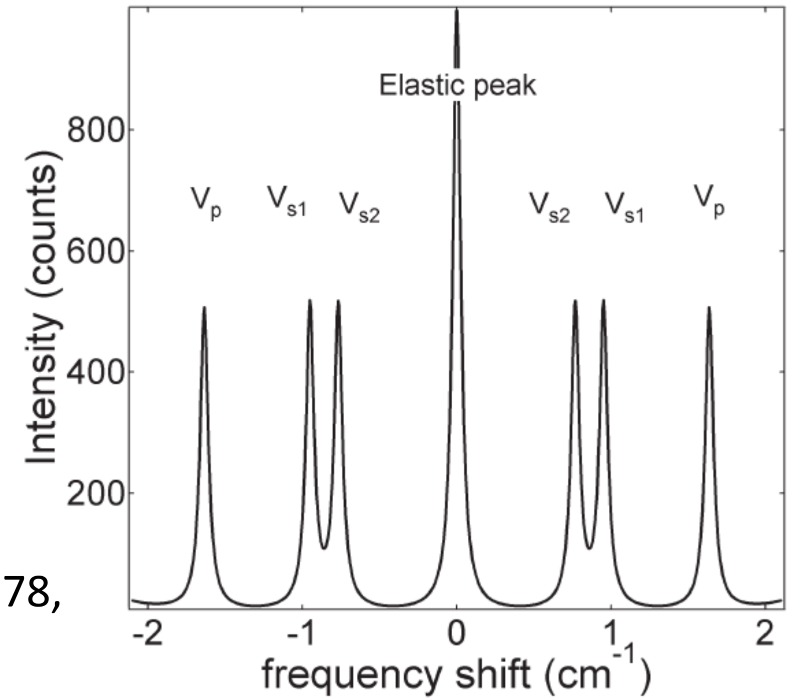
$$\omega = \omega_s - \omega_i = \pm \upsilon q$$

$$f_s = f_i \pm \frac{\upsilon q}{2\pi} = f_i \pm \frac{\upsilon}{2\pi} \frac{4\pi n}{\lambda} \sin\frac{\theta}{2} = f_i \pm \frac{2\upsilon n}{\lambda} \sin\frac{\theta}{2}$$

The BLS spectrum consists of doublets centered at the elastic frequency with frequencies (frequency shifts)

$$f = \pm \frac{\upsilon q}{2\pi}$$

S. Speziale, et al., 78, 543 (2014).



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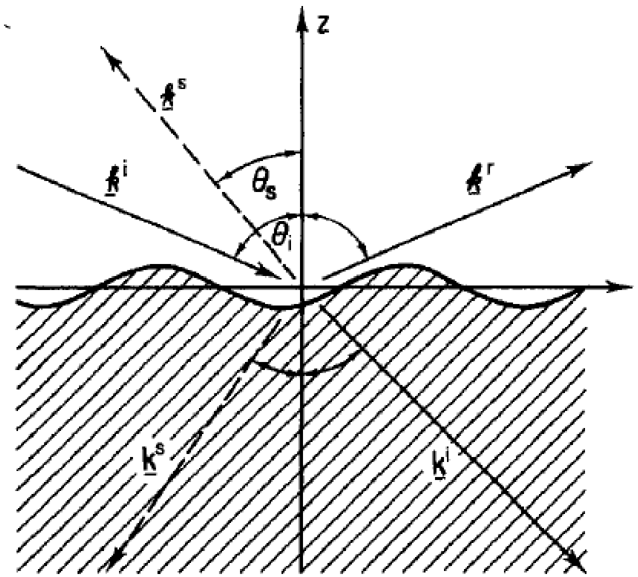
Scattering by Volume vs. Surface Phonons



Sandercock, J., Springer: 1982; pp 173-206.

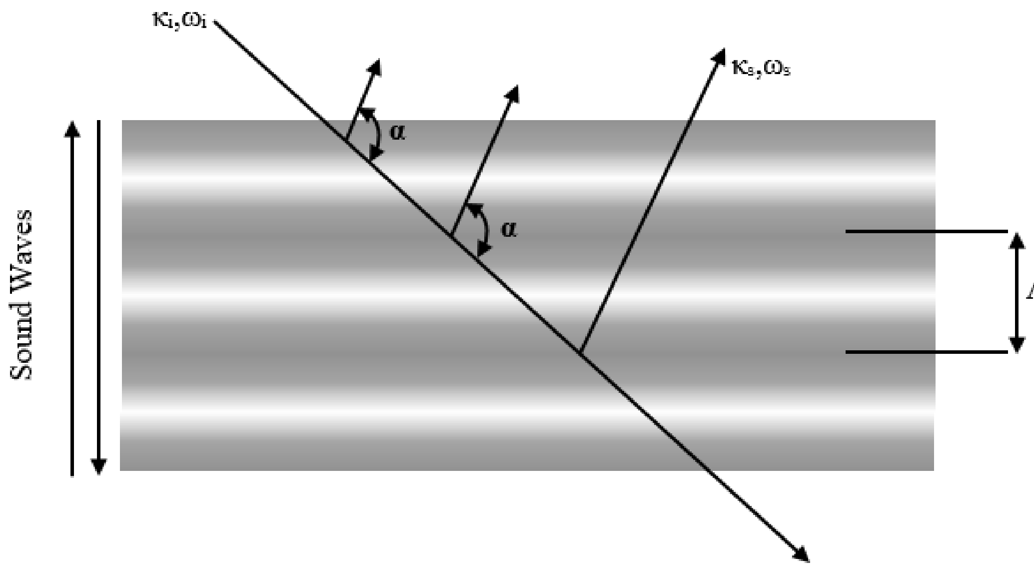
→ For the elasto-optic contribution (α is the scattering angle):

$$q = (4\,\pi\, n/\lambda)\,\sin(\alpha/2)$$

→ The ripple scattering is dominant for opaque materials.
→ For semi-opaque materials the elasto-optic effect can contribute to the scattering near the surface.

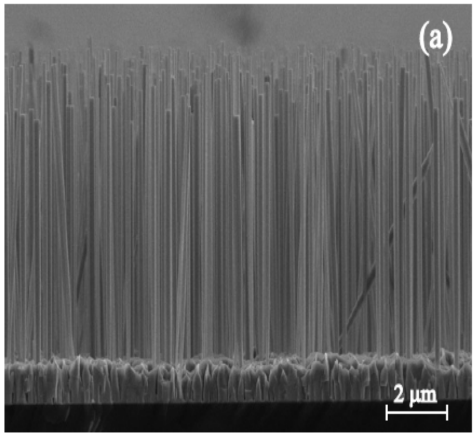
$$q = (4\,\pi/\lambda)\,\sin\theta$$

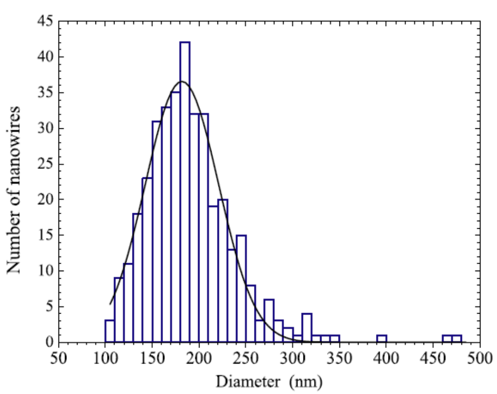
θ : Incident angle

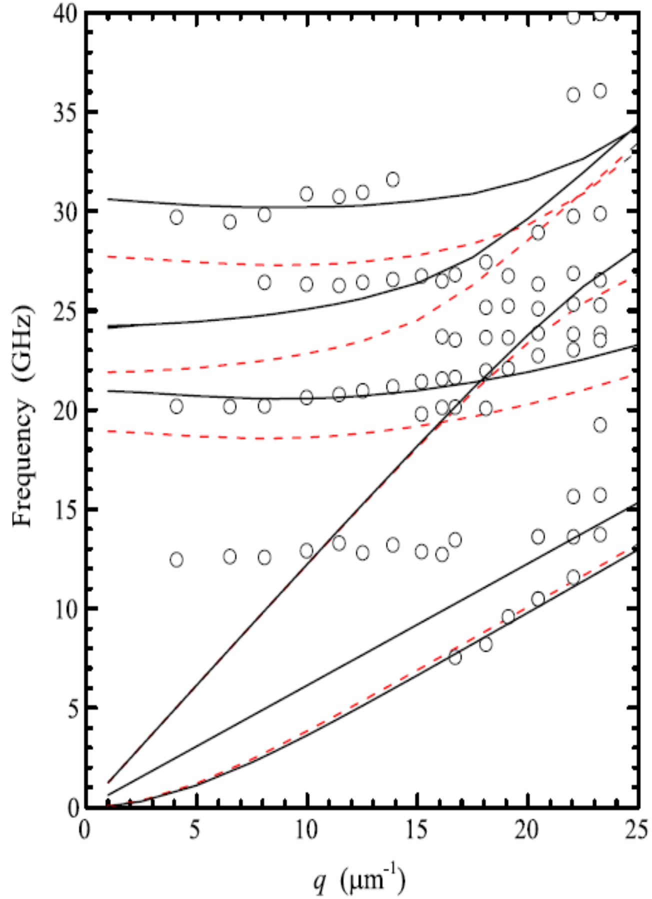


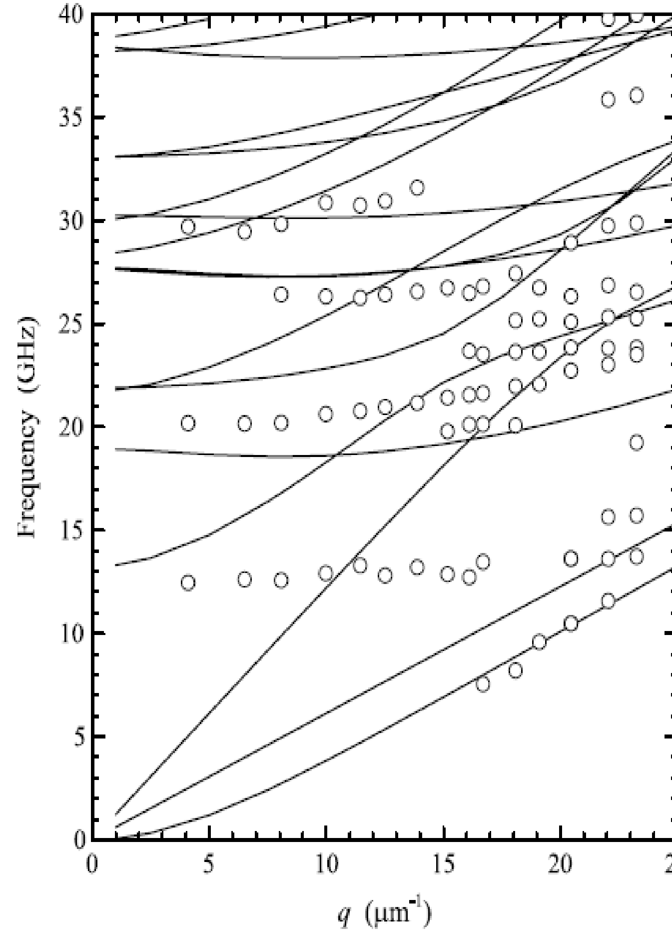
Early Experimental Studies

Brillouin - light-scattering measurements and finite-element modeling of vibrational spectra in mono-crystalline GaN nanowires







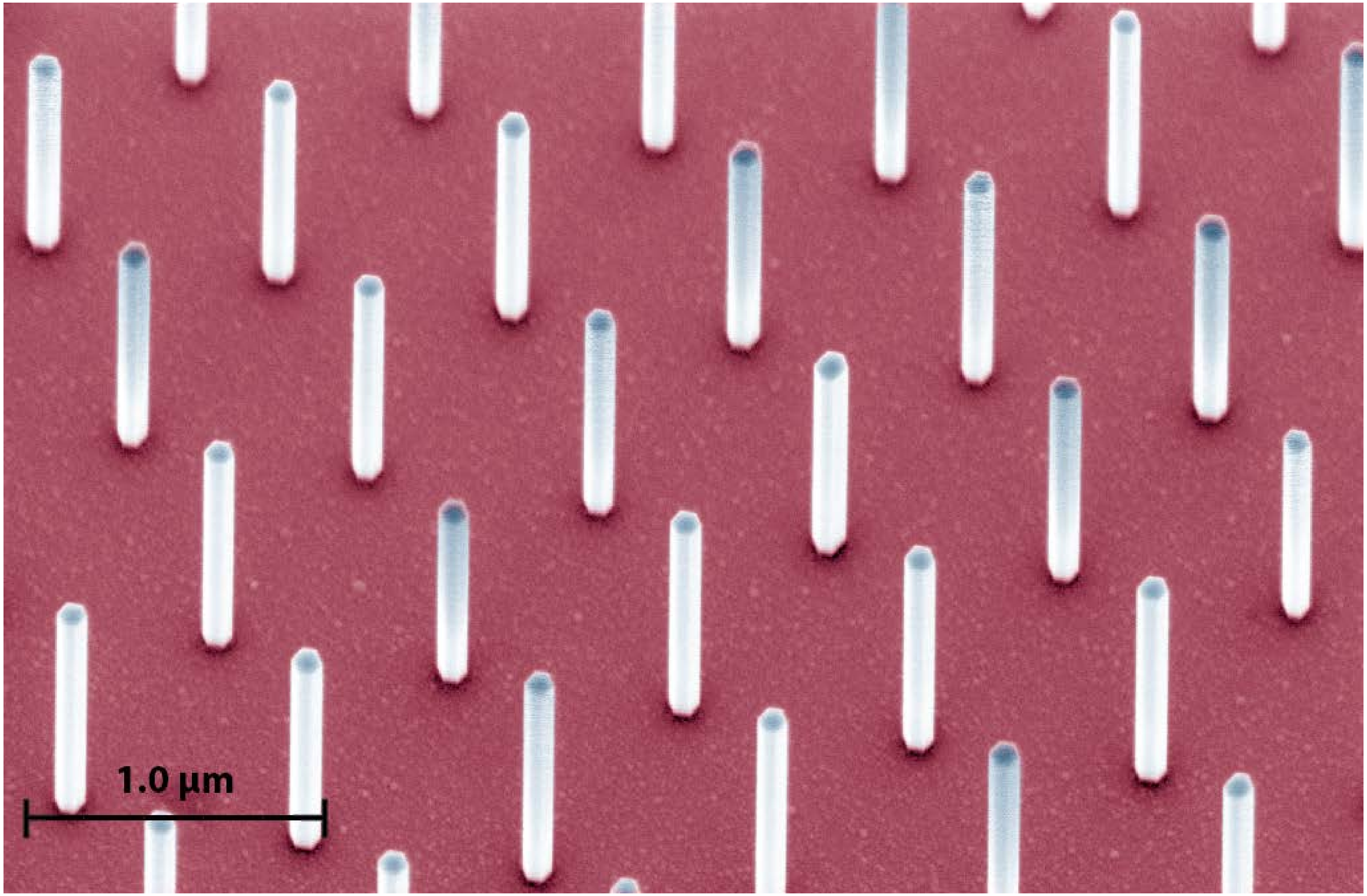


W.L. Johnson *et al.*, *Nanotechnology*, **23**, 495709 (2012).

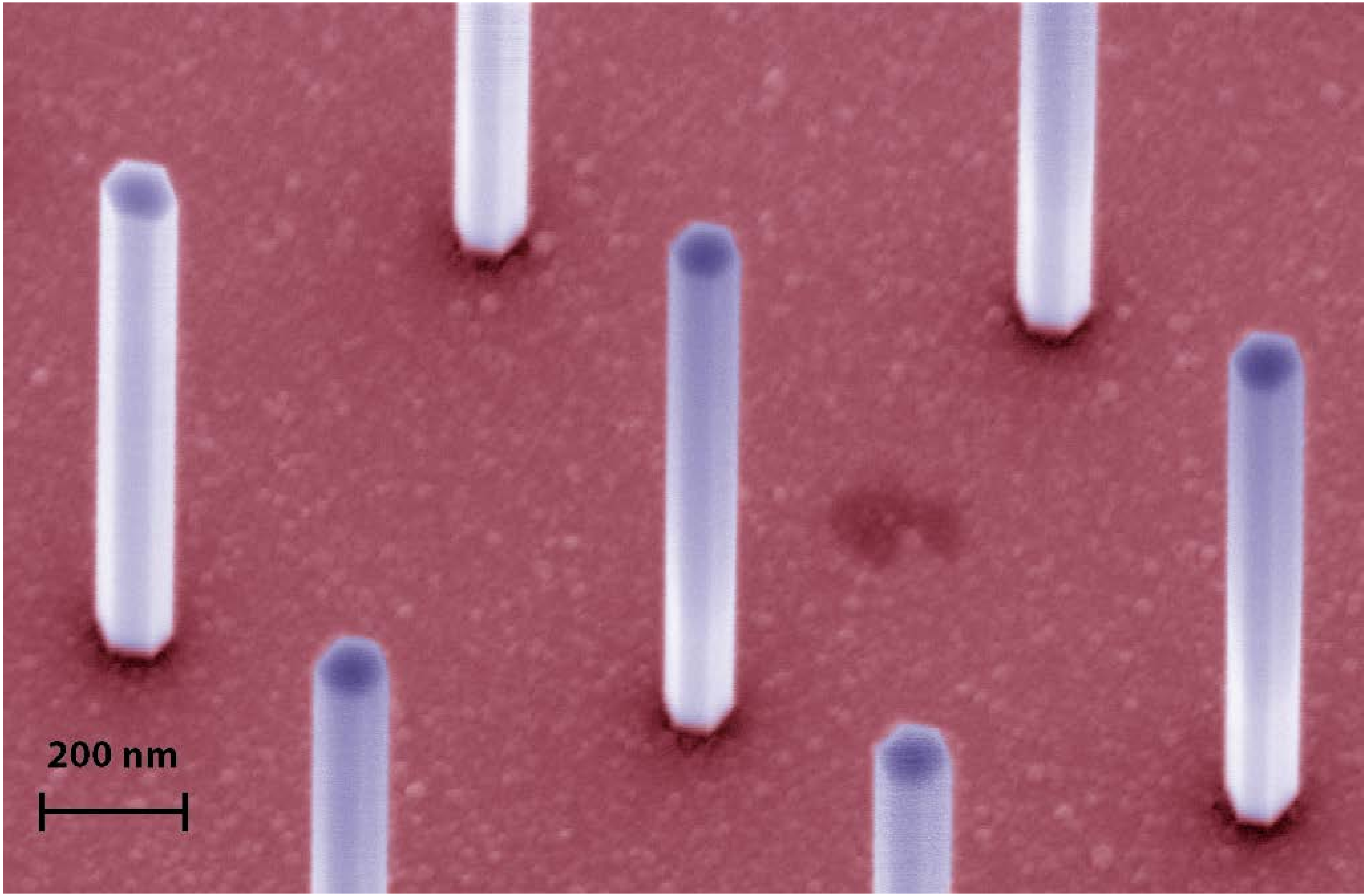
Selective Area Epitaxy of Nanowire Arrays

Metal-Organic Vapor Phase Epitaxy

Harri Lipsanen, Aalto University, Finland

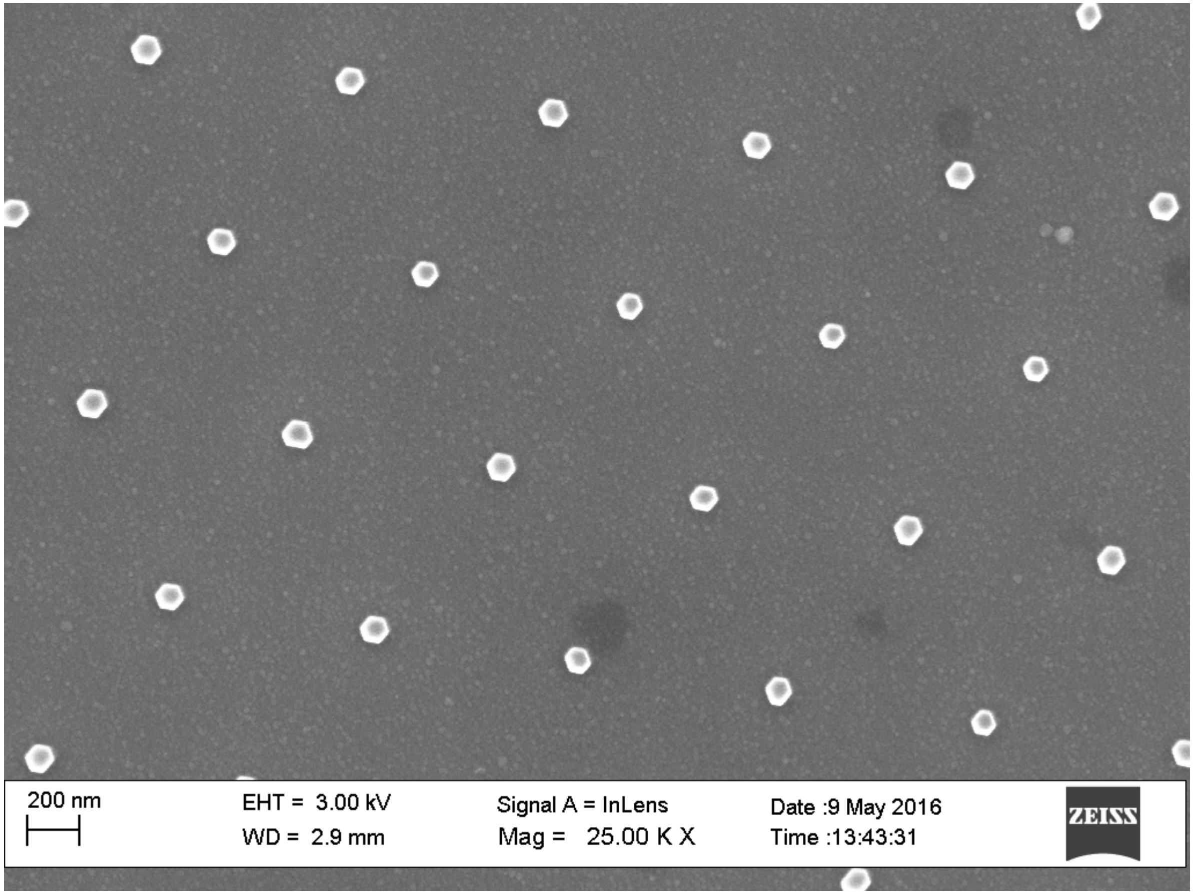


Nanowires with Controlled Diameter and Distance



High-Quality Uniform GaAs Nanowires

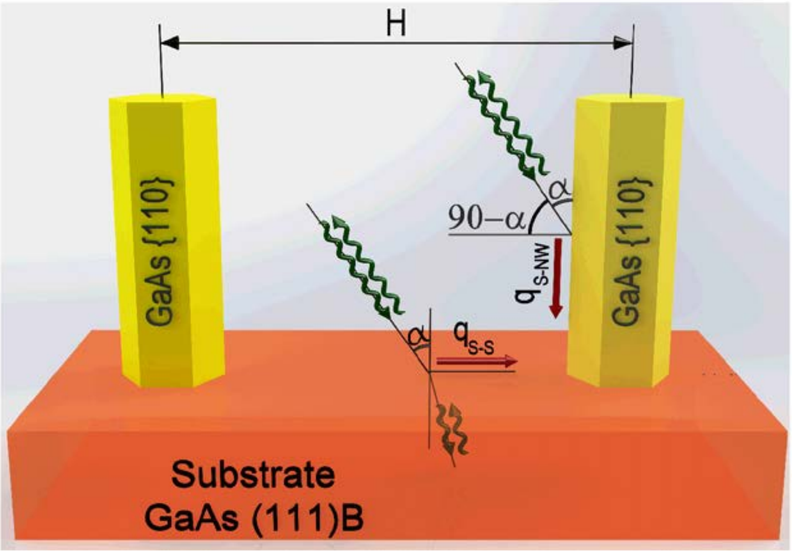
Large inter-NW distance: from 250 nm to 10 μm; Large NW length: L > 10×D

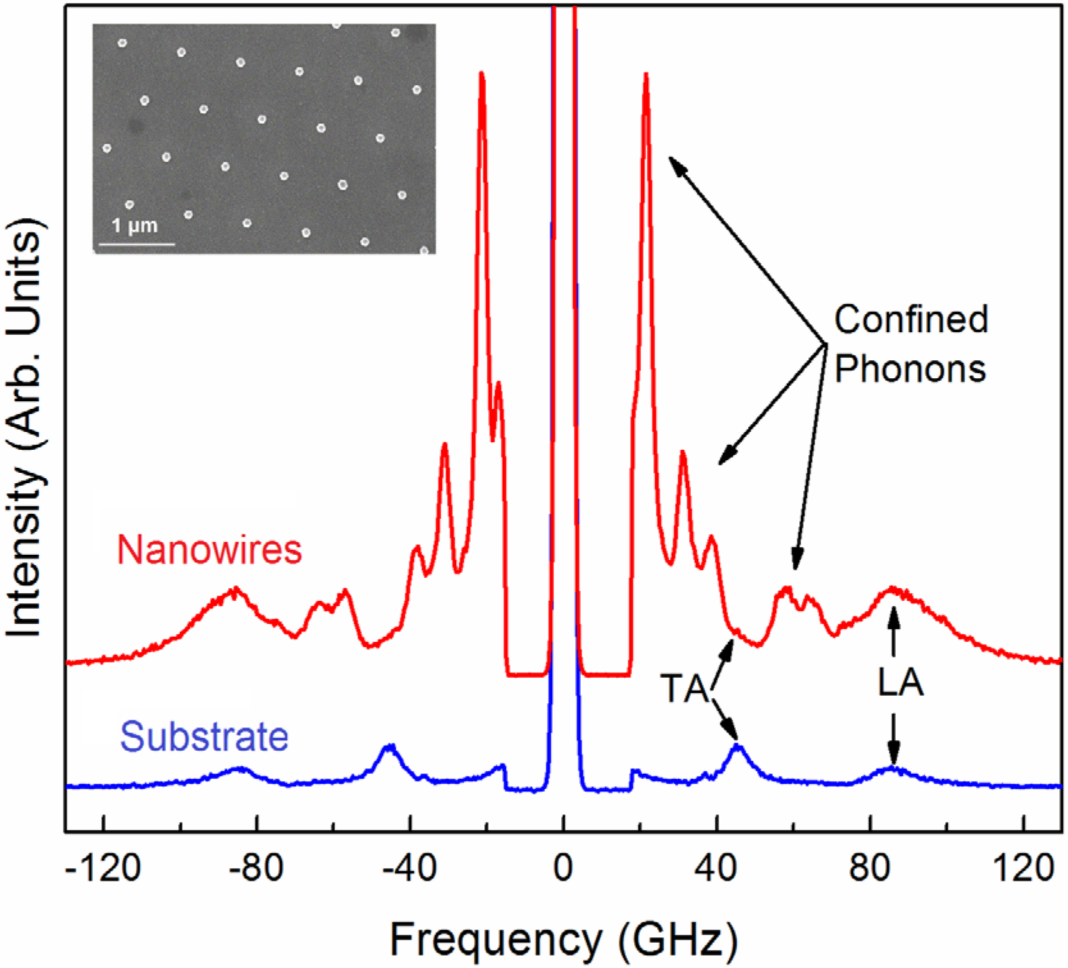


F. Kargar, et al., “Direct observation of confined acoustic phonon polarization branches in free-standing semiconductor nanowires,” Nature Com., 7, 13400 (2016).

Relative standard deviation in diameter D is ~ 3%

Acoustic Phonon Spectrum of GaAs Nanowires





Probing phonon wave vectors:

$$q_B = 4\pi n/\lambda \quad q_B = 97.6\ \mu m^{-1}$$

$$q_{S-S} = (4\pi/\lambda)\sin(\alpha)$$

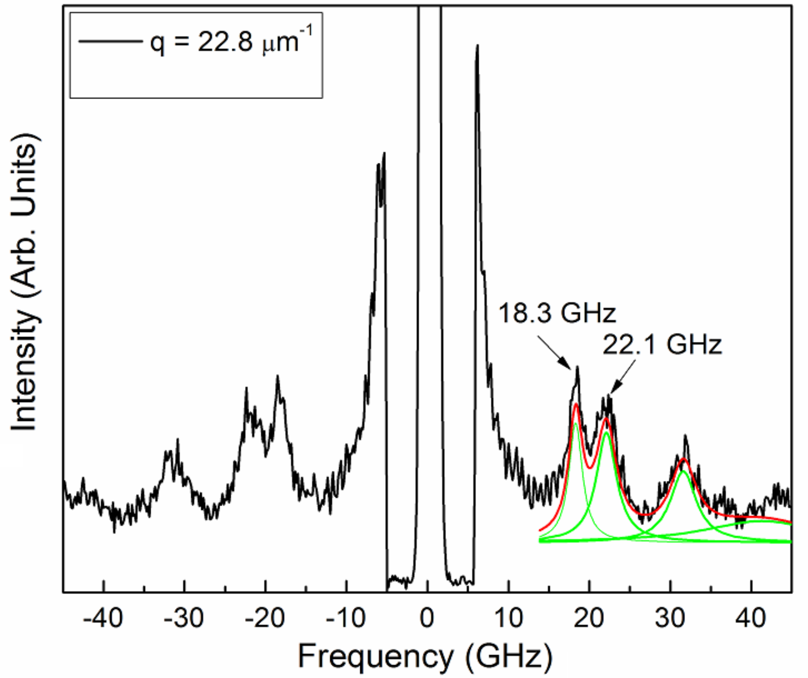
$$q_{S-NW} = (4\pi/\lambda)\cos(\alpha)$$

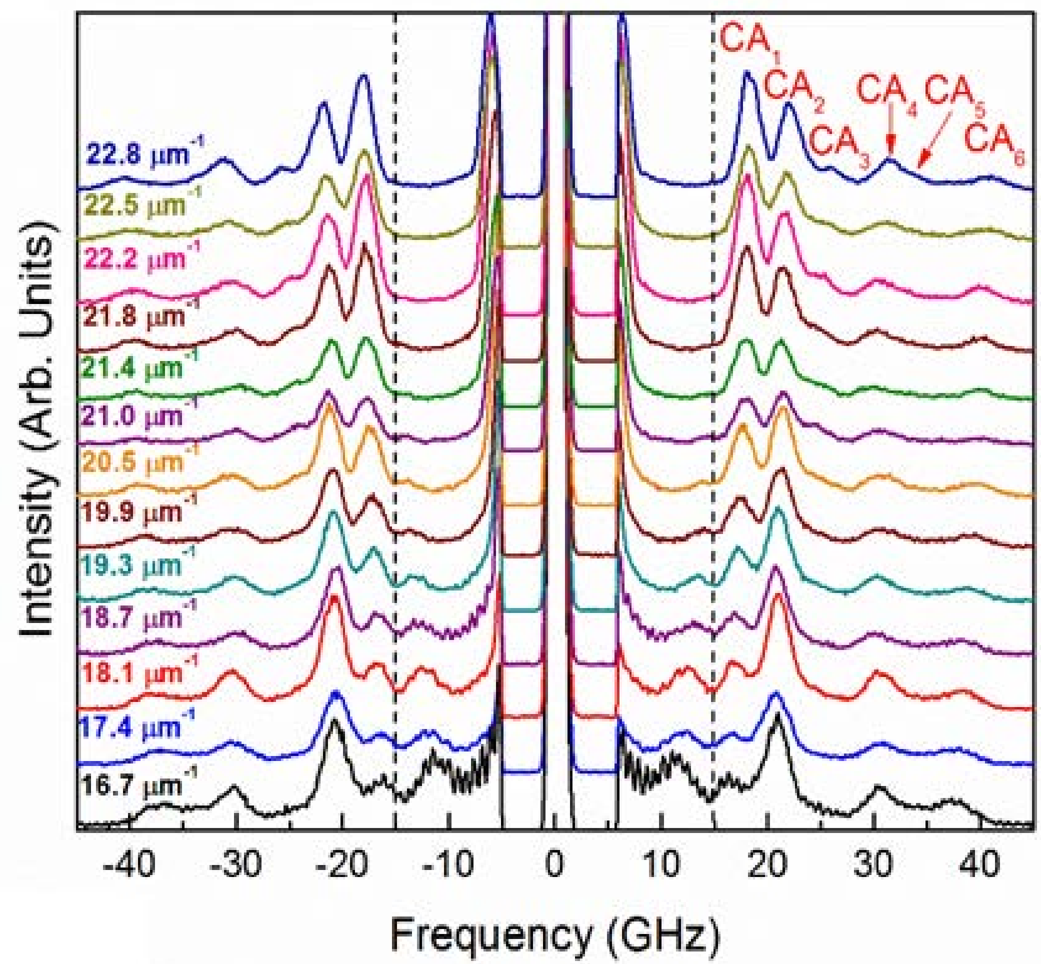
Substrate phonons: $\Delta\omega/\omega = 2n_2/n_1$

D=122 nm

Confined Acoustic (CA) Phonons in Nanowires

Phonon Spectrum Deconvolution





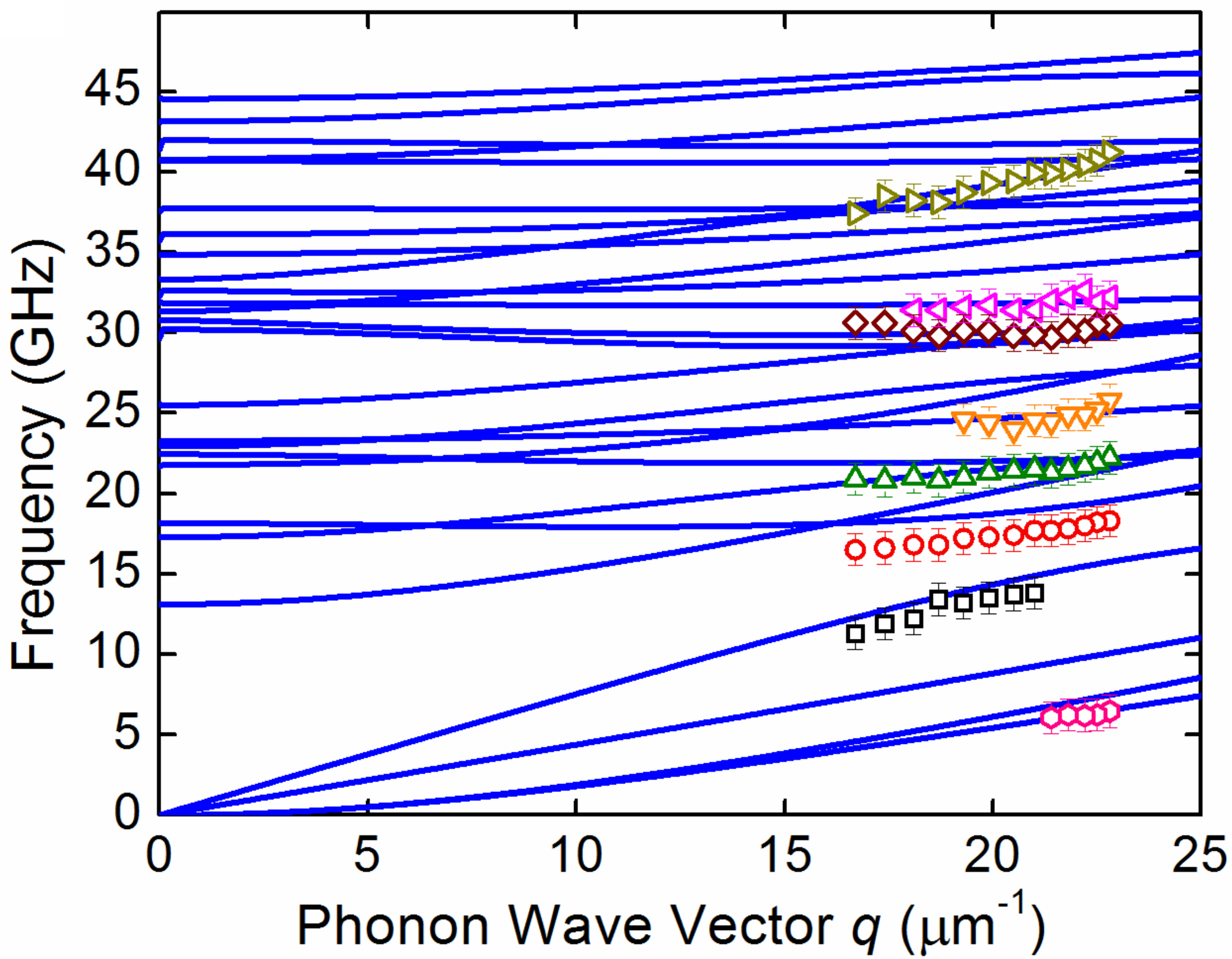
1 THz = 33.36 cm^{-1} = 4.1 meV

$$q_{S-NW} = (4\pi/\lambda)\cos(\alpha)$$

D=122 nm

Changing incident angle one can get dispersion branches.

Confined Acoustic Phonon Dispersion in Nanowires

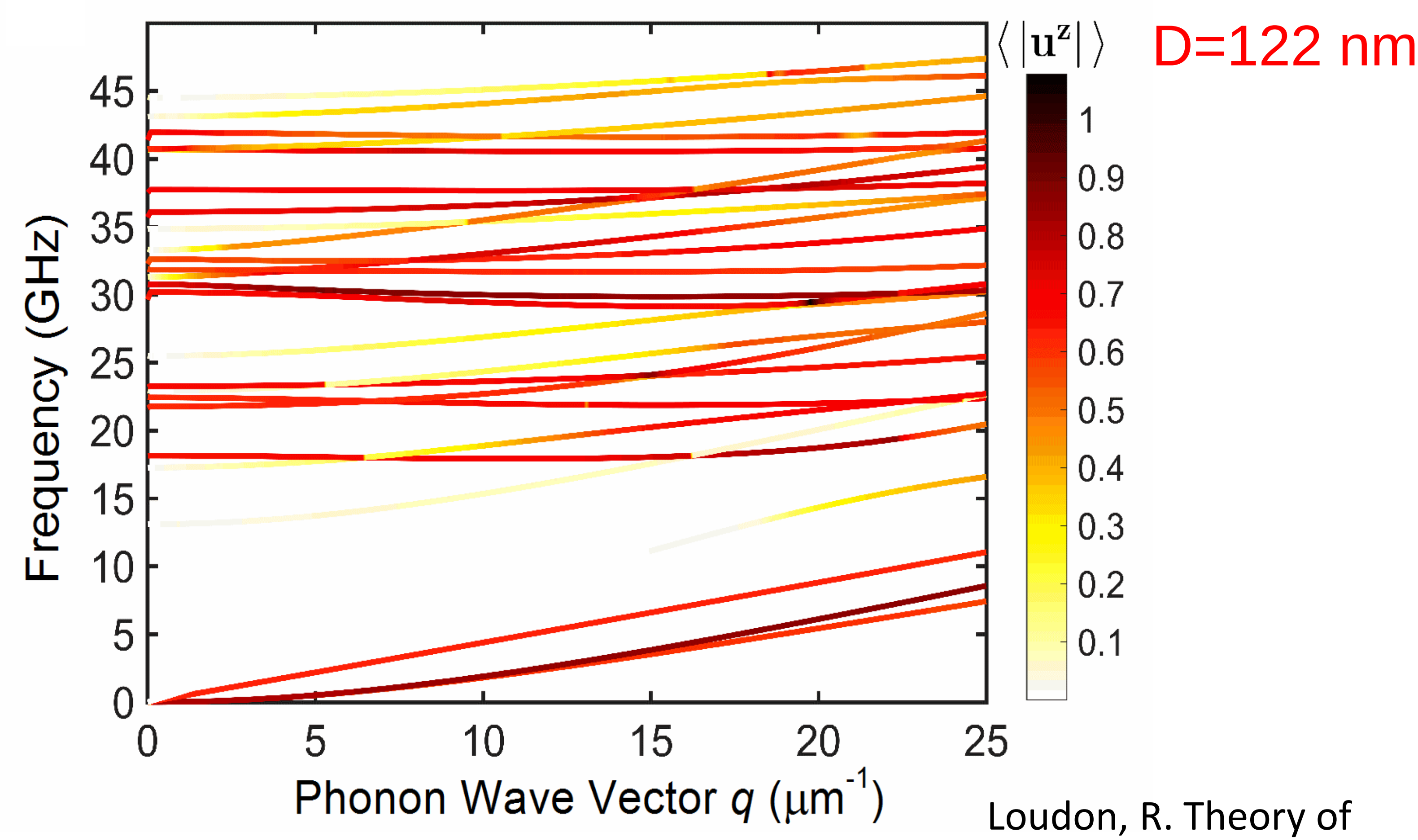


Calculations: Roger Lake, UC Riverside

Measured and calculated phonon dispersion for a GaAs nanowire along [111] direction. The experimental data points are indicated with symbols.

D=122 nm

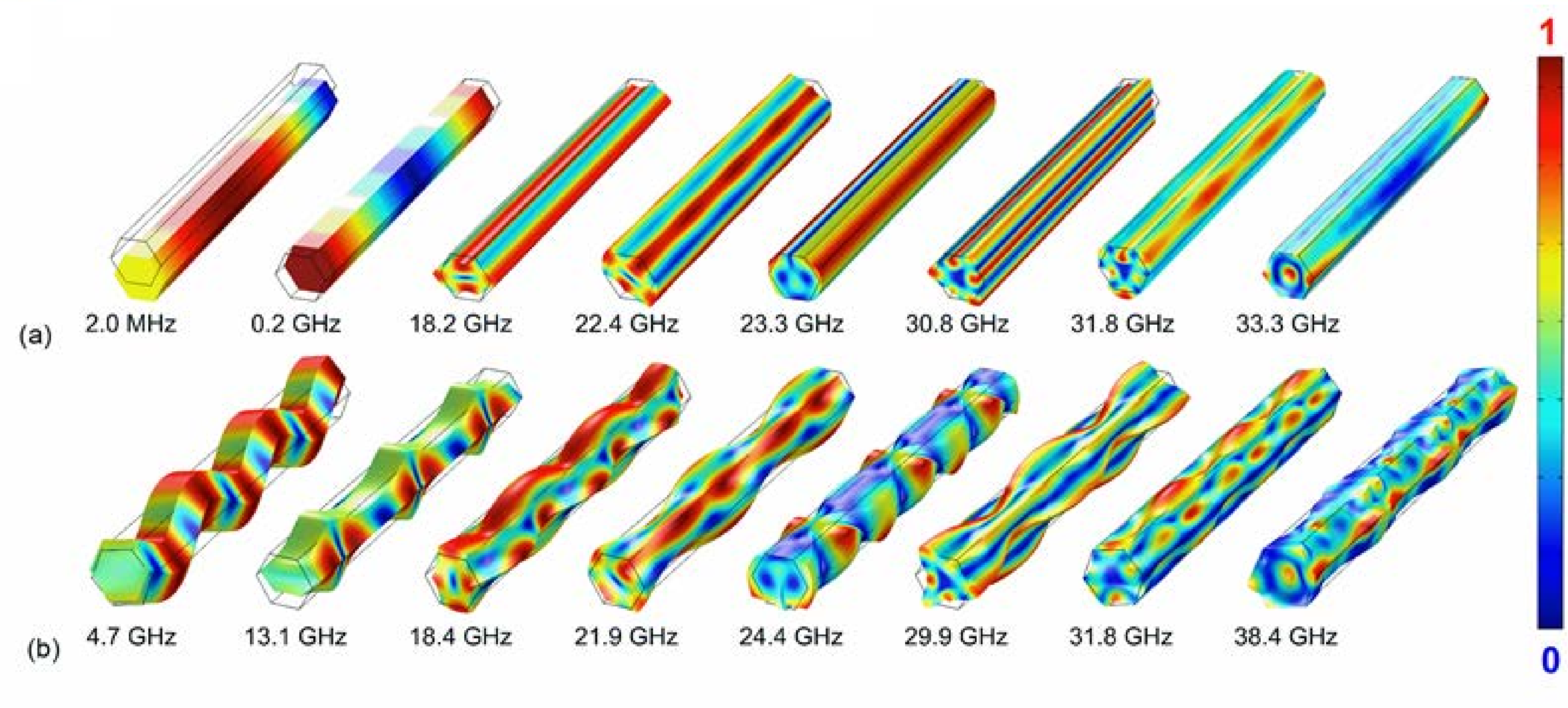
Optically Active Confined Acoustic Phonon Modes



Loudon, R. Theory of surface-ripple Brillouin scattering by solids. Phys. Rev. Lett. 40, 581 (1978).

$$d^2\sigma / d\Omega \, d\omega_s = \left(\zeta \ \omega_I^4 / 16\pi^2 c^4\right) F^2 \left\langle \left| u^z(0) \right|^2 \right\rangle_{q_x,\omega}$$

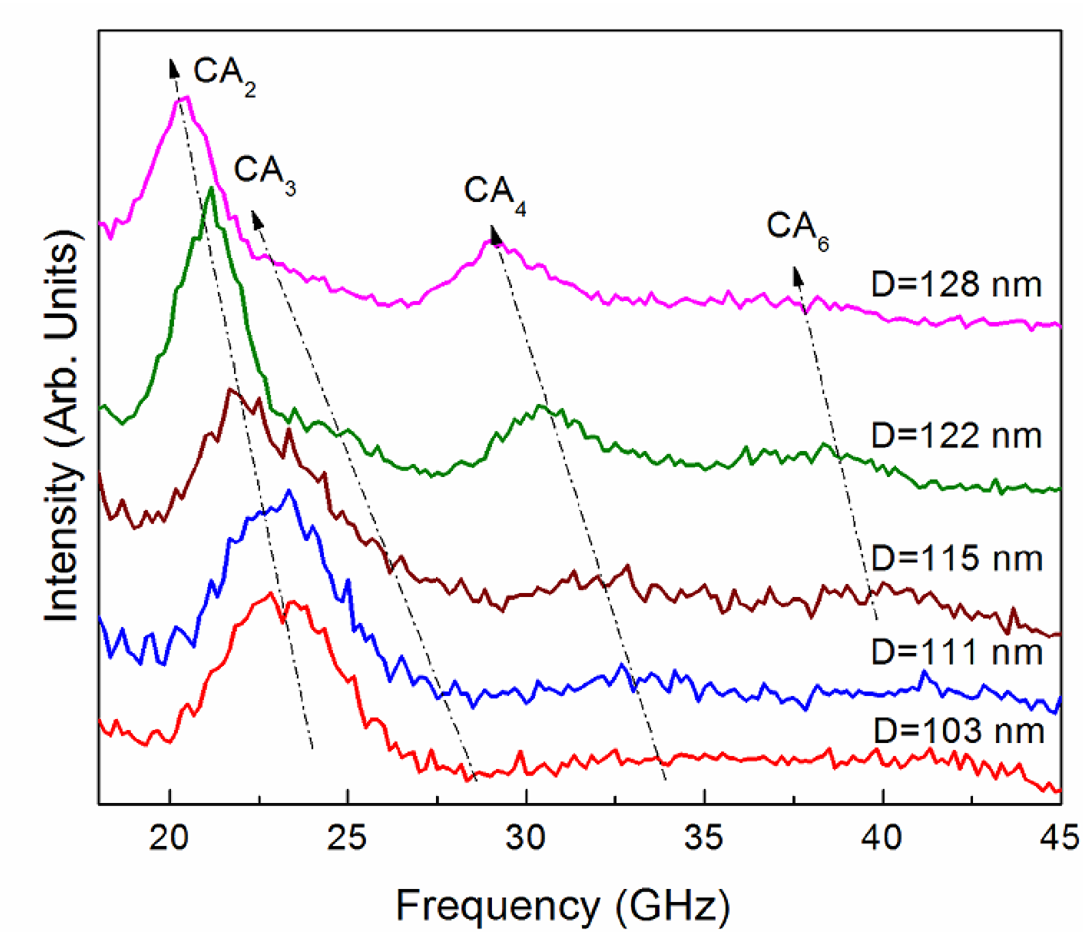
Confined Acoustic Phonon Branches in Nanowires

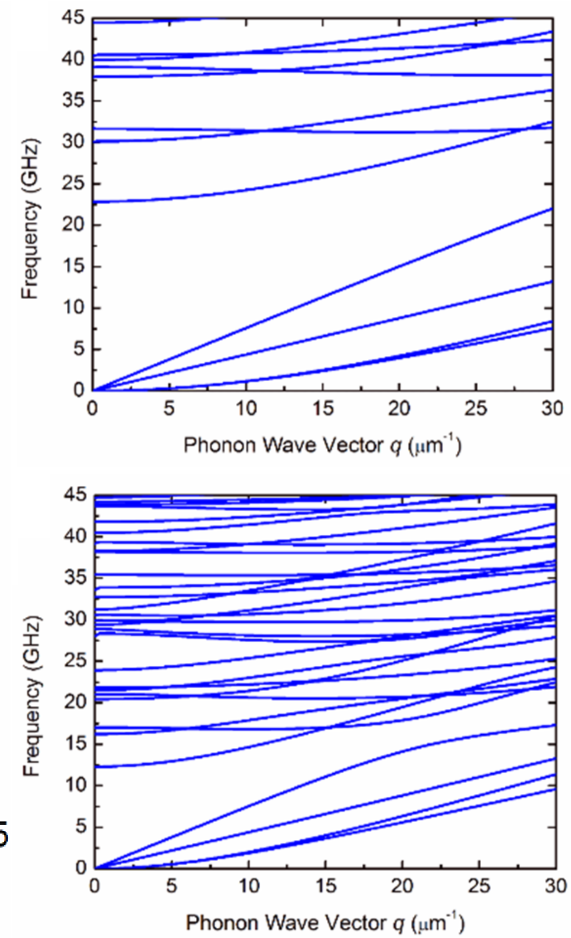


The normalized displacement field of the Brillouin-active phonon modes calculated for a 1-μm long NW for (a) $q_{S\text{-}NW}$ = 0.3 μm^{-1} and (b) $q_{S\text{-}NW}$ = 18.0 μm^{-1}. The red color corresponds to stronger displacement.

Elasticity equation: $\rho\left(\partial^2 u(r)/\partial t^2\right) = \partial S(r)/\partial x_i$ $\quad C_{ijkl}^{[111]} = \Sigma_{\alpha\beta\gamma\delta} U_{i\alpha} U_{j\beta} U_{k\gamma} U_{l\delta} C_{\alpha\beta\gamma\delta}^{[001]}$

Confined Acoustic Phonon Energies in Nanowires

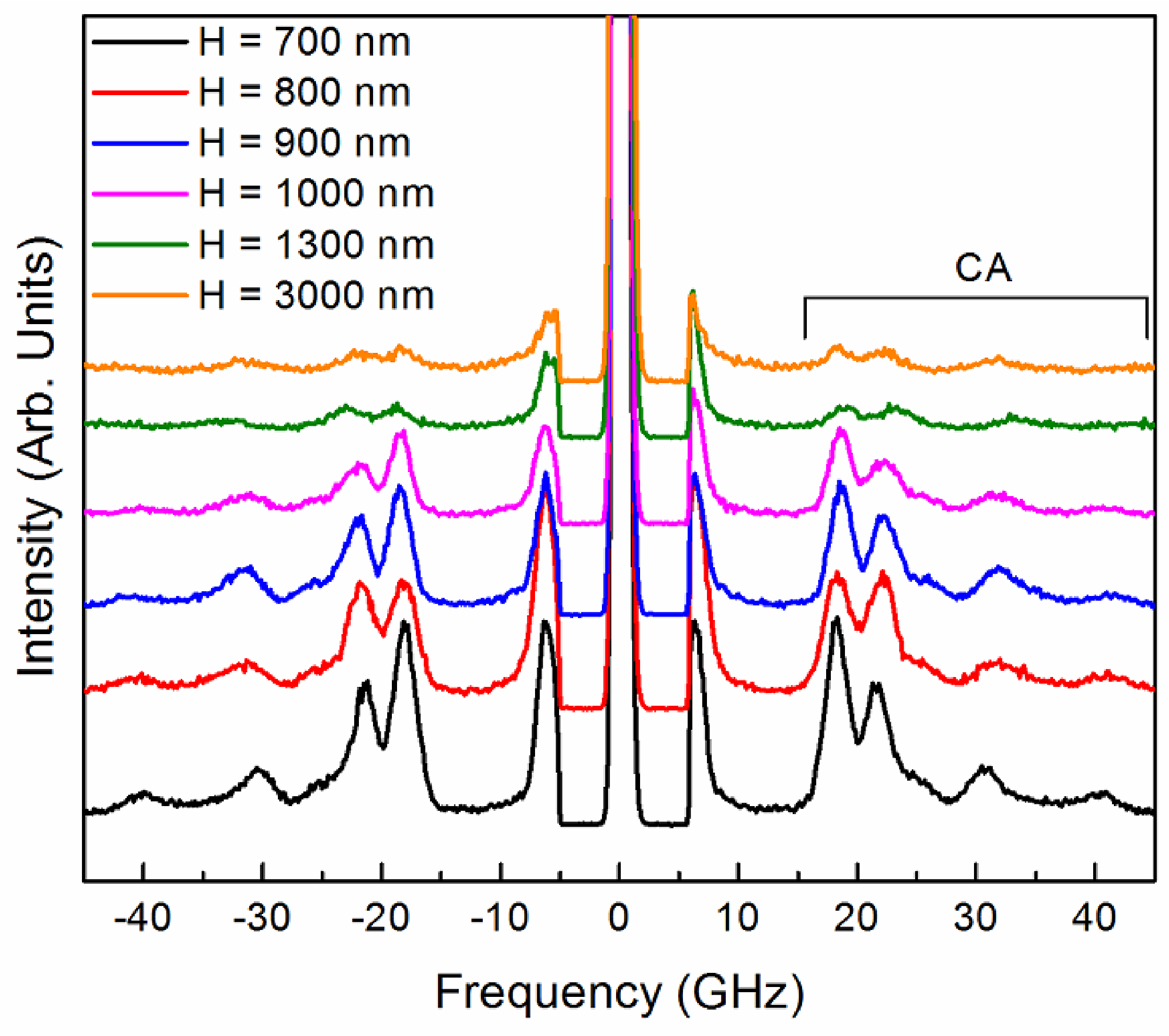




Phonon dispersion of GaAs NW along [111] direction for D = 70 nm (top) and D = 130 nm (bottom).

Brillouin spectrum for NWs with different diameter at a constant probing phonon wave vector q_S-NW=18.1 μm^{-1}. The decrease in the frequency of CA phonons with increasing D is visible. The CA branches show strong diameter dependence even for relatively large D values in the range from ~103 nm to ~128 nm.

Phonon Confinement in Individual Nanowires

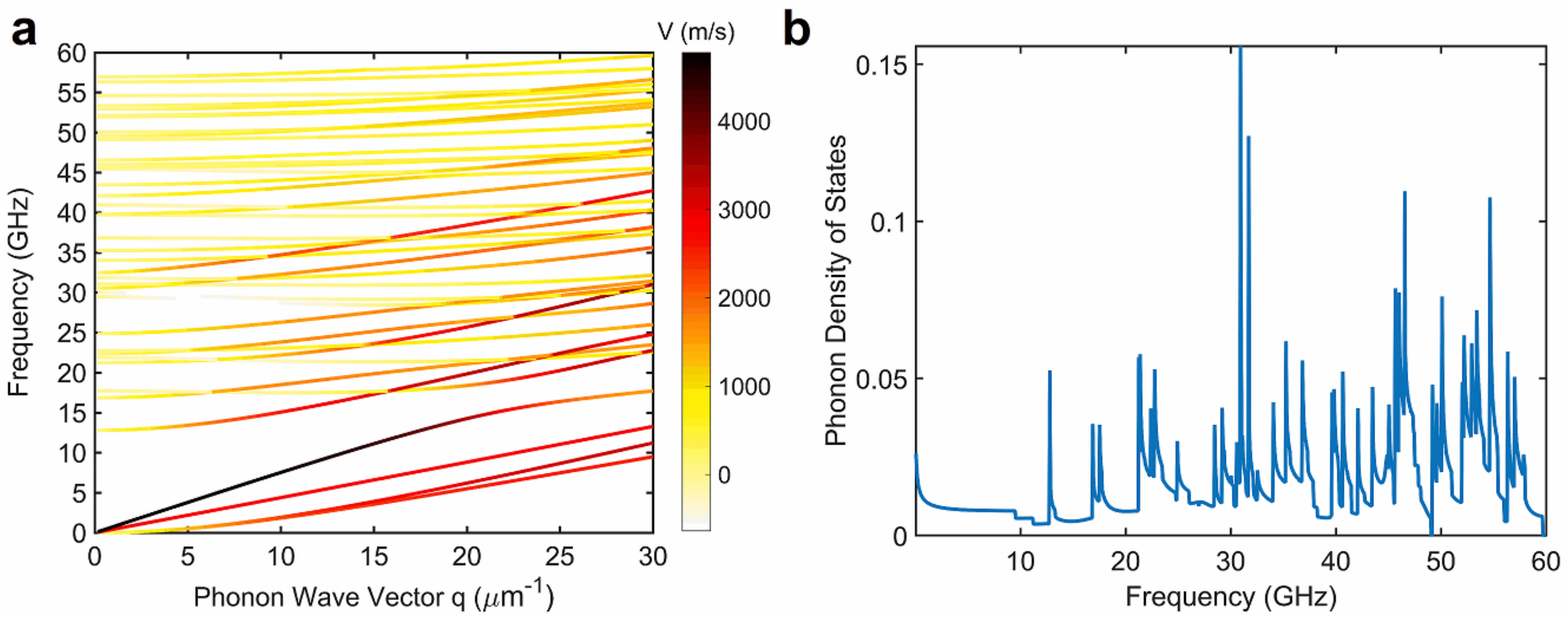


→ Measured spectrum for NWs with the constant diameter D = 122 nm and varying inter-NW distance H. The data are presented for the same fixed accumulation time of 30 minutes.

→ The spectral position of the CA peaks does not depend on H.

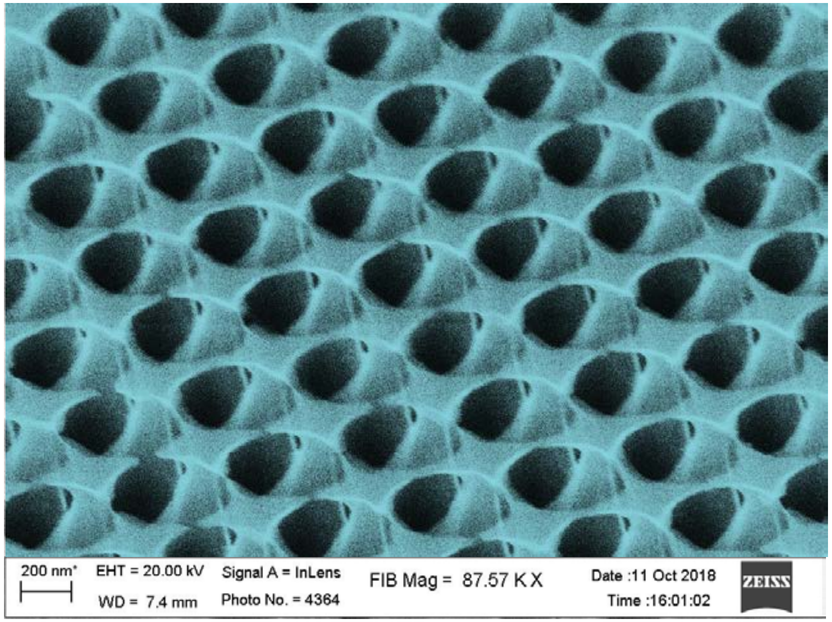
F. Kargar, et al., "Direct observation of confined acoustic phonon polarization branches in free-standing semiconductor nanowires," Nature Com., 7, 13400 (2016).

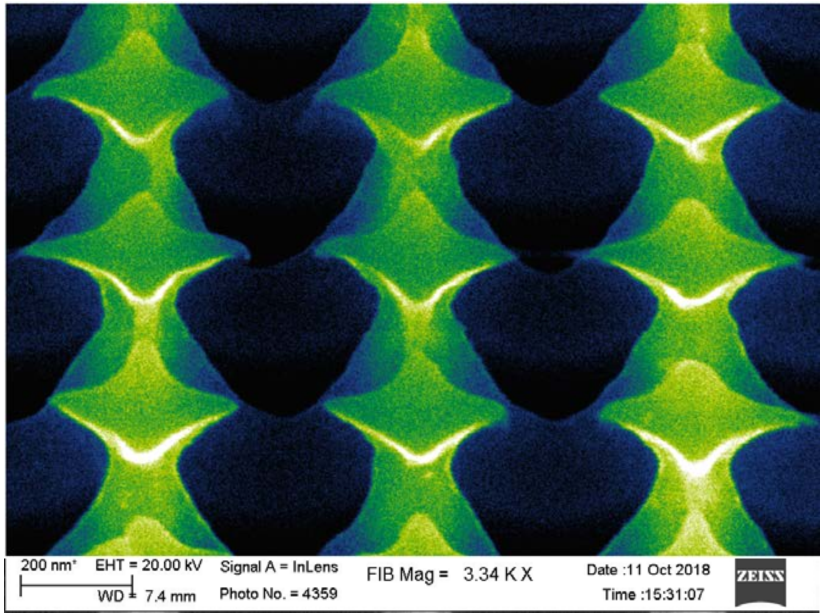
Phonon Group Velocity and Density of States



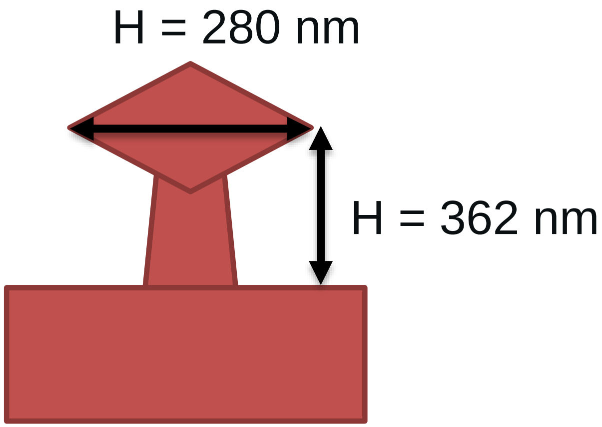
(a) Calculated phonon dispersion indicating the relative contribution of each branch to the mode-averaged phonon group velocity. The change in the color from yellow to black corresponds to increasing contribution to the mode-average phonon group velocity. (b) Calculated phonon density of states. The data indicates that the effect of the confined phonons is significant, leading to reduction of the average group velocity. The phonon density of states is modified as compared to bulk owing to the emergence of the confined phonon bands.

Tuning the Phonon Dispersion



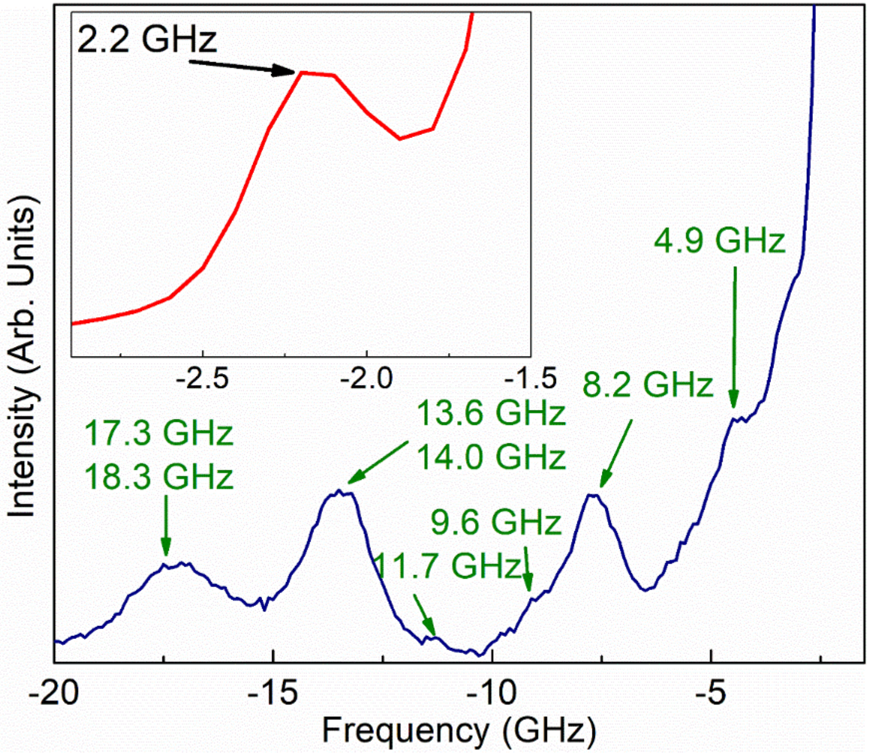


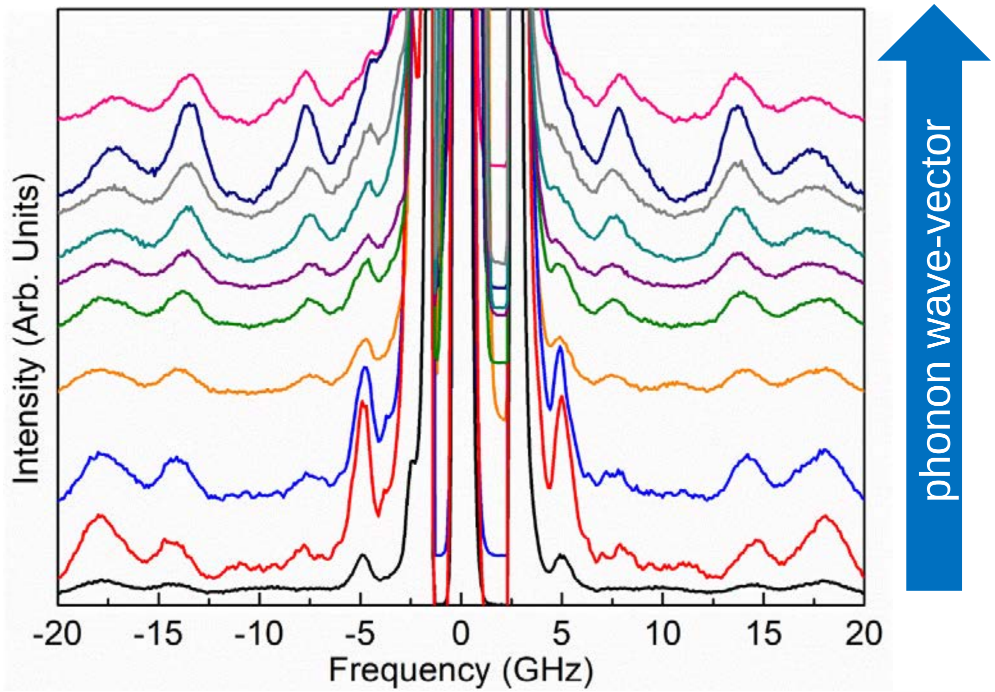
- SEM images of the ordered pillars with specific design
- This pillar array behaves as phononic and photonic material
- Periodicity: 500 nm; Height: 362 nm



Phonon Spectrum of Pillars with Hats

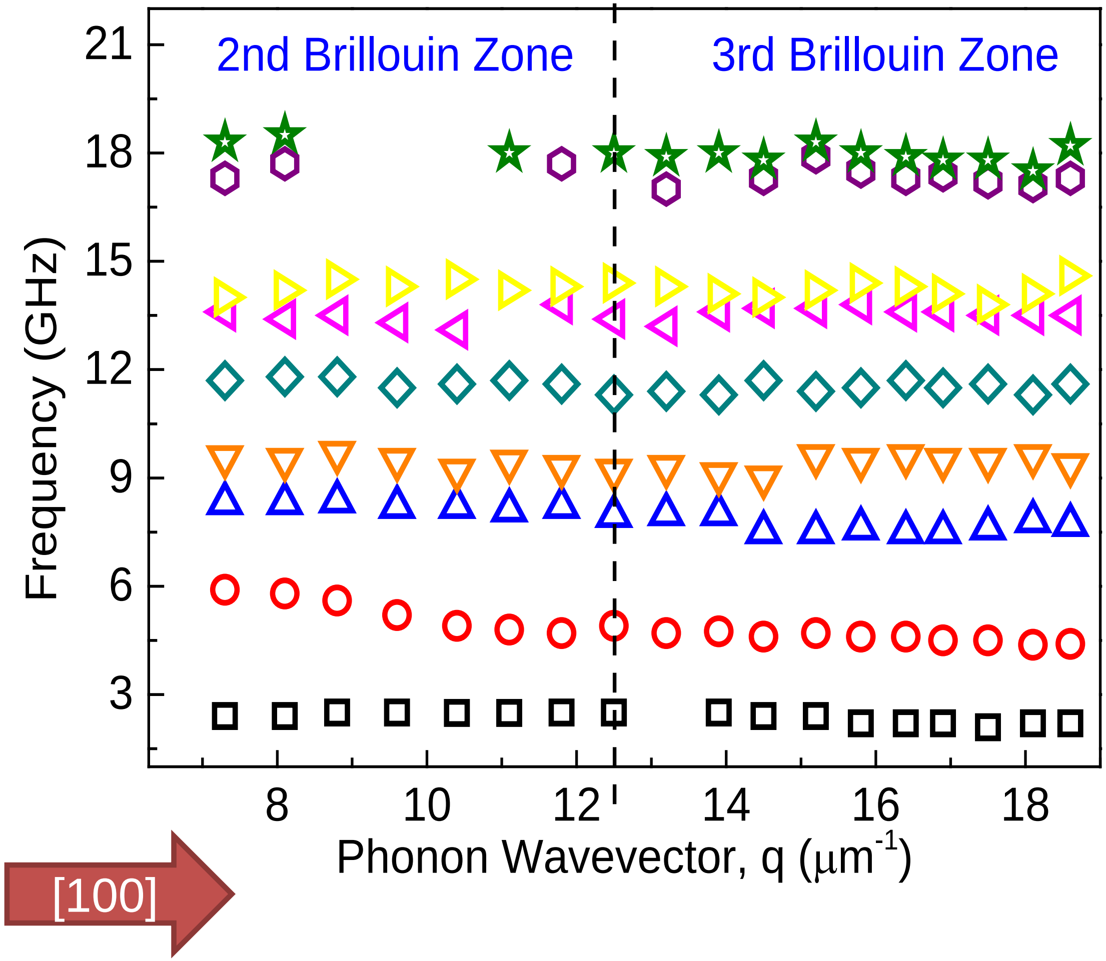
Nine distinct peaks attributed to the phonon spectrum changes due to periodicity or confinement





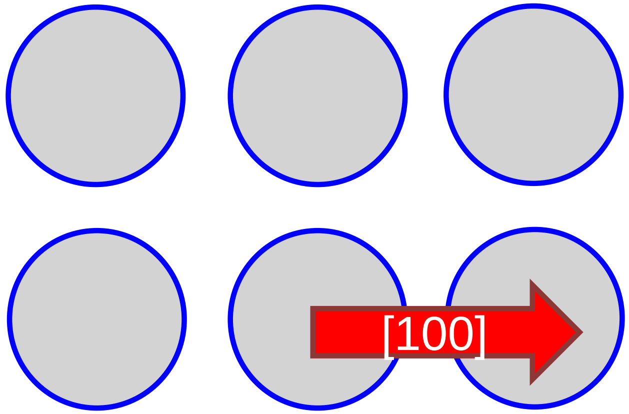
- Evolution of the spectrum with increasing phonon wave-vector
- Samples are patterned and opaque – spectrum is dominated by the surface ripple mechanism
- The phonon wave-vector is changed by changing the angle of incidence

Phonon Dispersion Along [100] Direction



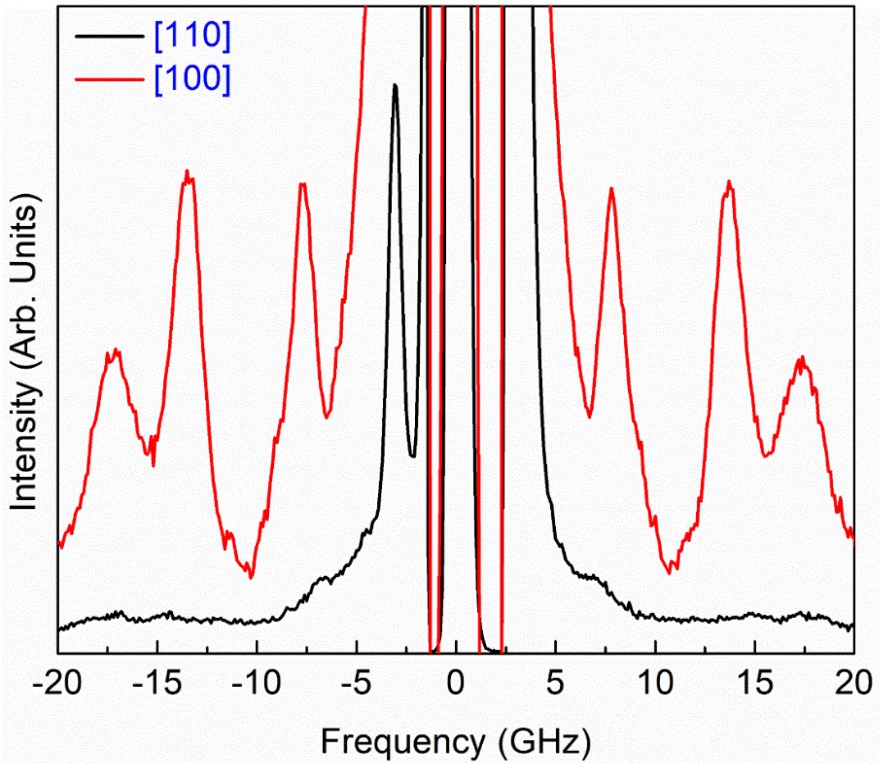
→ Phonon dispersion in [100] direction

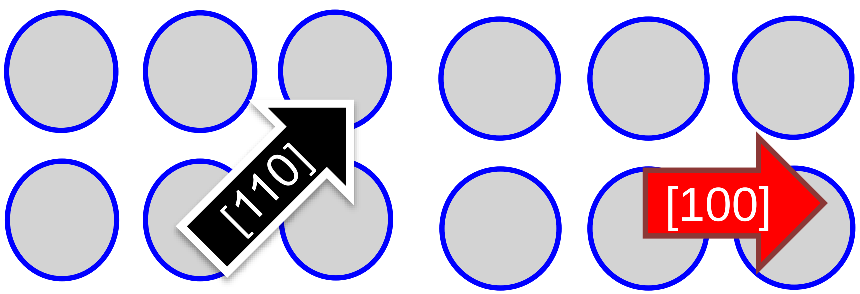
→ Reduced Brillouin zone (BZ) in the periodic structures, probes the phonons from Γ point up to the 3rd Brillouin zone.



→ Peaks for 1st BZ were instrument limited as a result of reflected light transmitted to the interferometer

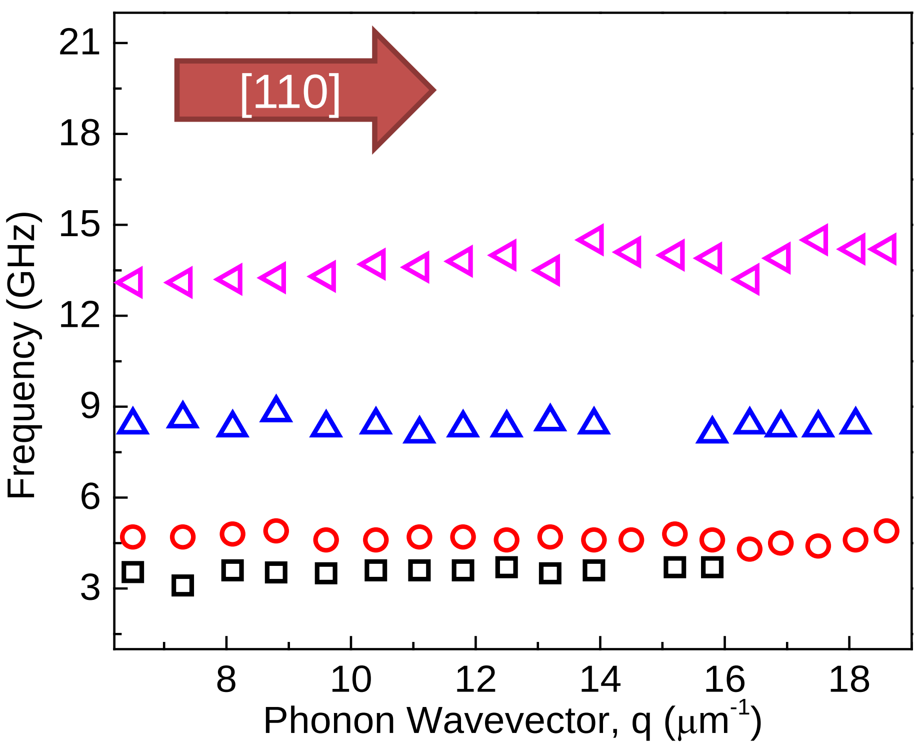
Localization of Phonons in the Hats



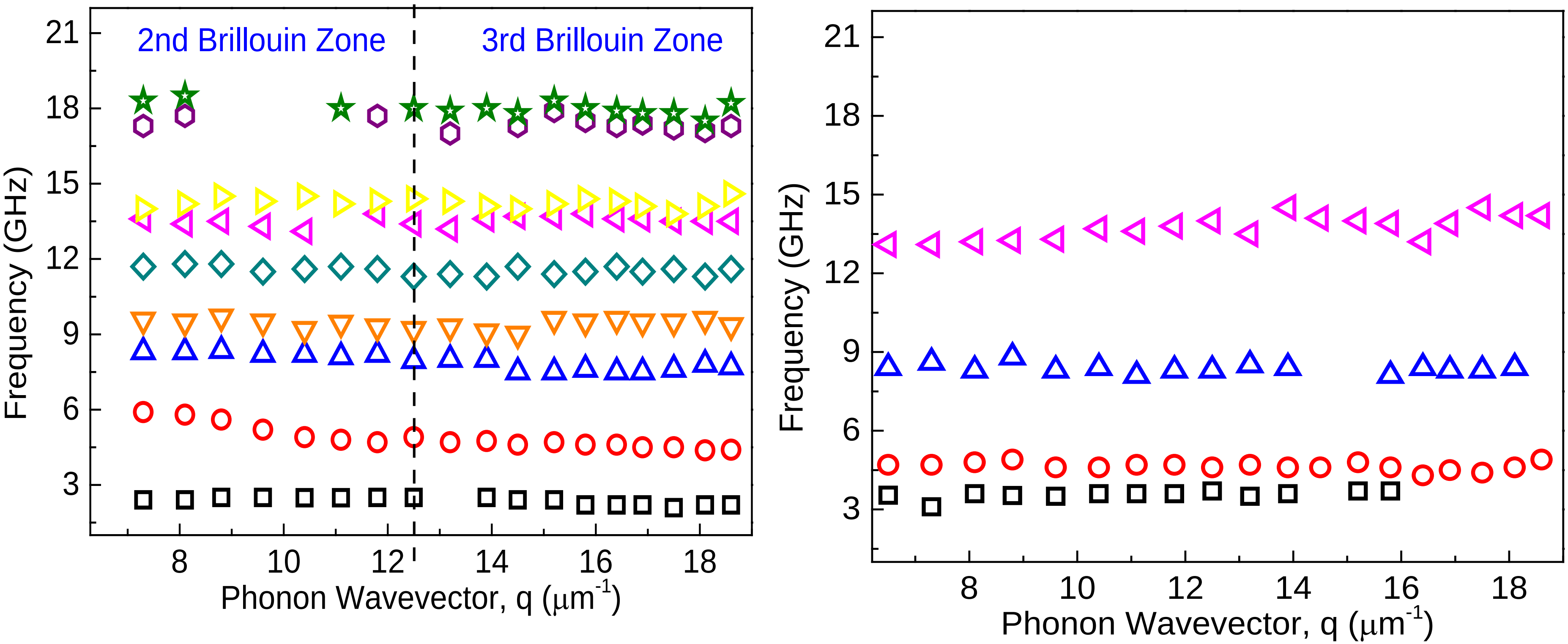


→ Different crystallographic direction: some modes remain the same while others disappear or changed

→ The phonon modes at the same energy are most likely localized phonons



Phonon Modes at Different Directions

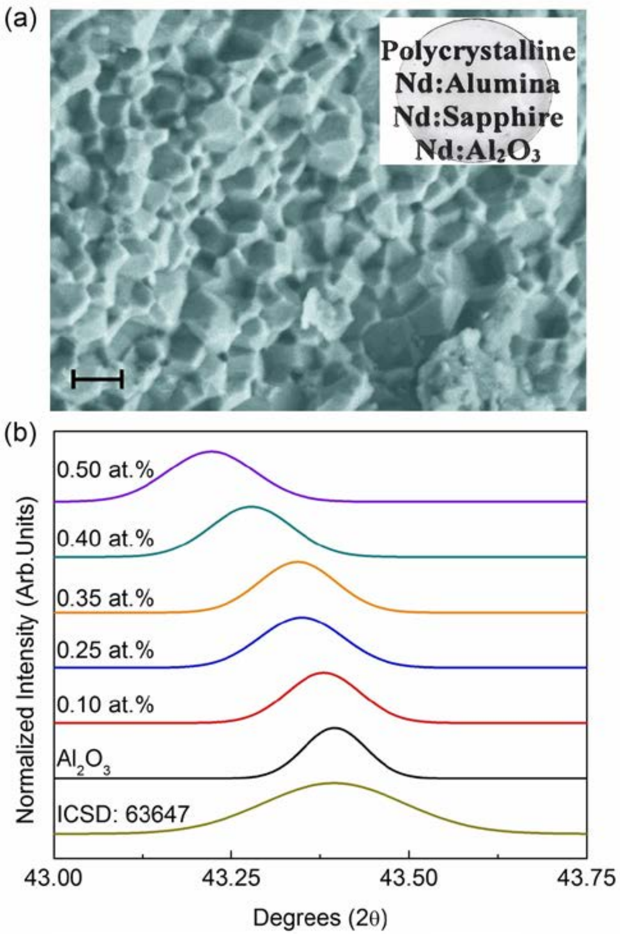


→ Constant energy modes – localized phonons

→ Disappearing phonon modes – result of the rotation and changed dispersion or weak interaction with light

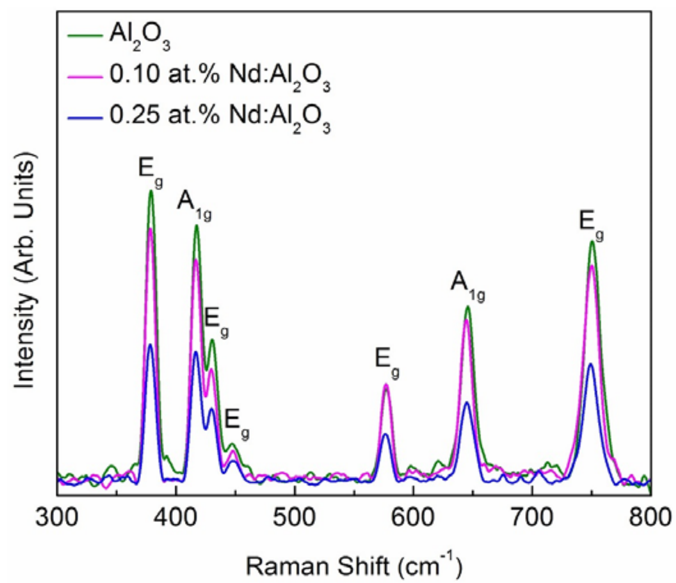
Engineering Phonons with Dopants

Collaboration: Javier Garay (UCSD)
Current Activated Pressure Assisted Densification (CAPAD) method



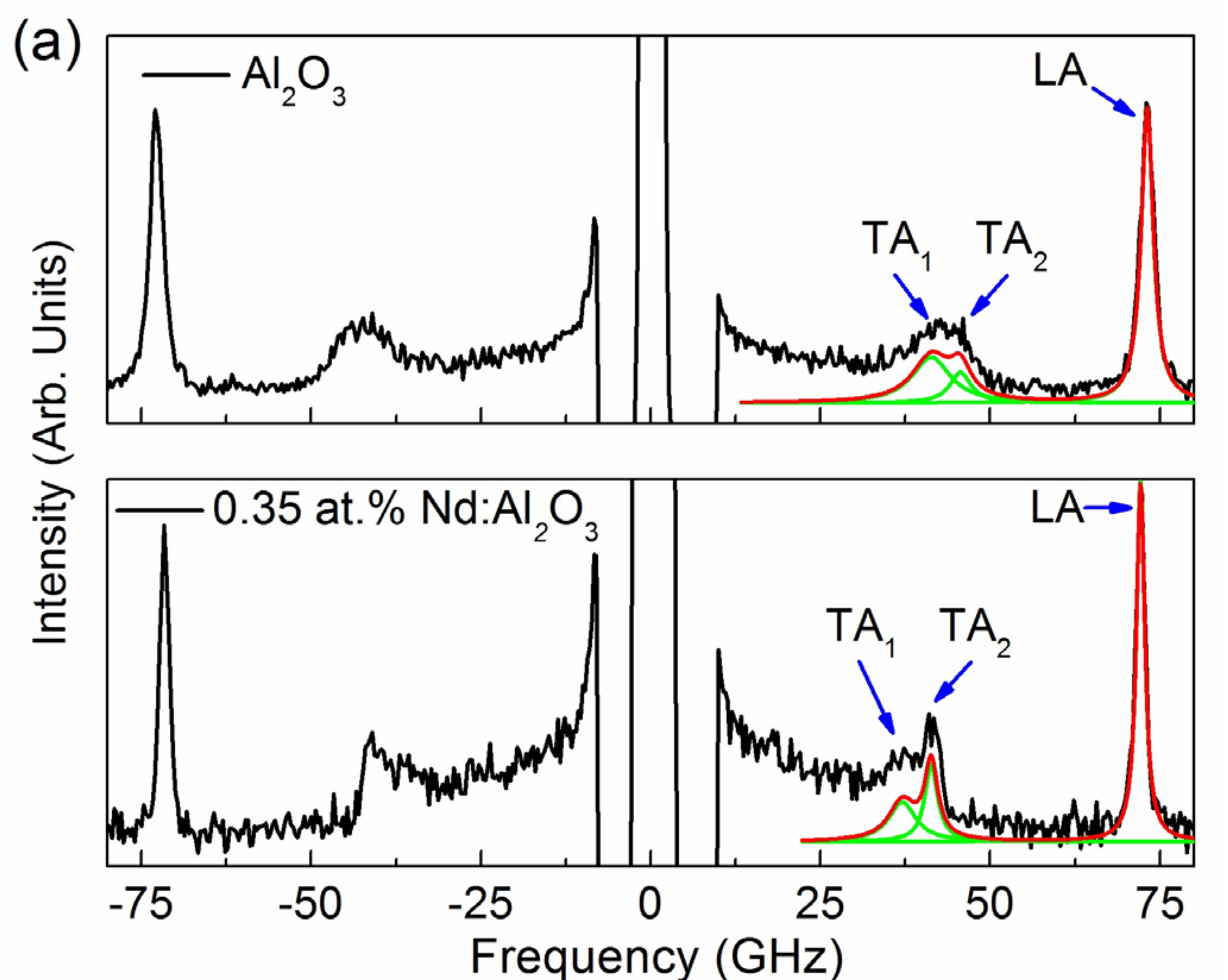
Element	Ionic Radius (nm)	Radius ratio	Weight (amu)	Weight ratio
Al	0.057 (3+)	1	26.98	1
Cr	0.064 (3+)	1.12	52	1.92
Er	0.088(3+)	1.54	167	6.19
Nd	0.115 (3+)	2.02	144	5.34

Transparent Al_2O_3 crystals with Nd, Cr, Er, and a combination of atoms used as substitutional dopants. The ionic radius and atomic mass of atoms are different from those of the host Al atoms.



F. Kargar, *et al*., "Acoustic phonon spectrum engineering in bulk crystals via incorporation of dopant atoms," Appl. Phys. Lett., 112, 191902 (2018).

Brillouin Spectrum of Al_2O_3 with Nd



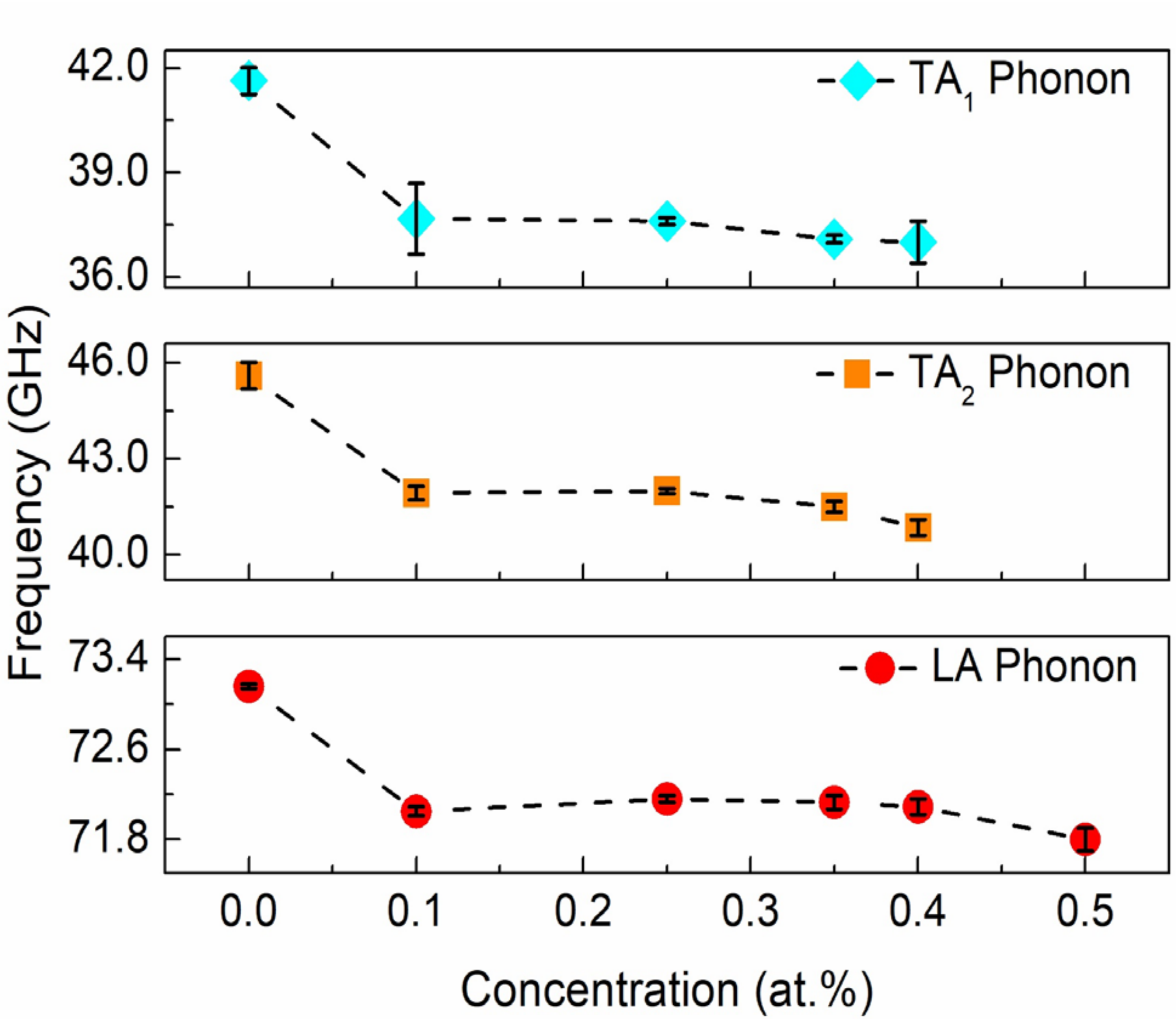
n=1.767 at λ=532 nm

Evolution of the spectrum with increasing Nd doping.

Decrease in frequency of LA and TA phonons of pure Al_2O_3 with increasing the Nd density to 0.1% and more.

F. Kargar, *et al.*, "Acoustic phonon spectrum engineering in bulk crystals via incorporation of dopant atoms," Appl. Phys. Lett., 112, 191902 (2018).

Change in the Frequency of Phonons

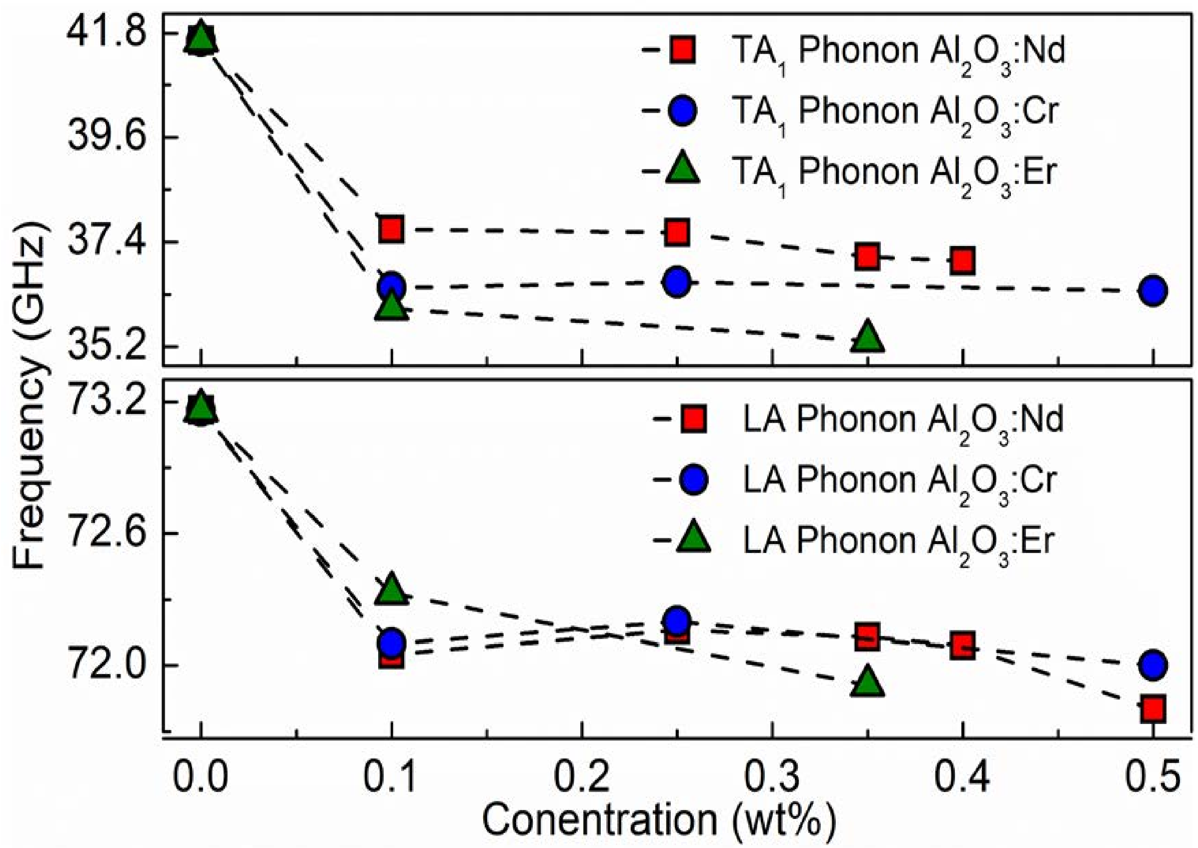


Peak position of LA and TA phonon polarization branches in Brillouin spectra versus Nd density.

The frequency of LA and both TA phonon branches decreases with increasing Nd concentration non-monotonically.

The frequency and velocity of the transverse acoustic phonons decrease by ~4 GHz and ~600 m/s, respectively, at the Nd density of only ~0.1 %.

Effect of the Dopants on Phonons



$$\Gamma = \sum_i f_i \left[\left(1 - M_i / \overline{M}\right)^2 + \varepsilon\left(\gamma\left(1 - R_i / \overline{R}\right)\right)^2 \right]$$

Due to the small concentration of dopants, the atomic mass difference, is unlikely to change the frequency of vibrations.

The atomic mass ratio between Al and Nd and Er is large while it is smaller for Cr as dopant.

Lattice distortion created by larger atoms is the possible mechanism.

Abrupt decrease at the smallest concentration of dopants (0.1%) followed by a much weaker dependence at higher concentrations: adding a few more atoms would not substantially increase the plane separation.

Conclusions and Outlook

→ We proved conclusively the confined nature of acoustic phonons in individual nanowires

→ Observed up to 10 confined phonon polarization branches

→ Confined phonon branches are optically active owing to their hybrid nature

→ Phonon confinement start to take place in nanowires with diameters as large as D=128 nm – order of magnitude large than the grey phonon MFP

→ One can engineer phonon thermal transport and phonon – electron interaction at low temperature; confinement effects are expected even at room temperature in certain structures

→ We demonstrated engineering of the fundamental acoustic phonon dispersion branches in bulk materials via introduction of dissimilar atoms

Acknowledgements

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