Phonons and Thermal Conductivities of Ge Quantum Dot Superlattices

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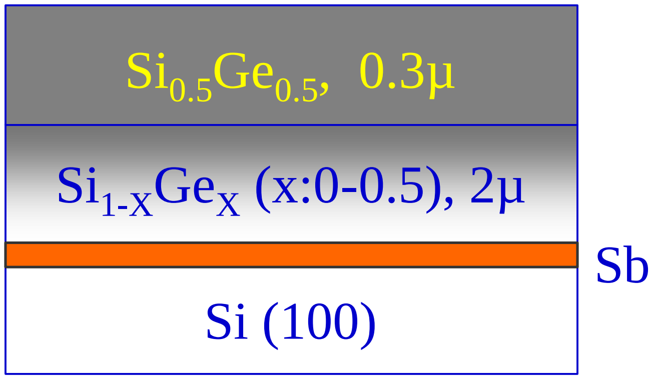
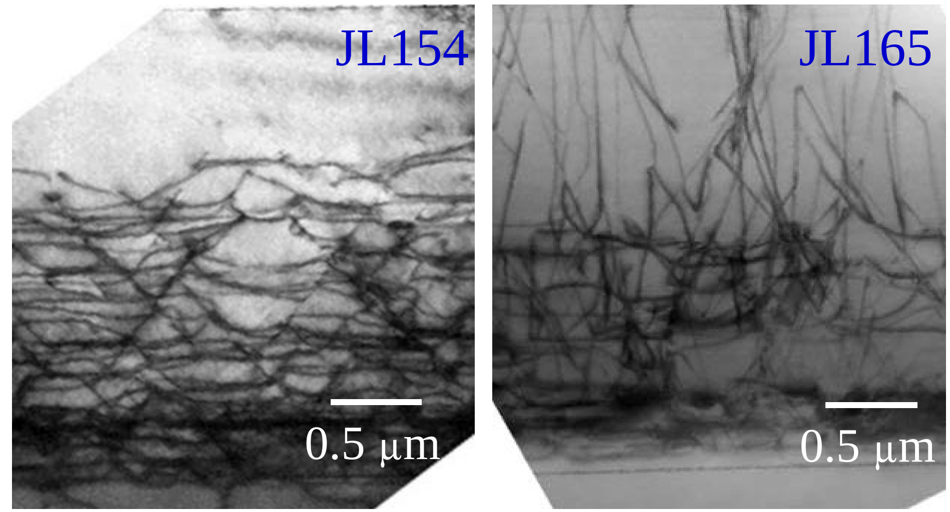
The fifth-year DoD MURI-ONR program on thermoelectrics review

Outline

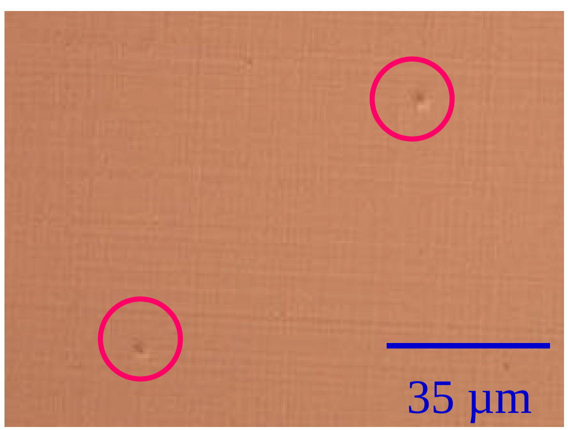
- Part I: Review for research work in five years
  - SiGe growth
    - Graded SiGe buffer layer growth technique by Sb mediation
    - Relaxed SiGe layers by low-temperature Si
    - Si/Ge quantum well superlattices
    - Ge quantum dot superlattices
  - Phonon measurement and modeling
    - Lattice thermal conductivity reduction due to phonon confinement effect—quantum wells and quantum wires
    - Multiple scattering effect/Strain relaxation effect in Ge quantum dot superlattices
    - Thermal conductivity modeling of Ge quantum dot superlattices
- Part II: The fifth-year research
  - Optical phonons in Ge QD SLs: Strain relaxation effects
  - Acoustic phonons in Ge QD SLs: multiple scattering effects
  - Thermal conductivity of Ge QD SLs measurements/modeling

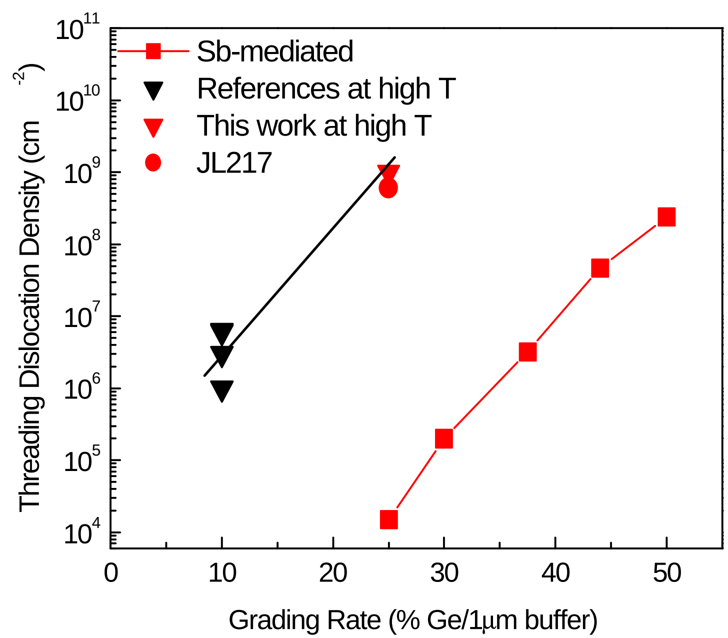
Sb-mediated SiGe buffers





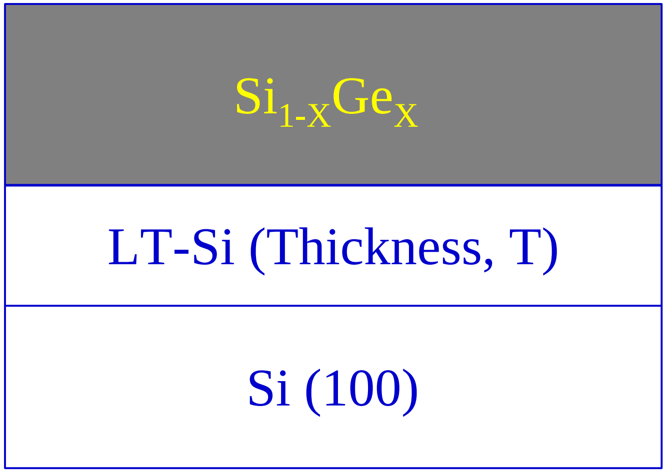
JL154: 1 ML Sb before growth
Growth T=510°C

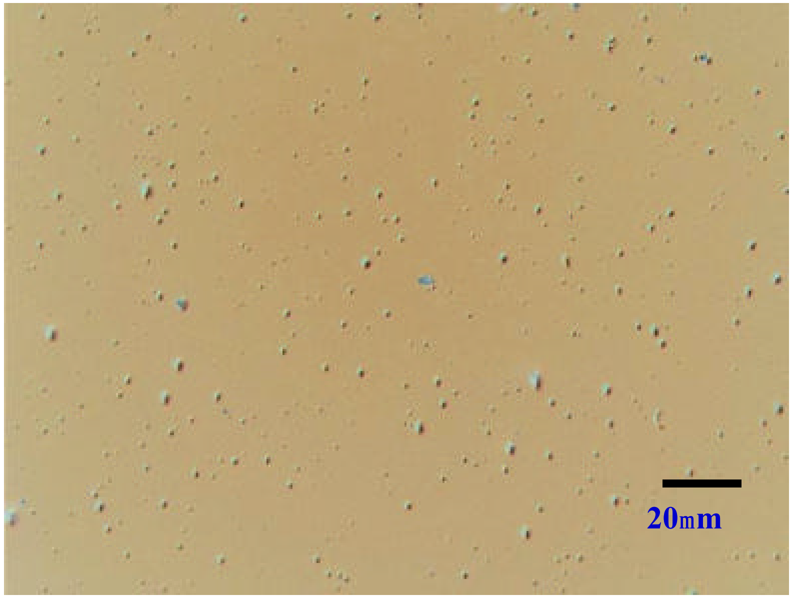




JL154: 1.5×10^{4} cm^{-2}

LT-Si assisted SiGe buffer

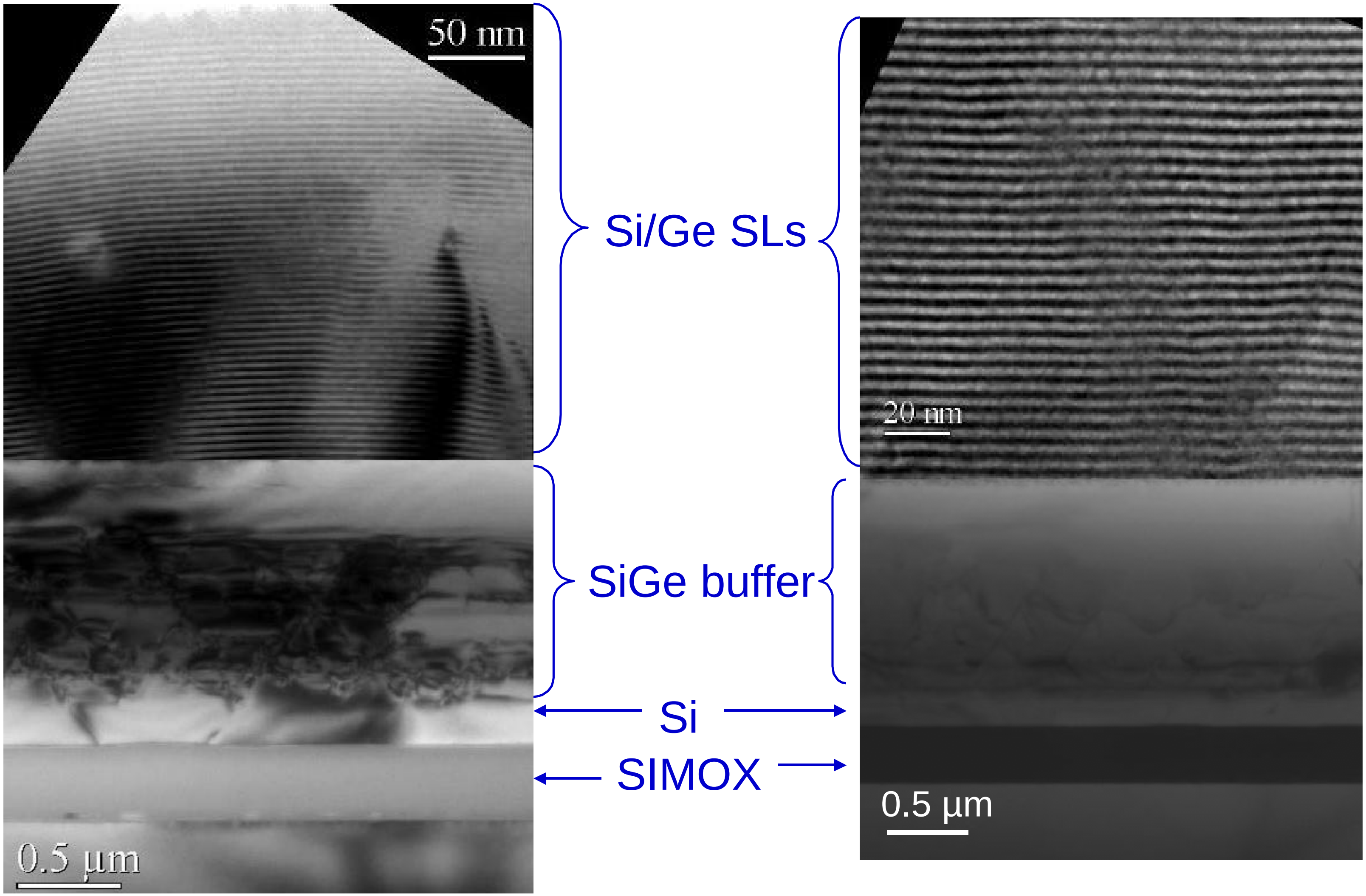




- Mechanism: Point defect injection
  - 100% relaxation
- How to get low dislocation density and smooth surface
  - Thickness of LT Si layer
  - Temperature of LT Si layer

- 500nm $Si_{0.7}Ge_{0.3}$ film on 200 nm 400°C LT Si buffer
- Threading dislocation density: 1.5×10^5 cm^{-2}

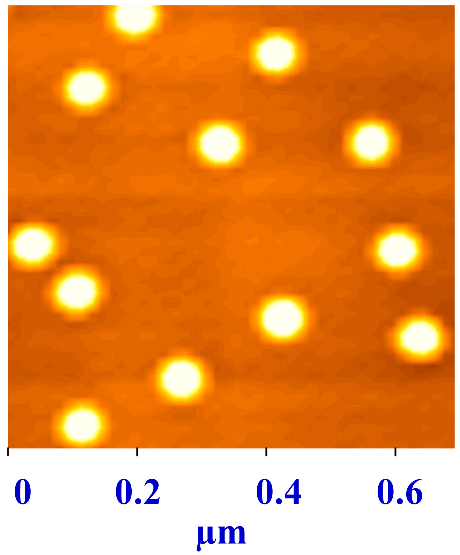
Si/Ge Quantum-well SLs

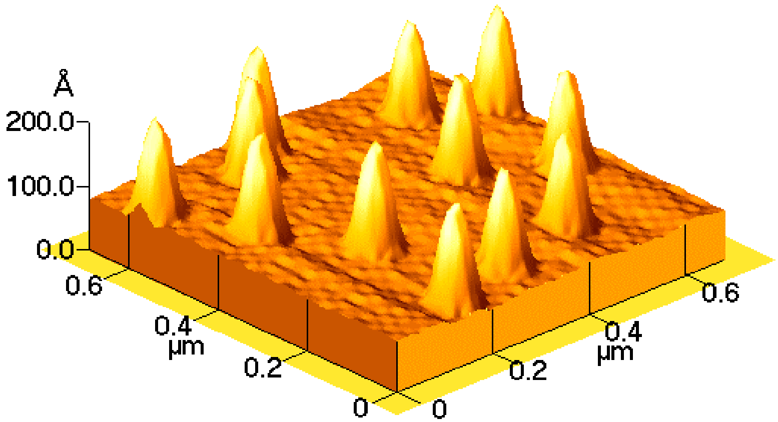


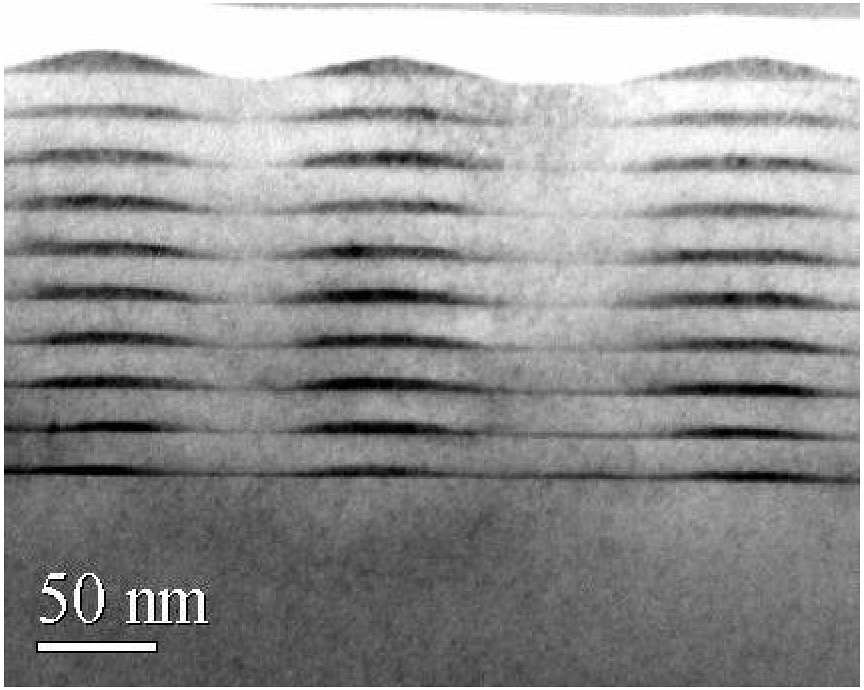
JL215 (250-period Si(20Å)/Ge(20Å))

JL197(100-period Si(20Å)/Ge(20Å))

Ge Quantum Dots



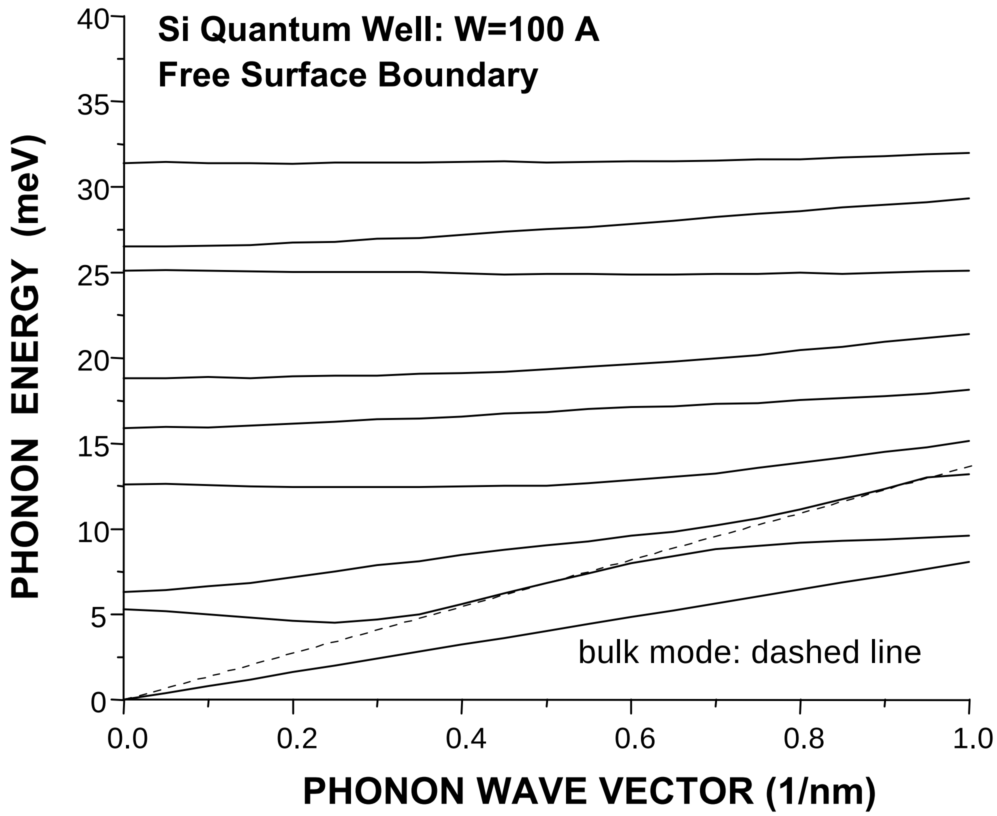




JL210 10-period Ge 1.5 nm/Si 20 nm

Phonon Confinement Effect in Quantum Well

acoustic phonon spectrum modification in quantum wells



Phonon scattering on point defects is increased due to phonon velocity reduction:

$$\frac{1}{\tau} = \frac{V\omega^4}{4\pi v_G^3}\Gamma,$$

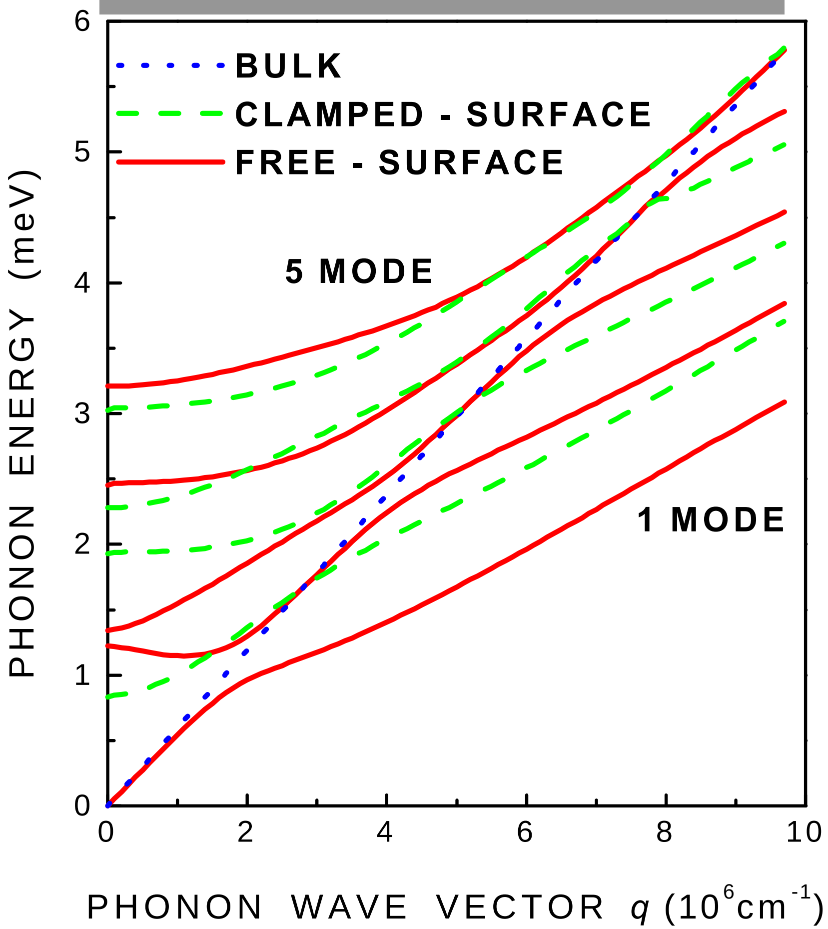
$$\Gamma = \sum f_i[1-(M_i/M)]^2$$

decreased slope = smaller group velocity

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

Phonon Confinement Effect in Quantum Wires

DISPERSION RELATIONS



Several branches of the lowest order longitudinal modes in Si wires of diameter 200 A are depicted.

- *Elastic continuum approximation*

$$L = \frac{1}{2}\int\left[\rho u'^2 - \lambda \xi_{i,j}^2 - 2\mu \xi_{i,j}^2\right]dr,$$

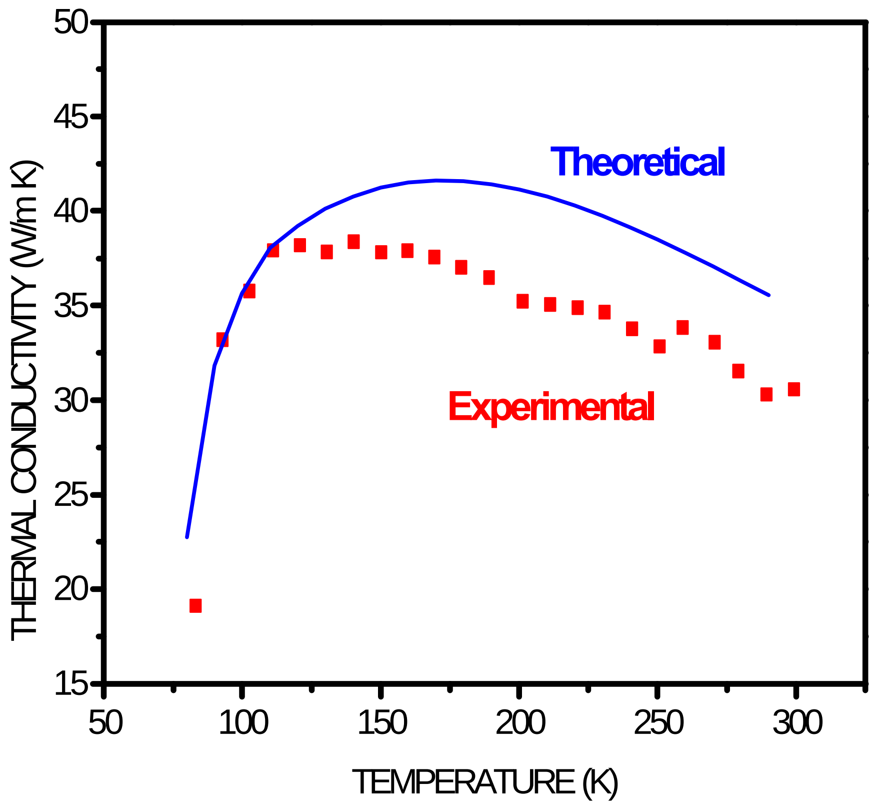
$$\frac{\partial^2 u}{\partial t^2} = s_t^2 \nabla^2 u + \left(s_l^2 - s_t^2\right) grad\ div\ u,$$

where L - Lagrangian, u - displacement, $\xi_{i,j}$ – strain tensor, ρ - density, λ, μ Lame constants

Free-standing or Clamped surface wire
ANY mode
group velocity ≤ sound velocity

Average phonon group velocity decrease

In-plane Heat Transport of Ge Quantum Dot Superlattices



$$k_l = \frac{k_B}{2\pi^2 v_g}\left(\frac{k_B}{\hbar}\right)^3 T^3 \int_0^{\frac{\theta}{T}} \frac{\tau_c x^4 e^x}{(e^x-1)^2}dx,$$

where $x=\hbar\omega/k_B T$, θ - Debye temperature ,

v_g - phonon group velocity,

τ_C - combined relaxation time:

$$\frac{1}{\tau_C} = \sum_i \frac{1}{\tau_i} = \frac{1}{\tau_U} + \frac{1}{\tau_B} + \frac{1}{\tau_M} + \frac{1}{\tau_D}$$

$$\frac{1}{\tau_D} = \frac{v_g \sigma_V}{V}$$

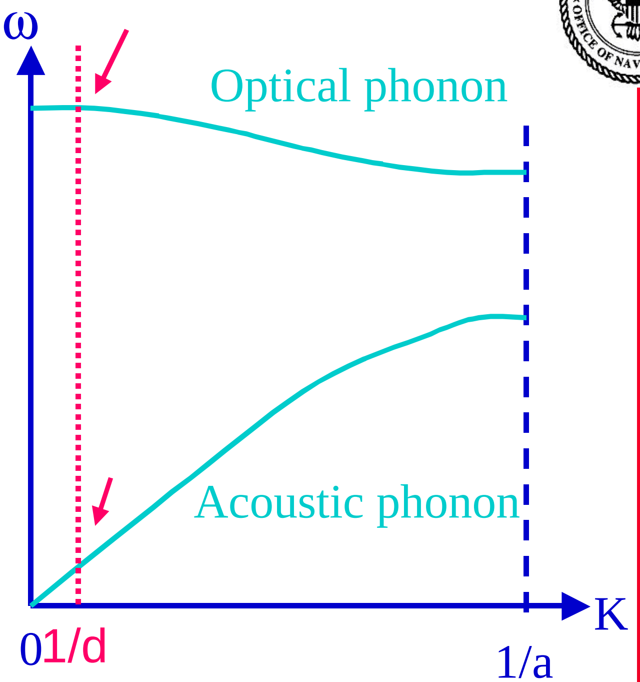
where σ_V - total cross section of the dot ensemble in volume V

Si/Ge Quantum Dot Superlattice (JL183 sample), dot height 7.0 nm, 75 nm in base, dot concentration 4×10^9 dots/cm^2.

Summary for Part I

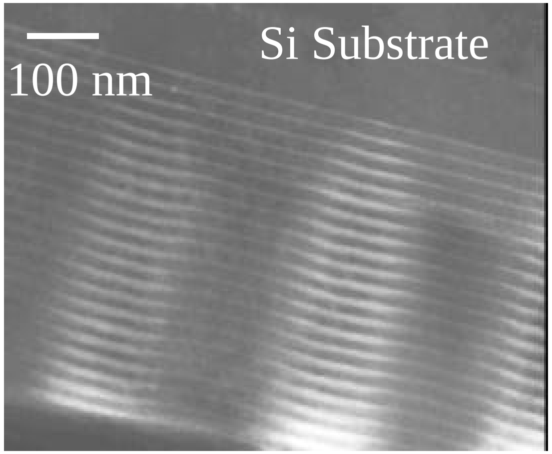
- SiGe samples
  - ~150 samples
- Graduate Students Graduated based on SiGe samples
  - Xiangzhong Sun, Taka Koga, Theo Boca, Jeffery Yao, Weili Liu, Bao Yang, Jianlin Liu
- Theory developed
  - Phonon confinement effect (quantum wells, wires)
  - Phonon scattering by dots
- Publications
  - Book chapters: 1
  - Journals: 19
  - Conference proceedings: 10
- Patent
  - Sb-mediated SiGe graded buffer layer technique

Part II: Fifth-year Research

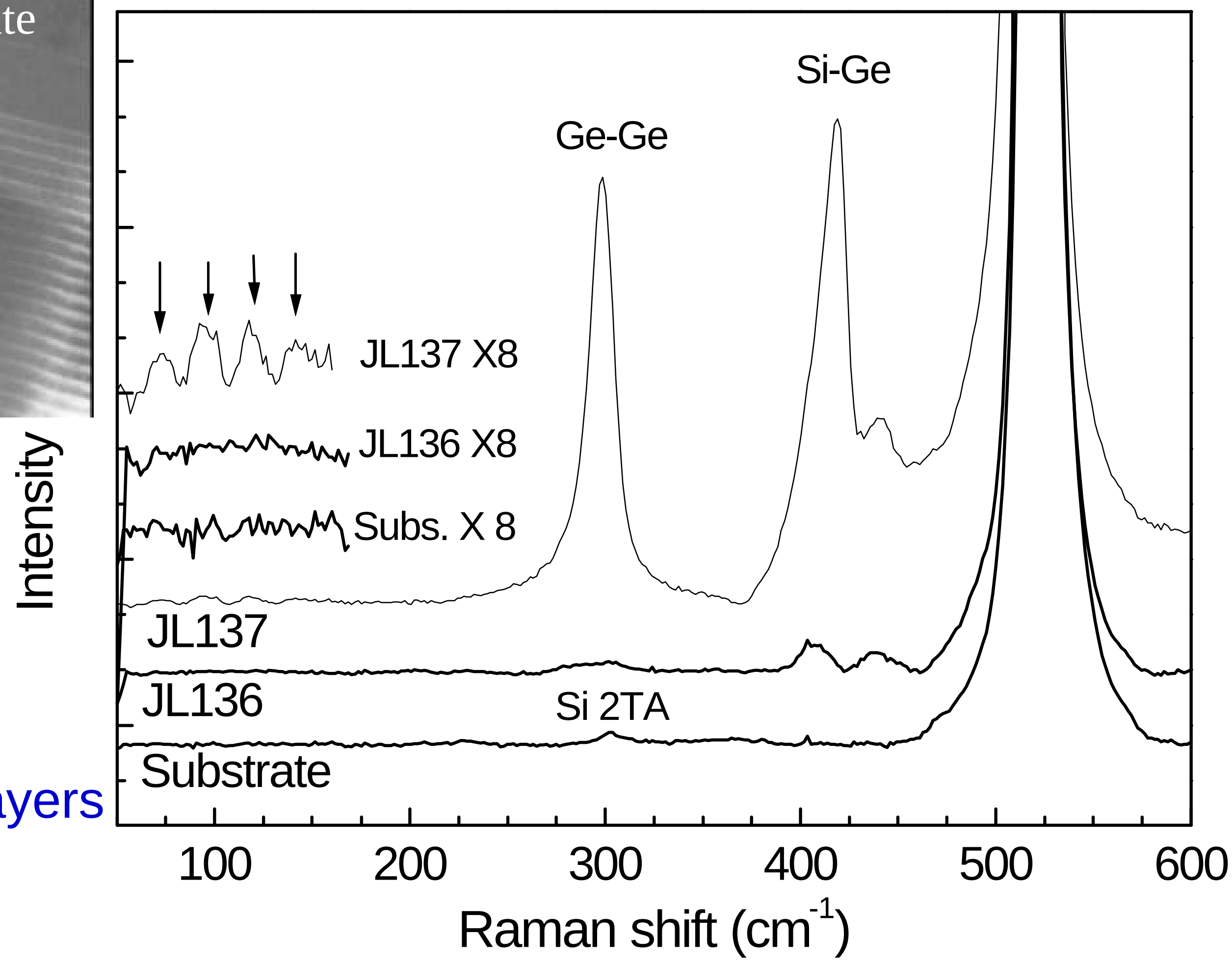


- Material growth service samples:
  - Si/Ge superlattices on LT-Si
- Wang's group:
  - Optical phonons in Ge QD SLs: Strain relaxation effects
    - PRB revised version submitted
  - Acoustic phonons in Ge QD SLs: multiple scattering effects
    - PRL revised version submitted
  - Thermal conductivity of Ge QD SLs measurements/modeling
    - PRB in preparation

Raman Scattering to Probe Acoustic and Optical Phonons



JL137

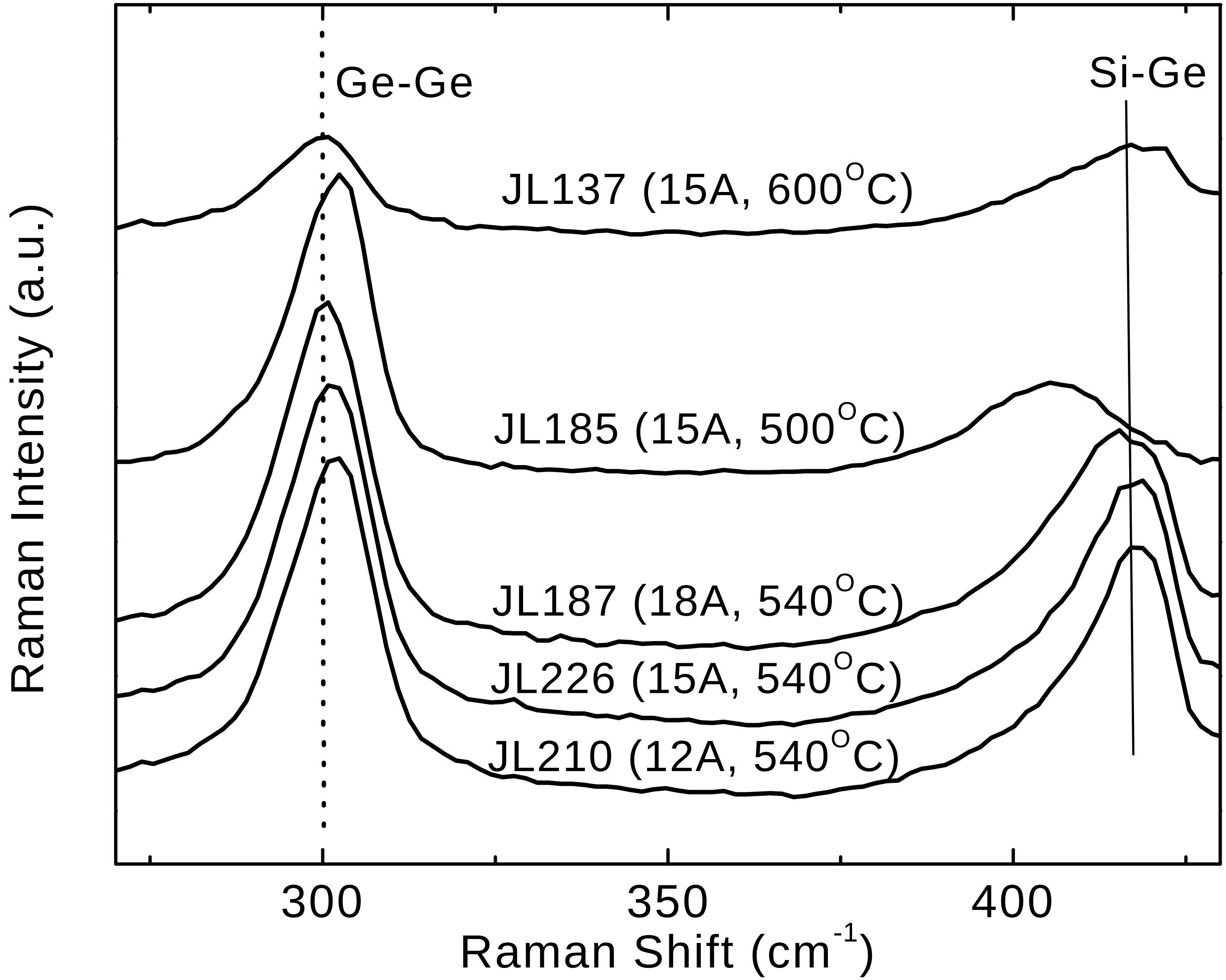


- JL137: dots
- JL136:Wetting layers
- Si substrate

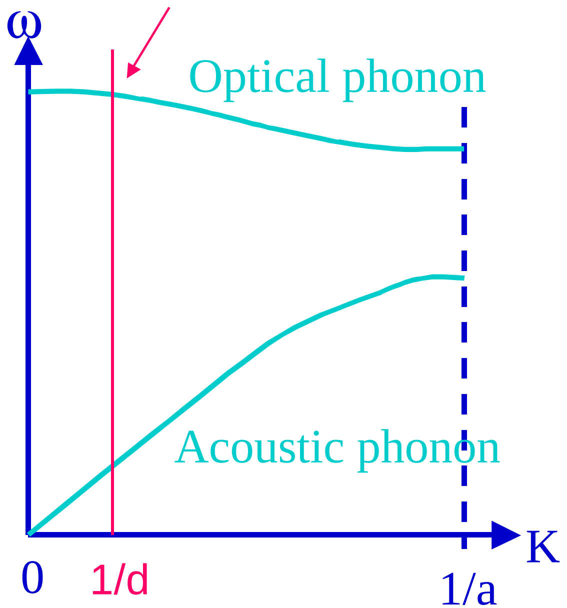
Quantum Dot Superlattices

Sample	Growth T (°C)	Period	Ge layer Thickness (Å)	Si layer Thickness (nm)	Dot base (nm)	Dot height (nm)	Density (cm^{-2})
JL185	500	10	15	20	114.7	15.1	5.9×10^8
JL210	540	10	12	20	110.4	11.9	3.6×10^9
JL226	540	10	15	20	122.0	14.0	4.1×10^9
JL187	540	10	18	20	122.2	16.0	3.5×10^9
JL137	600	22	15	20	175.5	10.2	2.6×10^8
JL138	600	22	12	20	152.4	10.0	1.4×10^8
JL136	600	25	6	20	-----	----	------

Optical Phonons



Optical Phonon Analysis



$$\omega = \omega_0 + \frac{1}{2\omega_0}\left[p\varepsilon_{zz} + q\left(\varepsilon_{xx} + \varepsilon_{yy}\right)\right]$$

p, q: deformation potentials

ω_0: Bulk Ge phonon position

ε_{xx}=0.042, ω=317.4 cm^{-1}

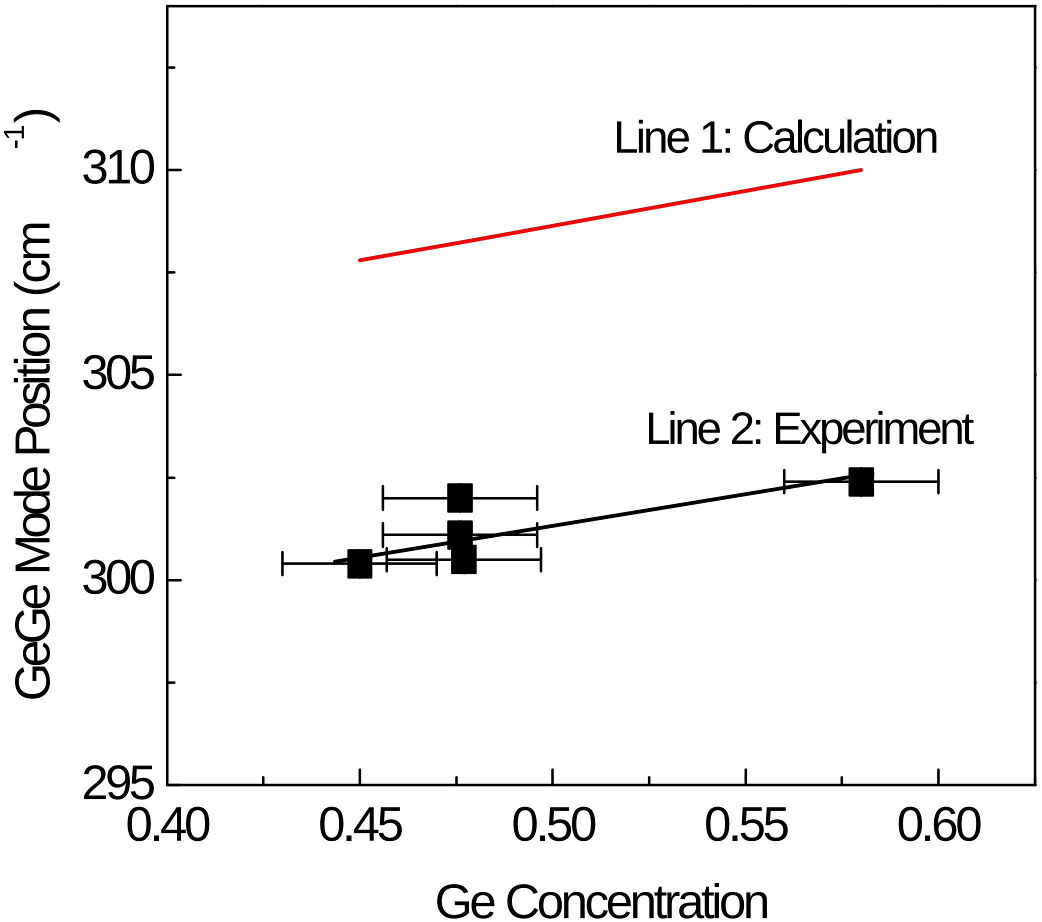
Conclusion: dot relaxed

Sample	ω (Cm^{-1})	ε_{xx} (10^{-3})
JL210	302	-4.8
JL226	301.1	-2.65
JL187	300.5	-1.2
JL185	302.4	-5.8
JL137	300.4	-0.96

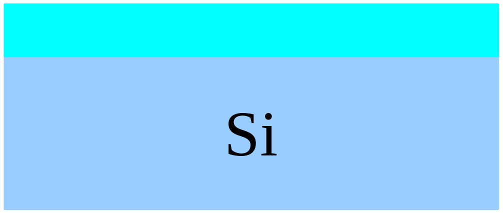
Strain Relaxation by Si/Ge Interdiffusion

$$\frac{I_{GeGe}}{I_{SiGe}} \cong B\frac{x}{2(1-x)}$$

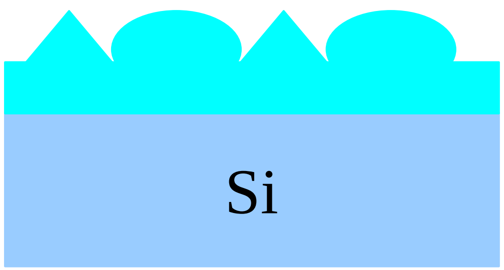
Sample	$\frac{I_{GeGe}}{I_{SiGe}}$	Ge composition in dots
JL210	1.45	0.48
JL226	1.45	0.48
JL187	1.45	0.48
JL185	2.14	0.58
JL137	1.29	0.45



Relaxation From Dot Morphology Transition



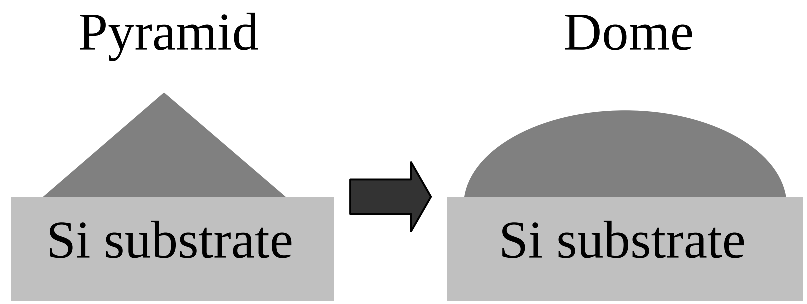
Layer-by-layer
T_{Ge}<6Å



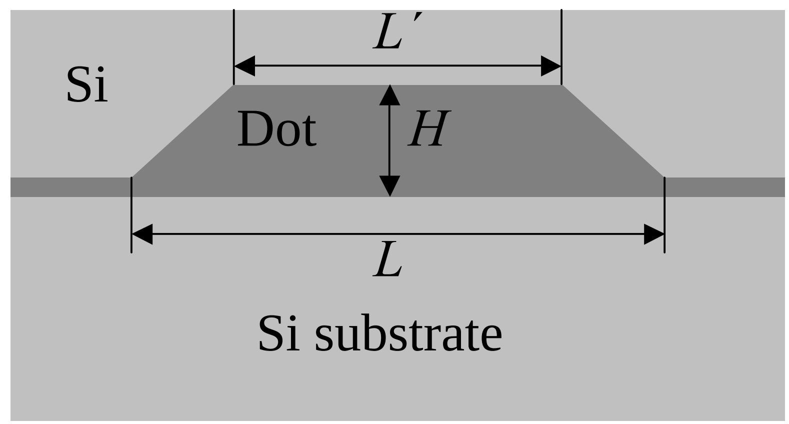
Pyramid/dome dots
6Å<T_{Ge}<20Å

(Stranski-Krastanov mode)

Surface dot relaxation:

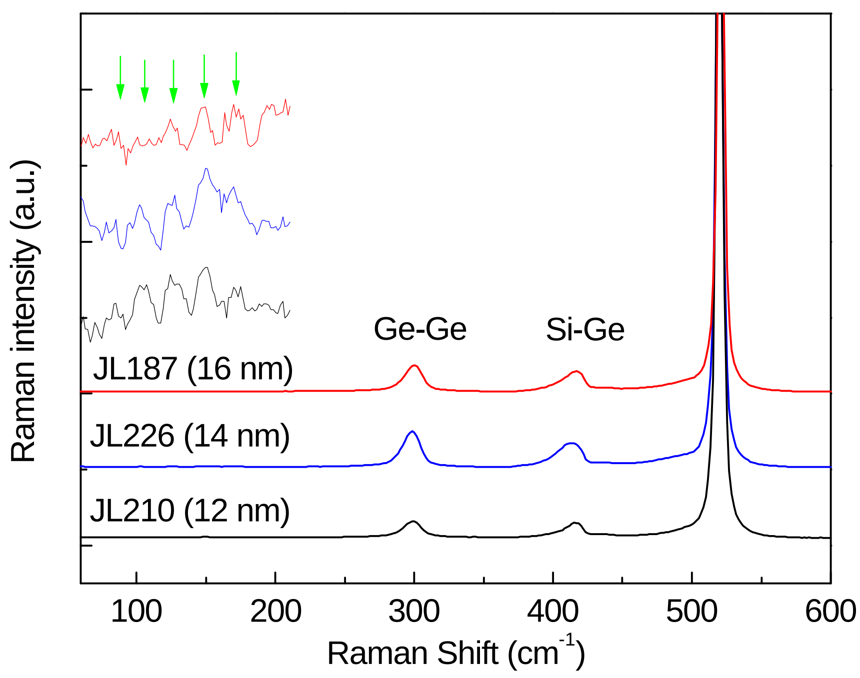


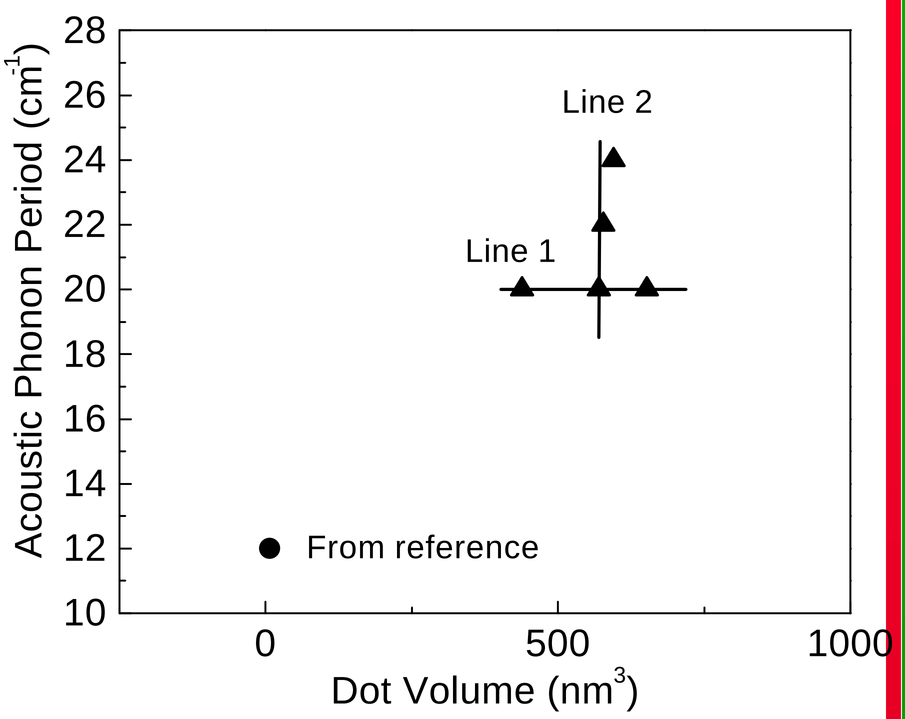
Embedded dot relaxation:



$$\Delta\varepsilon_{xx} = \frac{2C_{11}}{C_{12}} \cdot \frac{H}{L-L'} \ln\left[\left(1+\frac{L-L'}{L'}\right)^2 - 1\right]$$

Dependence of Acoustic Phonons On Dot Sizes

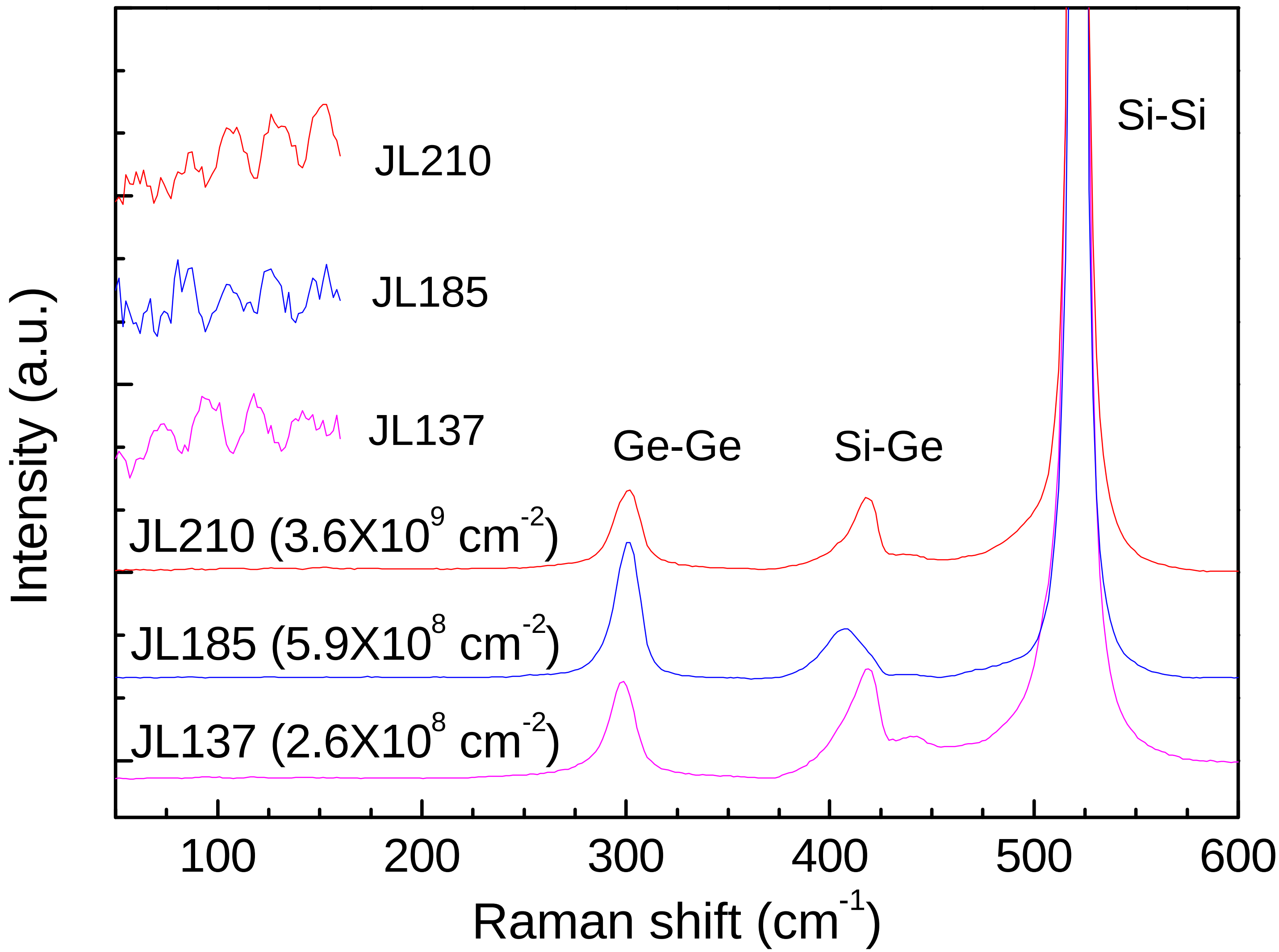




- Acoustic phonon peak positions Independent on dot size/volume
- Conclusion: Not confined or folded
- What are they

• A. Milekhin, et al. Eur. Phys. J. B 16, 355(2000)

Dependence of Acoustic Phonons on Dot Densities



Multiple Scattering Effects (Calculation/Experiment Comparison)

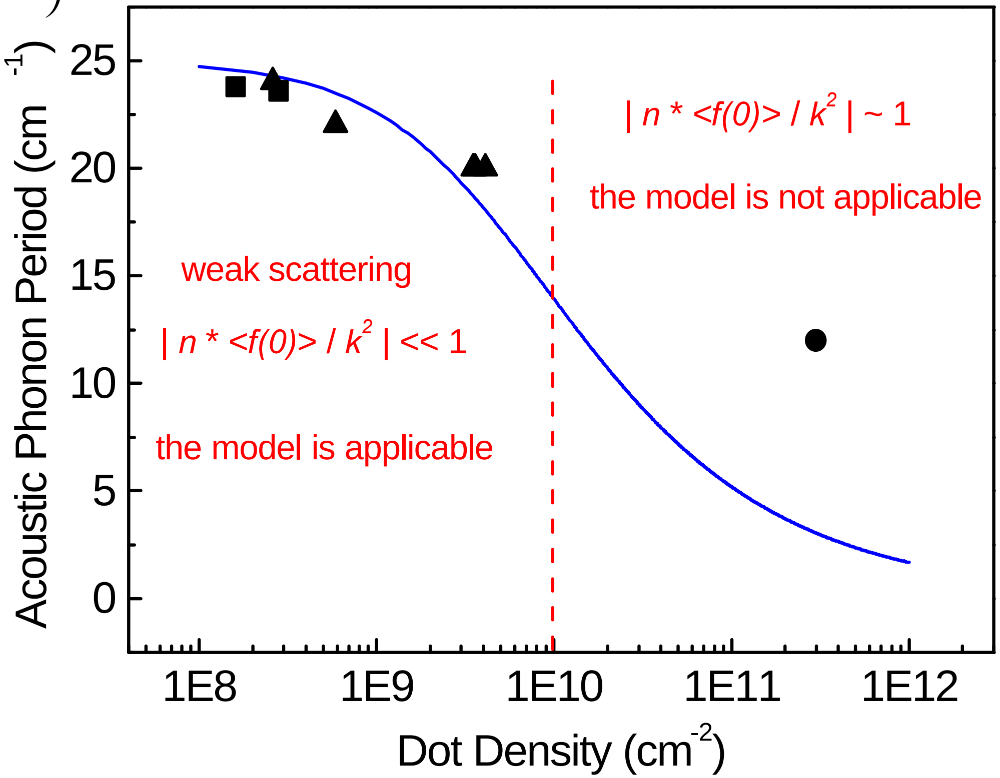
$$k'^2 = k^2\left(\left[1+\frac{2\pi n\langle f(0)\rangle}{k^2}\right]^2 - \left[\frac{2\pi n\langle f(\pi)\rangle}{k^2}\right]^2\right)$$

$$\langle f(\theta)\rangle = \int d\alpha\, p(\alpha)\, f(\theta;\alpha)$$

$$|n\langle f(\theta)\rangle / k^2| \ll 1$$

$$k'^2 \approx k^2 + 4\pi n\langle f(0)\rangle$$

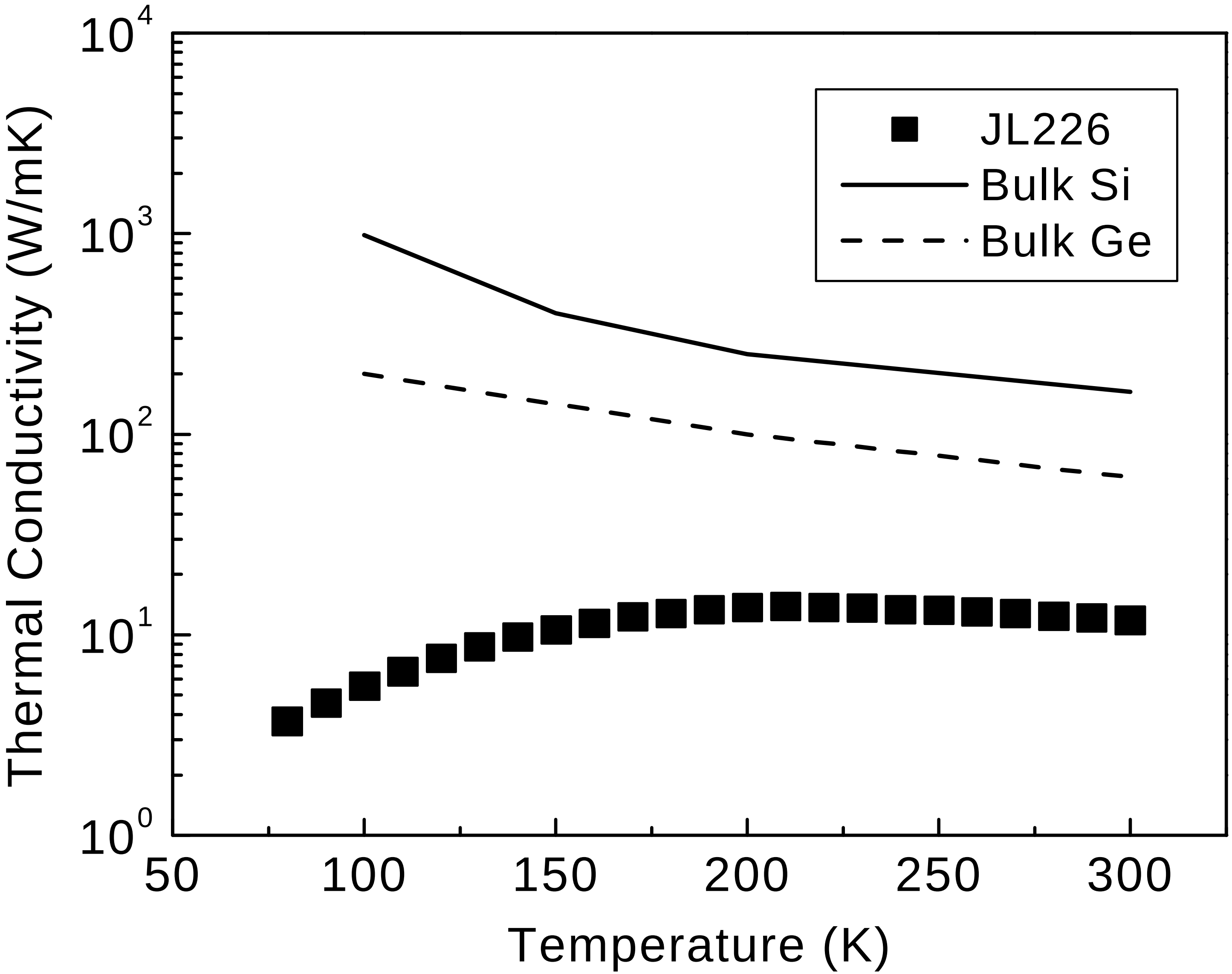
Phonon peak period dependence on quantum dot density



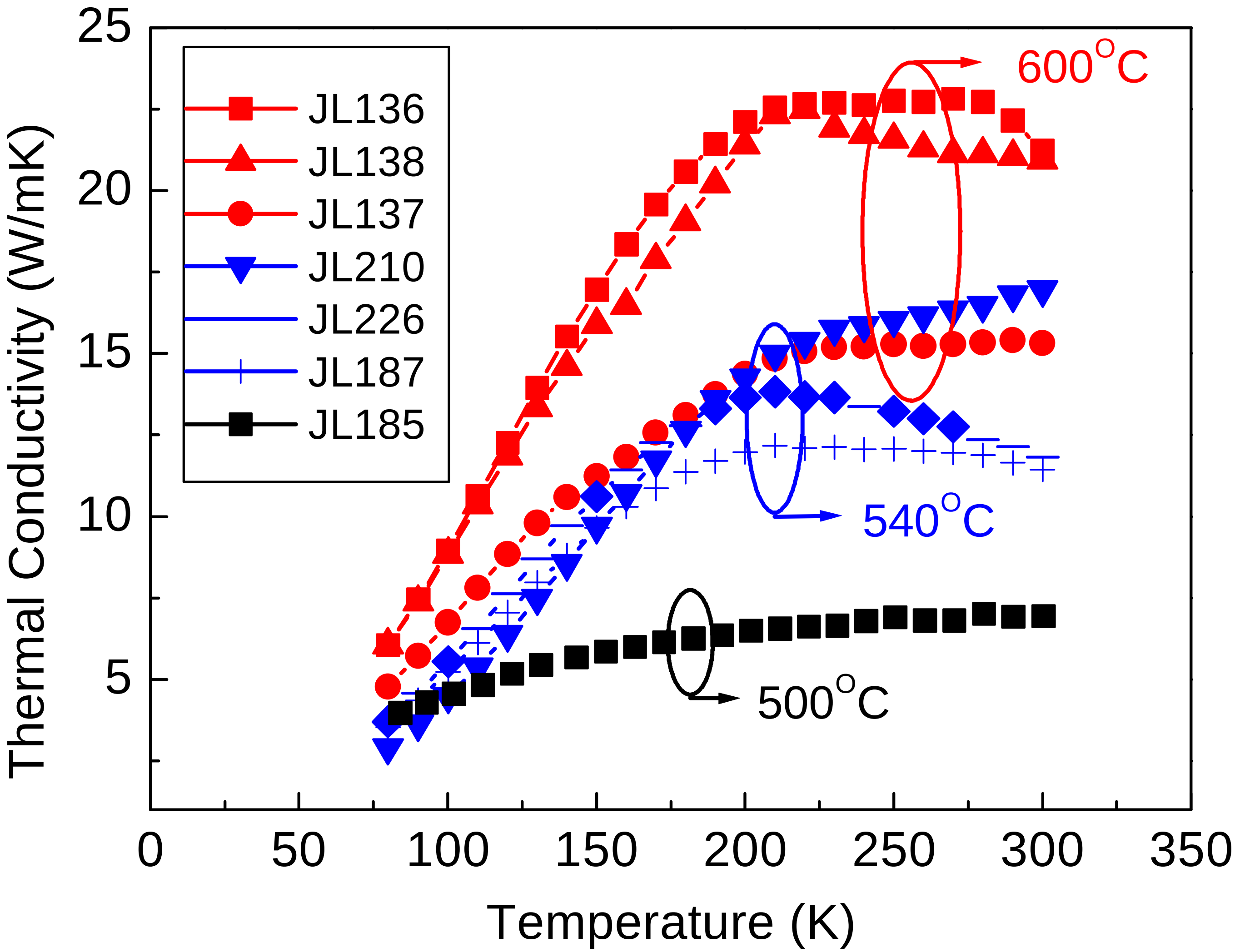
k' - modified phonon vector; *k* - phonon vector in the absence of quantum dots;

n – number of dots per unit volume; *f(0)*, *f(π)* - forward and backward scattering respectively

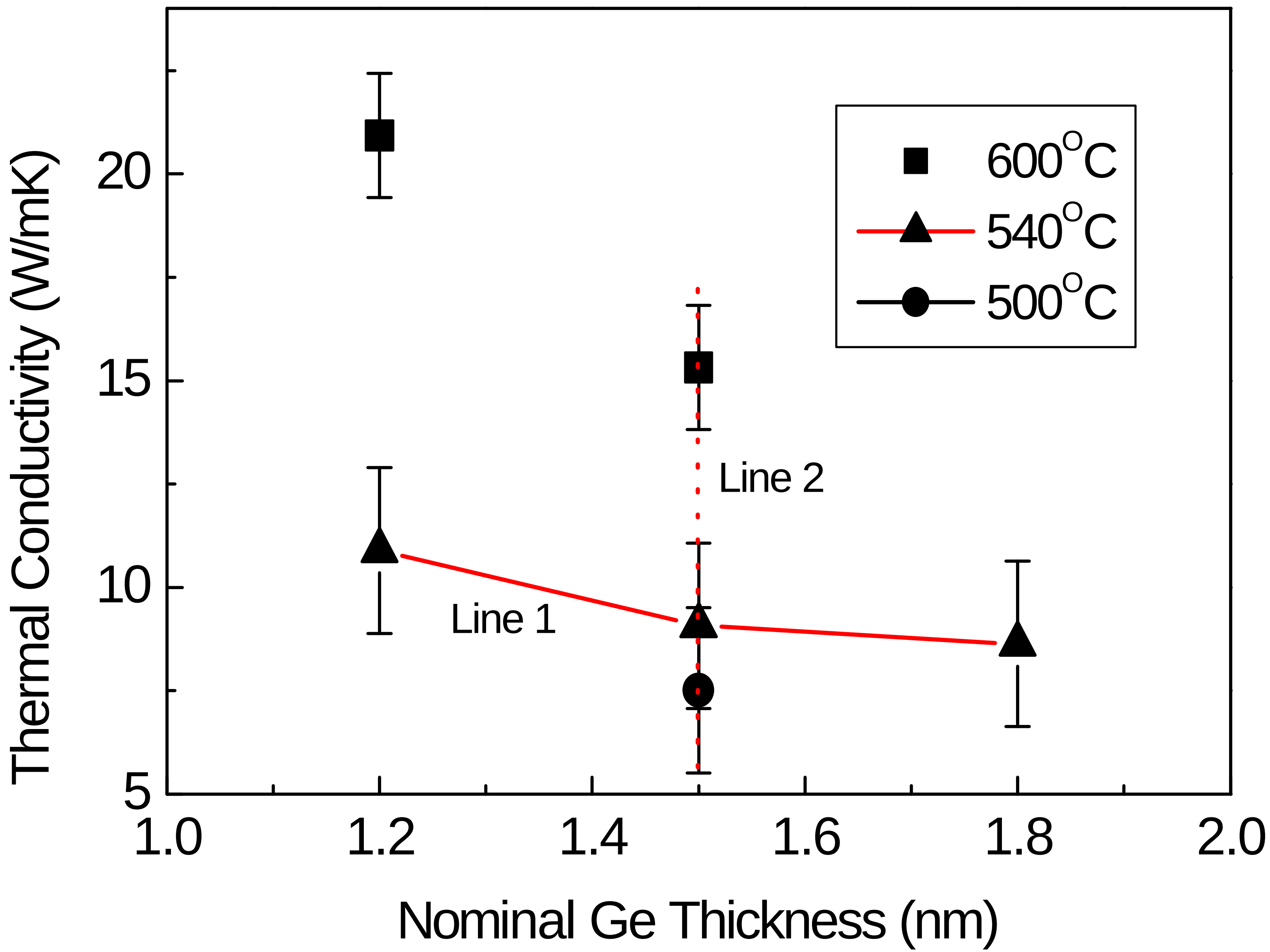
Thermal Conductivity Comparison



Cross-plane Thermal Conductivity



Thermal Conductivity Dependences



Cross-plane Thermal Conductivity Modeling

$$\kappa = \frac{1}{3}\sum_i \int dk v_{g_i}^2(k)\tau_{C_i}(k)S_i(k)$$

$$\frac{1}{\tau_C} = \sum \frac{1}{\tau} = \frac{1}{\tau_M} + \frac{1}{\tau_B} + \frac{1}{\tau_U} + \frac{1}{\tau_D}$$

$$\frac{1}{\tau_D} = \frac{v_g \sigma_V}{V}$$

QD scattering cross-section:

$$\sigma = \frac{\pi}{k^2}\sum_{m=0}^{\infty}(2m+1)\left|1+R_m\right|^2$$

Reflection coefficient:

$$R_m = \frac{h'^*_m(ka) + i\beta h^*_m(ka)}{h'_m(ka) + i\beta h_m(ka)}$$

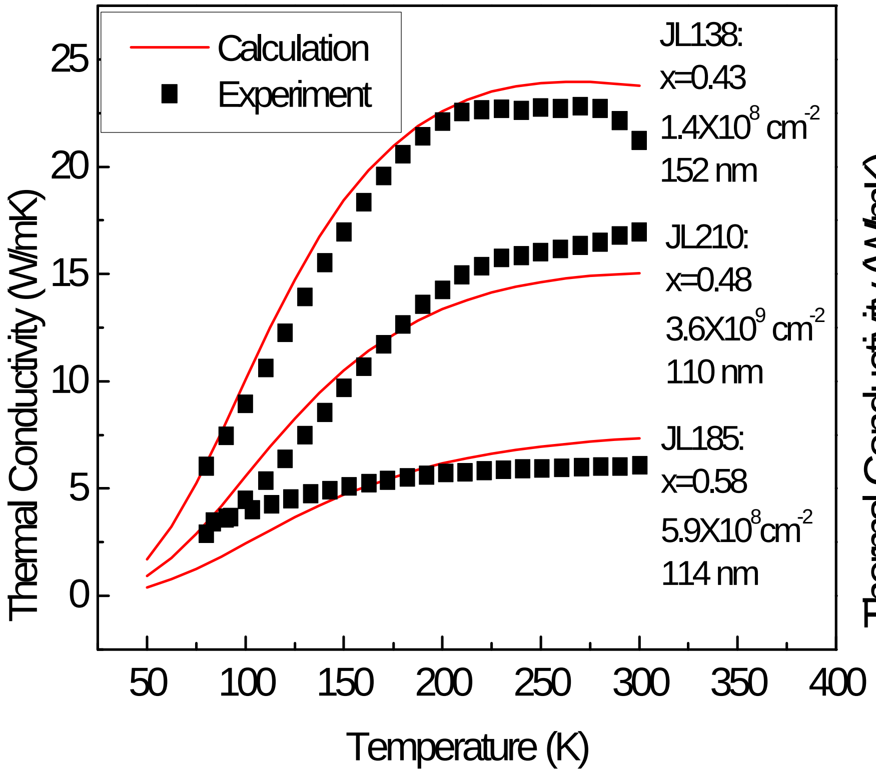
$$\beta_n = i\frac{\rho c}{\rho_e c_e}\left[\frac{j'_n(ka)}{j_m(k_e a)}\right]$$

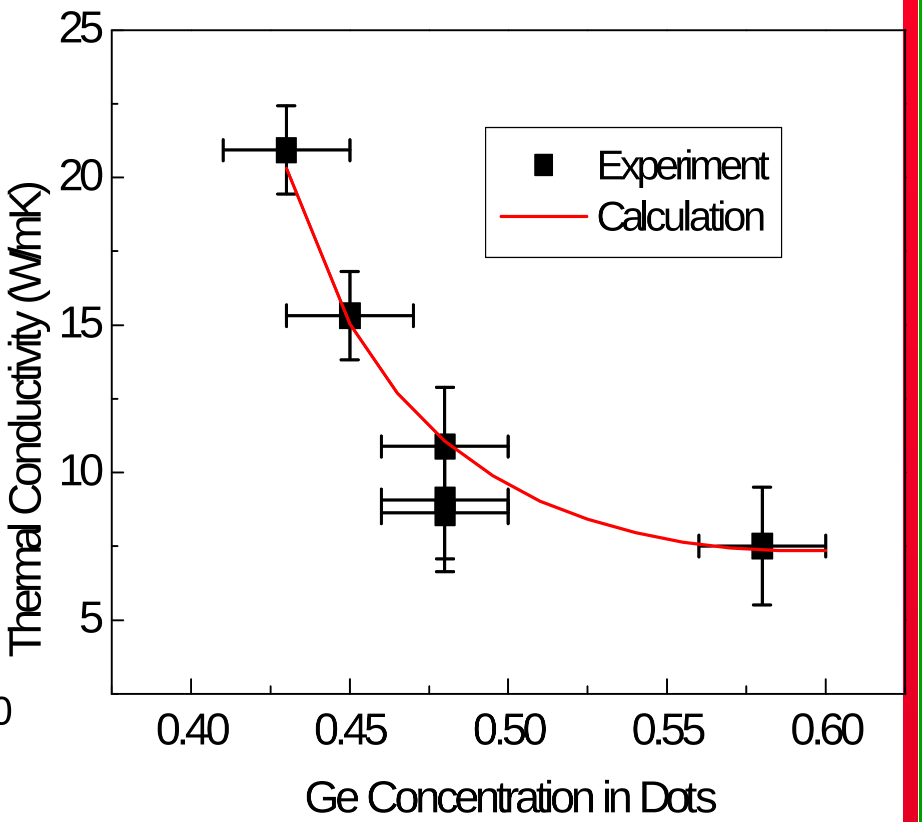
ρ-density, c-sound velocity, e-the parameter corresponding to dot material
j-Sphere Bessel functions

$$\rho_e = \rho_{Ge}\cdot x + (1-x)\rho_{Si}$$

$$c_e = c_{Ge}\cdot x + (1-x)c_{Si}$$

Experiment/Theory Comparison





- Dot Ge composition
- Dot density
- Dot base diameter

➢ Conclusion: Phonon scattering by SiGe quantum dots with different -sizes and densities

5th Year Accomplishments

- *MBE growth of service Si/Ge superlattices on LT-Si assisted SiGe buffers*
- *MBE growth of Ge quantum dots for phonon engineering*
  - **Dot size control by Ge thicknesses**
  - **Dot density control**
  - **Dot Ge composition control by growth temperature**
- *Phonon physics: Optical and acoustic phonons in Ge quantum dot superlattices*
  - **Raman scattering to probe acoustic and optical phonons**
  - **Optical phonon spectra: Strain relaxation effect**
  - **Acoustic phonons in superlattices with different dot sizes/densities: Multiple scattering effects**
- *Cross-plane thermal conductivity reduction in Ge quantum dot superlattices*
  - **Measurements by Gang's group**
  - **Cross-plane thermal conductivity modeling**

5^{th} Year Publications

- Journal Publications:

1. *"Modification of the three-phonon Umklapp process in a quantum wire"* A. Khitun and K.L.Wang Applied Physics Letters, vol.79, (no.6), AIP, 6 Aug. 2001. p.851-3.
2. *"Effect of the long-range order in a quantum dot array on the in-plane lattice thermal conductivity"* A. Khitun, A. Balandin, J.L. Liu, and K.L.Wang *Superlattices and Microstructures*, vol.30, (no.1), Academic Press,. p.1-8 (2001).
3. *"Acoustic phonons in self-assembled Ge quantum dot superlattices: multiple scattering effects",* J.L.Liu, A.Khitun, and K.L.Wang, Phys. Rev. Lett. (submitted)
4. *"Optical phonons in self-assembled Ge quantum dot superlattices: strain relaxation effects",* J.L.Liu, J.Wan, Z.M.Jiang, A.Khitun, and K.L.Wang, Phys.Rev.B (submitted)
5. *"Cross-plane thermal conductivity of the self-assembled Ge quantum dot superlattices',* J.L.Liu, A.Khitun, K.L. Wang, W.L.Liu, G. Chen, Phys. Rev B (in preparation)

- Conference Presentations:

1. *"In-plane thermal and electronic transport in quantum dot superlattice"* A. Khitun, J.L. Liu , K.L.Wang and G.Chen (to be published in 2001 *MRS proceedings*)

Acknowledgements

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- Contract monitors: Dr.John Pazik, etc.
- Program PI: Dr. Gang Chen
- Other co-Pis: Prof. Dresselhaus, etc.
- Post-docs, Students

Thank you!