Investigation of the effects of a Quantum Dot Crystal geometry on its Brillouin spectrum

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Acknowledgements

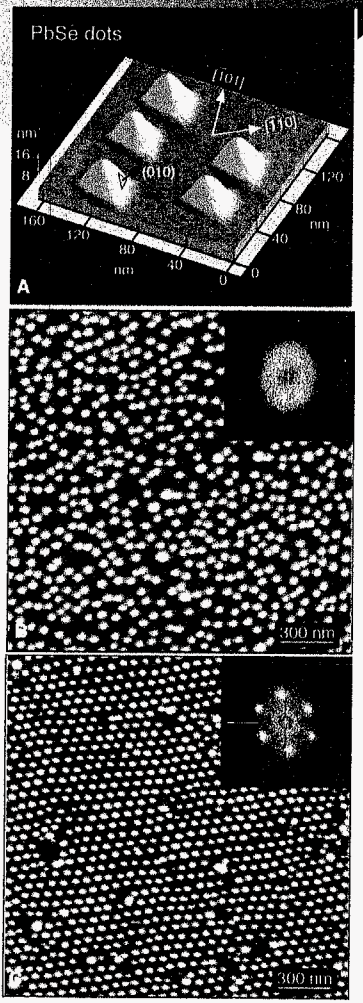
- Leonid P. Pryadko (Physics Department, UCR)
- Prof. Evgenii P. Pokatilov (Physics Department, Moldova State University)
- NSF Nanoscale Exploratory Research project ECS-0210282, NSF CAREER Award No. 0093959 to A.A.B. and AFOSR STTR Contract F49620.
- National Research Council Award at Jet Propulsion Laboratory (O.L.L.)

Motivation

- Fundamental Science
  - creation of artificial crystals with re-engineered properties
- Applications in Nanodevices
  - photonics
  - optoelectronics
  - utilization of nonlinear electrical transport properties
  - high-efficiency thermoelectric elements
- Most physical properties are determined by electron and phonon spectra

Artificial Quantum Dot Crystal

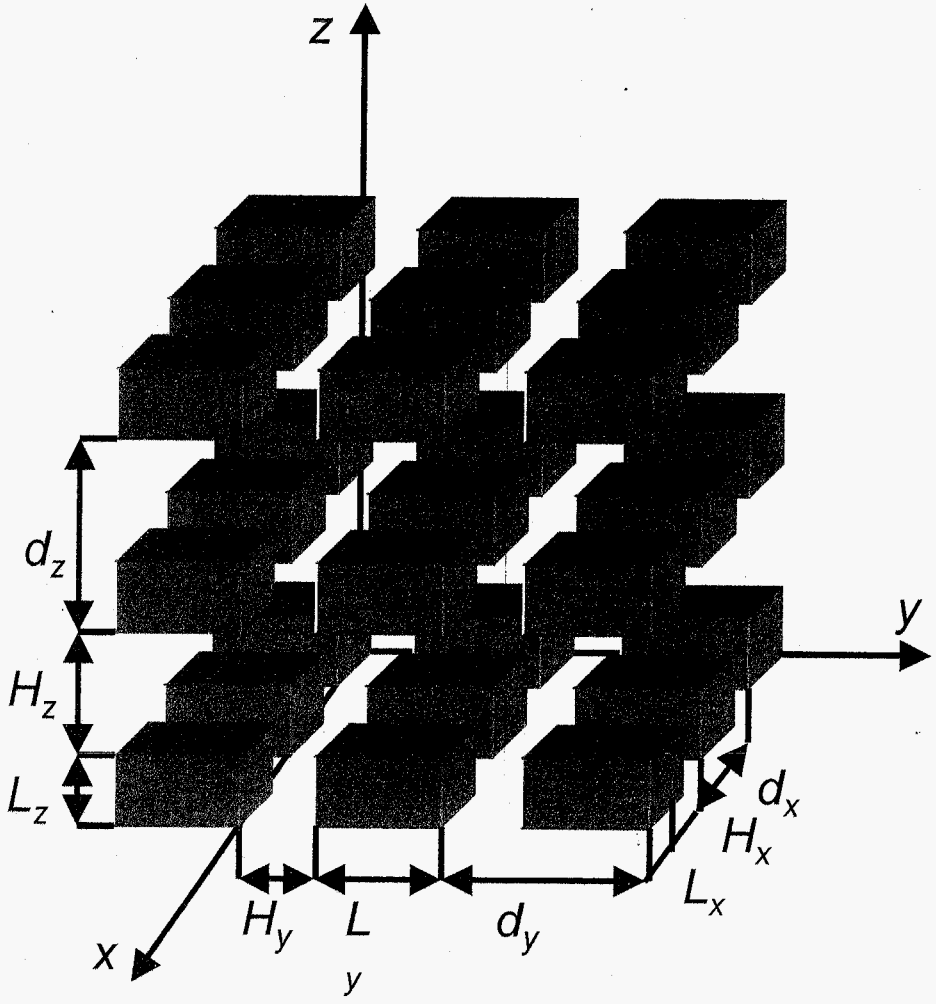
- If atoms of one material deposit onto the surface of another they may spontaneously form crystalline structures (quantum dots) with a high order of 3-dimensional regimentation



- (A) AFM image of pyramidal PbSe dots on PbTe (111) with {100} side facets.
- Large-scale AFM images of
  - (B) a PbSe single layer on $Pb_{1-x}Eu_xTe$
  - (C) the last PbSe layer of a 60-period PbSe/ $Pb_{1-x}Eu_xTe$ dot crystal. The insets show the 2D power spectra of the AFM images.

G. Springholtz, V. Holy, M. Pinczolits, G. Bauer, *Science* **282**, 734 (1998)

Quantum Dot Crystal (QDC) Structure



- *Three-dimensional (3D) ordered* array of interacting quantum dots forms the Quantum Dot Crystal (QDC)
- "…In the superlattice structure disorder and scattering is small enough to allow the *coherent* superlattice band states to built up…"

  C. Weisbuch, B. Vinter
  Quantum Semiconductor Structures

Applicability of the Continuum Approximation

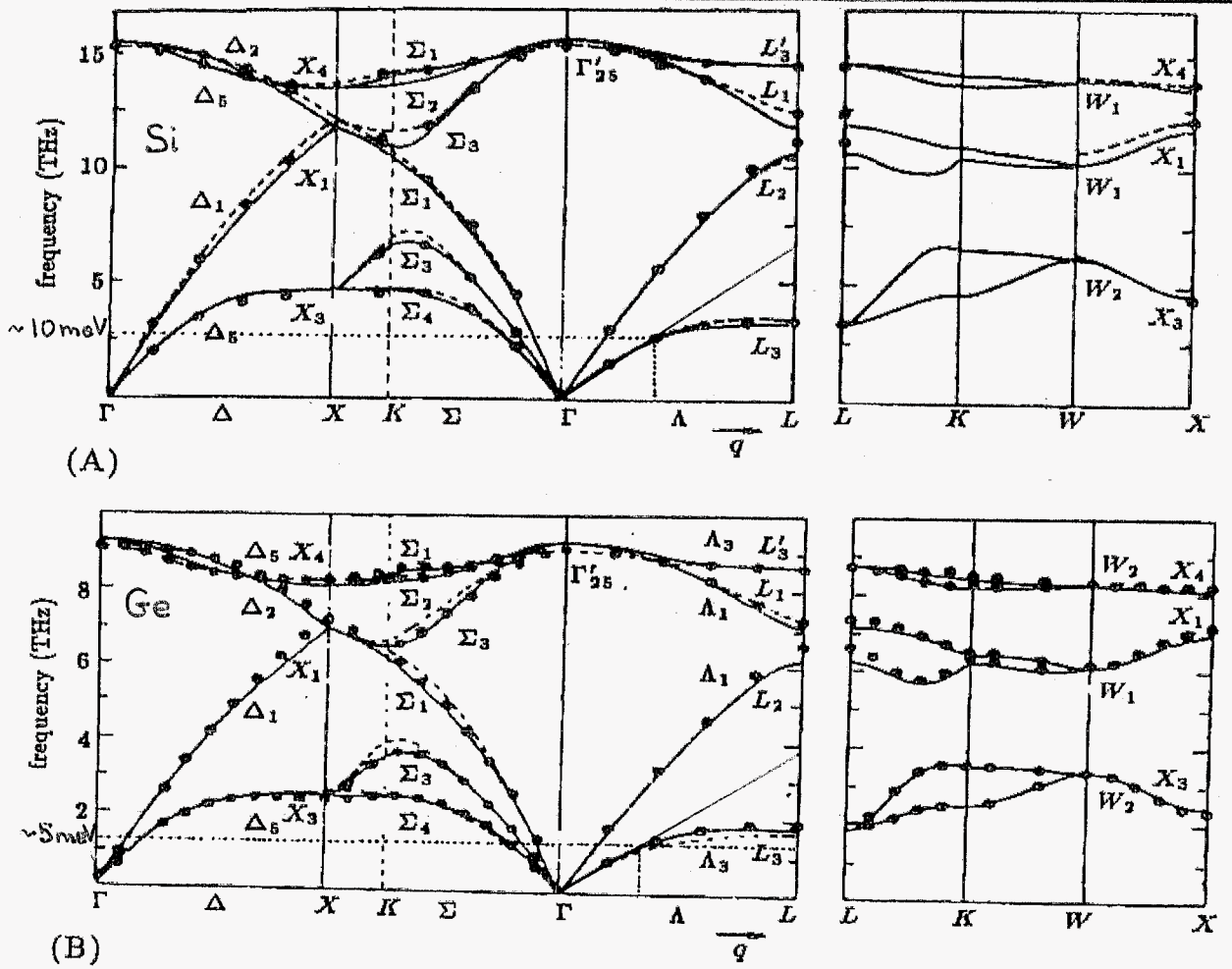


Figure 5.9: Phonon dispersion curves for: (A) Si, and (B) Ge. Solid curves: calculated by Weber (1977); filled circles: experimental values.

- Phonons can be formally treated in the continuum long-wave approximation as long as acoustic phonon dispersion in the bulk material remains linear.
- It corresponds to 2-3 lattice constants for Ge and Si.

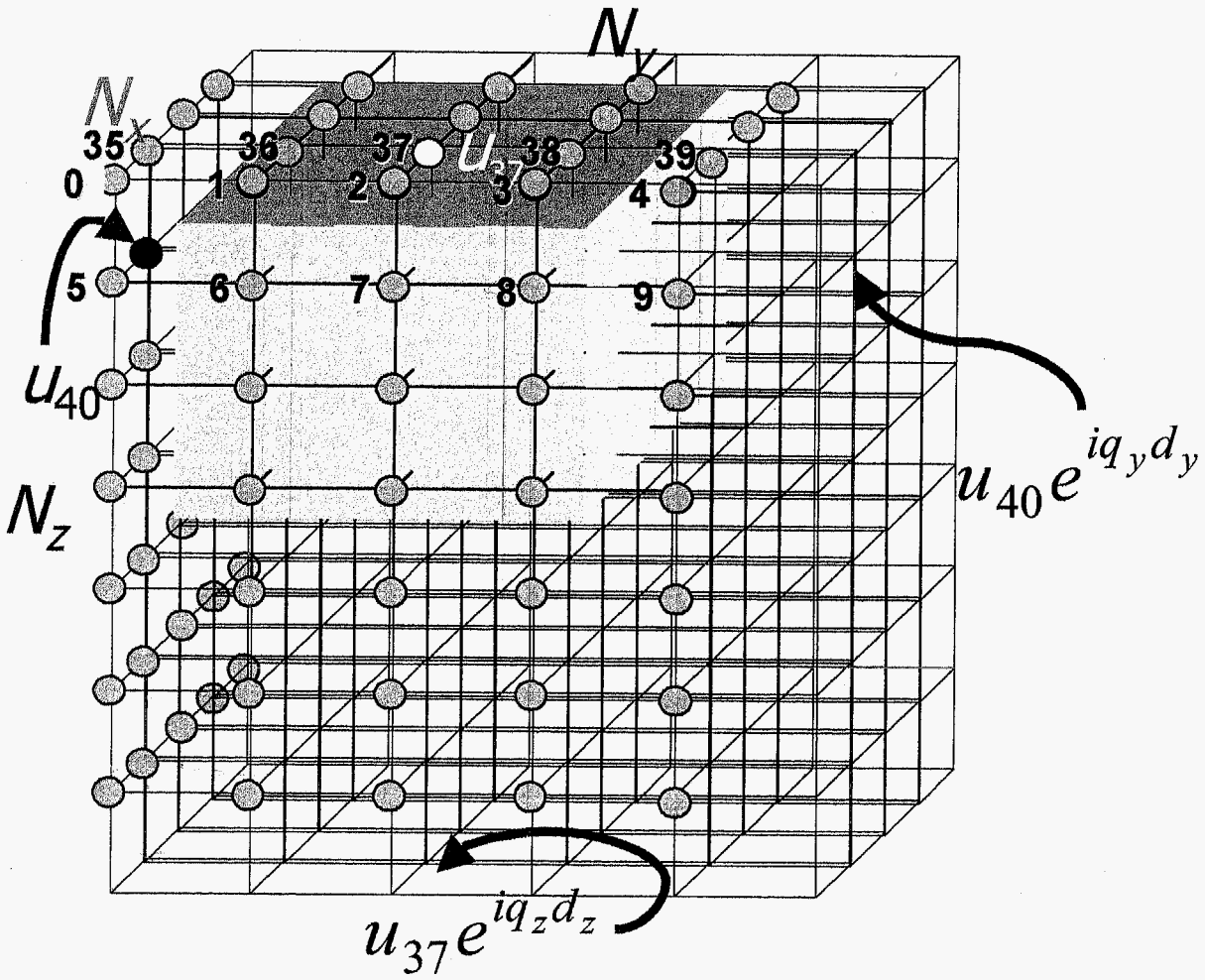
Elasticity Equation for Heterogeneous Cubic Crystal

- For x-component

$$\rho \frac{\partial^2 u_x}{\partial t^2} = \frac{\partial}{\partial x} c_{11} \frac{\partial u_x}{\partial x} + \frac{\partial}{\partial y} c_{44} \frac{\partial u_x}{\partial y} + \frac{\partial}{\partial z} c_{44} \frac{\partial u_x}{\partial z}$$
$$+ \frac{\partial}{\partial x} c_{12} \frac{\partial u_y}{\partial y} + \frac{\partial}{\partial x} c_{12} \frac{\partial u_z}{\partial z} + \frac{\partial}{\partial y} c_{44} \frac{\partial u_y}{\partial x} + \frac{\partial}{\partial z} c_{44} \frac{\partial u_z}{\partial x}$$

- For y-component replace $x \to y, y \to z, z \to x.$
- For z-component replace $x \to z, y \to x, z \to y.$
- In subscripts of elastic constants
  - $1 \equiv xx$ - strain along the displacement
  - $2 \equiv yy$ - strain perpendicular to the displacement
  - $4 \equiv yz$ - tangential strain

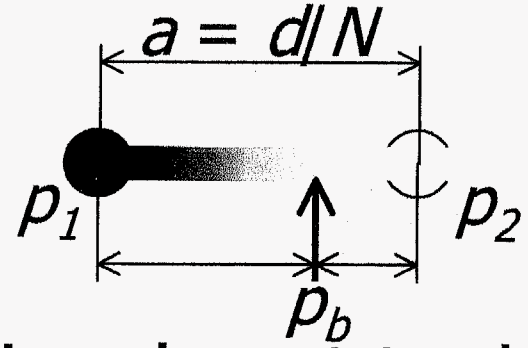
Grid and Boundary Conditions in QDC



- The coordinates of the closest to the origin QD vertex are (1/2,1/2,1/2)
- Quasi-periodic boundary conditions
- PDE for every node $\rightarrow 3(N_x N_y N_z)$ linear algebraic equations

Material Parameters

- Material parameters on QD's boundaries linearly change



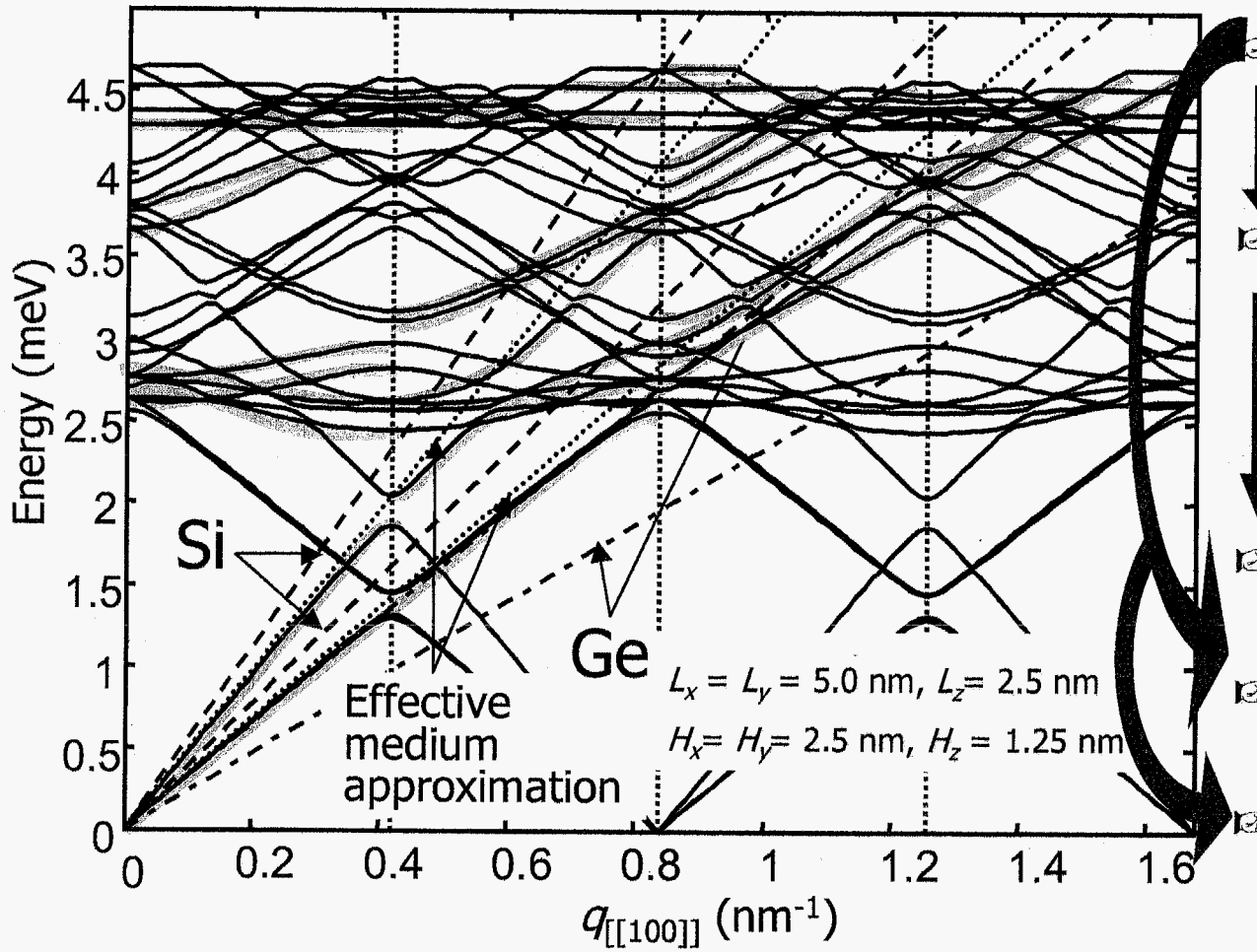
- On the boundaries closest to the origin:

$$p_b = \frac{1}{2}(p_1 + p_2)$$

- On the fare boundaries:

$$p_b = p_1\left[\frac{L}{a} - \left(n - \frac{1}{2}\right)\right] + p_2\left[\left(n + 1 - \frac{1}{2}\right) - \frac{L}{a}\right]$$

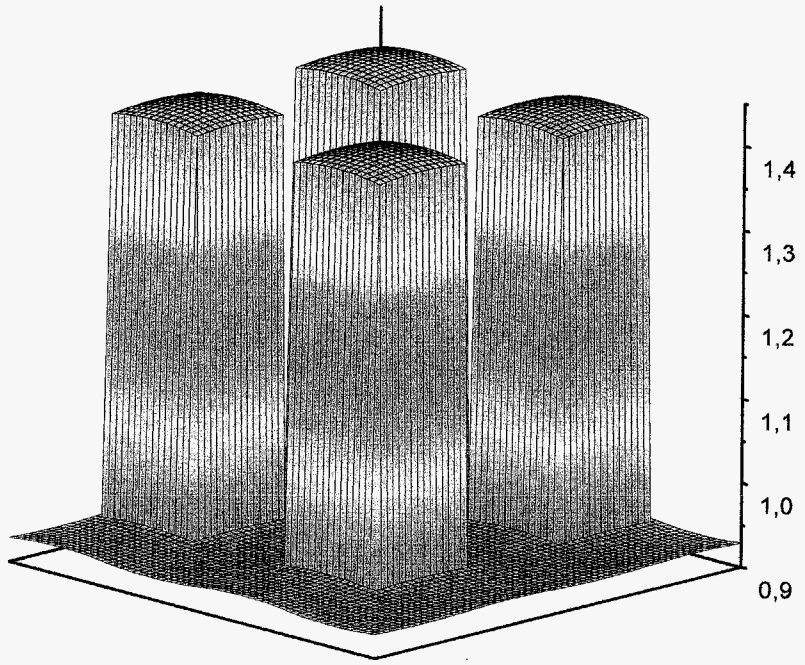
Phonon dispersion in tetragonal Ge/Si QDC



- Both quasi-acoustical and quasi-optical vibrations exist at low energy
- Quasi-optical waves oscillating inside or between quantum dots are almost localized
- Thermal conductivity decreases
- Phonon carrier scattering increases
- Phonon-assisted optical transitions are stimulated

Normalized to bulk intensity of phonon vibrations in QDC

- The lowest LA mode
- Ge/Si cubical QDC
  - L = 3 nm, D = 6 nm
  - c_{11}(Ge) = $12.40 \cdot 10^{10}$ N/m^2,
    c_{12}(Ge) = $4.13 \cdot 10^{10}$ N/m^2,
    c_{44}(Ge) = $6.83 \cdot 10^{10}$ N/m^2,
    c_{11}(Si) = $16.577 \cdot 10^{10}$ N/m^2,
    c_{12}(Si) = $6.393 \cdot 10^{10}$ N/m^2,
    c_{44}(Si) = $7.962 \cdot 10^{10}$ N/m^2;
    ρ(Ge) = $5.3234 \cdot 10^3$ kg/m^3,
    ρ(Si) = $2.3290 \cdot 10^3$ kg/m^3.
- The spectrum is normalized to the vibration intensity in $Ge_{0.113}Si_{0.887}$



Electromagnetic wave in QDC

- In effective medium approximation

$$\mathbf{D} = \hat{\varepsilon}\varepsilon_0 \mathbf{F}(\mathbf{r},\mathbf{k},\omega)\exp(-i\omega t)\ , \text{where}\quad \mathbf{F}(\mathbf{r},\mathbf{k},\omega) = \mathbf{A}\exp(i\mathbf{k}\cdot\mathbf{r})$$

- **A** is the light polarization vector: (**A**·**k**)=0,
- λ is the light wavelength in vacuum.

$$|\mathbf{k}| = \frac{2\pi\sqrt{\varepsilon}}{\lambda}$$

- Phonon vibrations cause local change of the dielectric susceptibility tensor ε
  - in cubic semiconductors

$$\varepsilon_{xx} = \varepsilon_{xx}^{\circ} + q_{1111}\frac{\partial U_x}{\partial x} + q_{1122}\frac{\partial U_y}{\partial y} + q_{1122}\frac{\partial U_z}{\partial z}$$

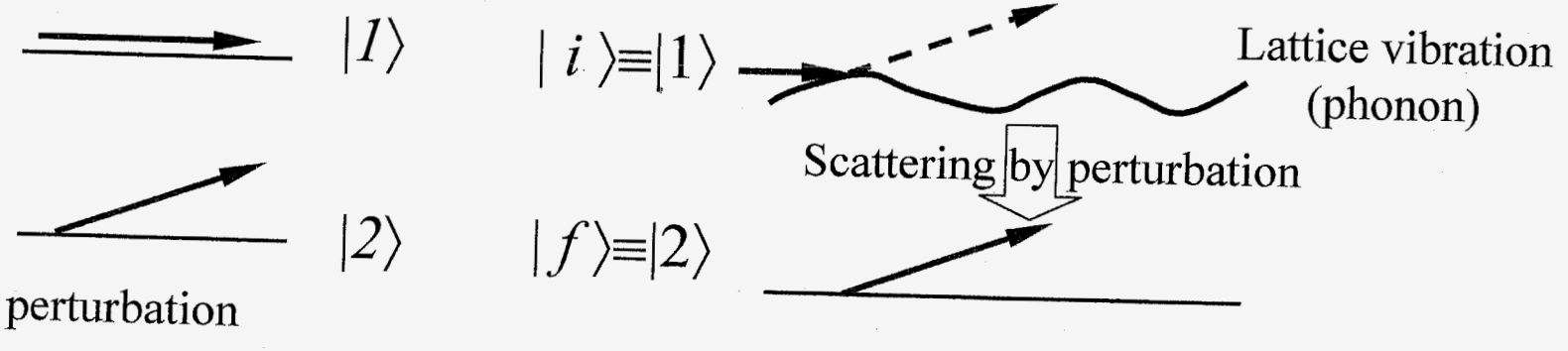
  - q_{ijkl} are components of the photoelastic tensor

Probability of anti-Stokes Raman scattering in QDC

$$P_{fi} \propto \left|\left\langle \mathbf{D}_f \middle| \mathbf{D}_i \right\rangle\right|^2 = \varepsilon^2 \varepsilon_0^4 \delta(\omega_f - \omega_i - \Omega)\delta(\mathbf{k}_f - \mathbf{k}_i - \mathbf{q})\left(\frac{V}{d_x d_y d_z}\right)^2 \left| \int_{EC} d\mathbf{r} \left\{ \exp(-i\mathbf{q}\cdot\mathbf{r}) \cdot \right.\right.$$

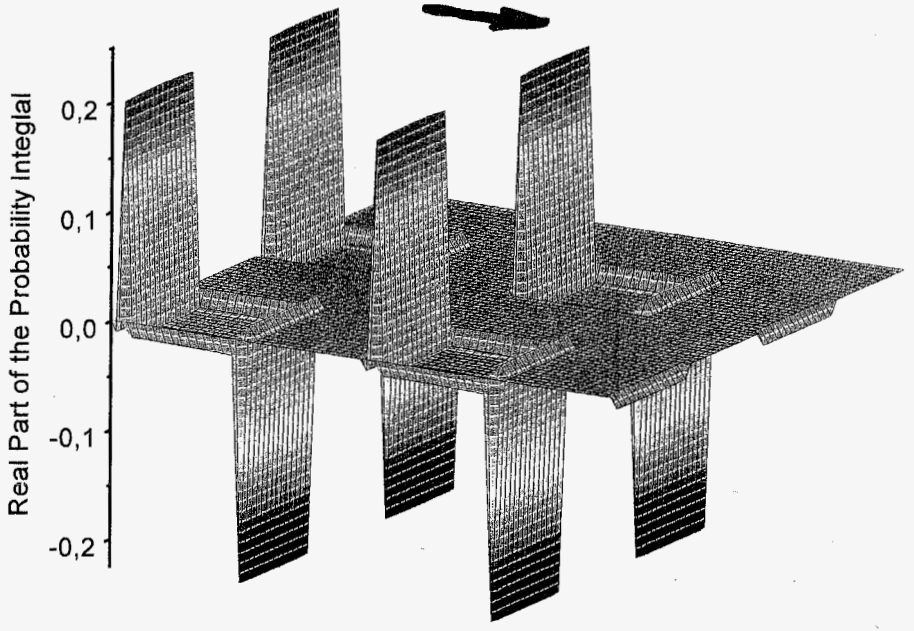
$$\left[\left(q_{1111} A_x^i A_x^f + q_{1122}\left(A_y^i A_y^f + A_z^i A_z^f\right)\right)\frac{\partial u_x}{\partial x} + \left(q_{1111} A_y^i A_y^f + q_{1122}\left(A_z^i A_z^f + A_x^i A_x^f\right)\right)\frac{\partial u_y}{\partial y} + \right.$$

$$\left.\left.\left.\left(q_{1111} A_z^i A_z^f + q_{1122}\left(A_x^i A_x^f + A_y^i A_y^f\right)\right)\frac{\partial u_z}{\partial z}\right]\right\}\right|^2$$

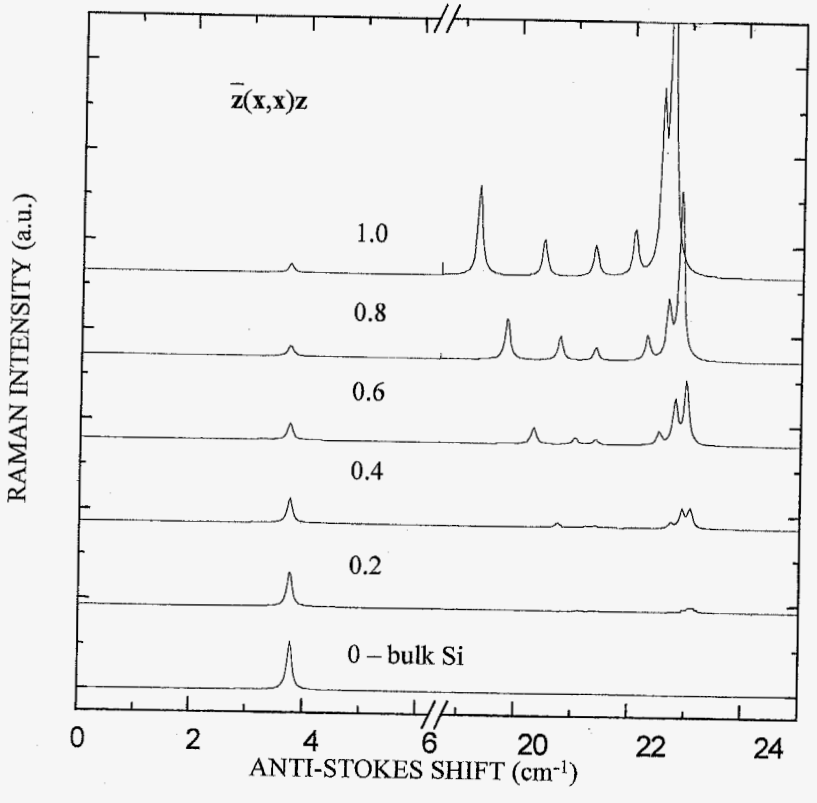


Spatial distribution of the light scattering in QDC

- The lowest LA mode
- Ge/Si cubical QDC
  - $L = 3$ nm, $D = 6$ nm
  - $q_{1111}(\mathrm{Ge}) = 40.0$,
    $q_{1122}(\mathrm{Ge}) = 35.0$,
    $q_{1111}(\mathrm{Si}) = 16.0$,
    $q_{1122}(\mathrm{Si}) = -2.0$;
    $\varepsilon^0(\mathrm{Si}) = 11.9$,
    $\varepsilon^0(\mathrm{Ge}) = 16.2$.

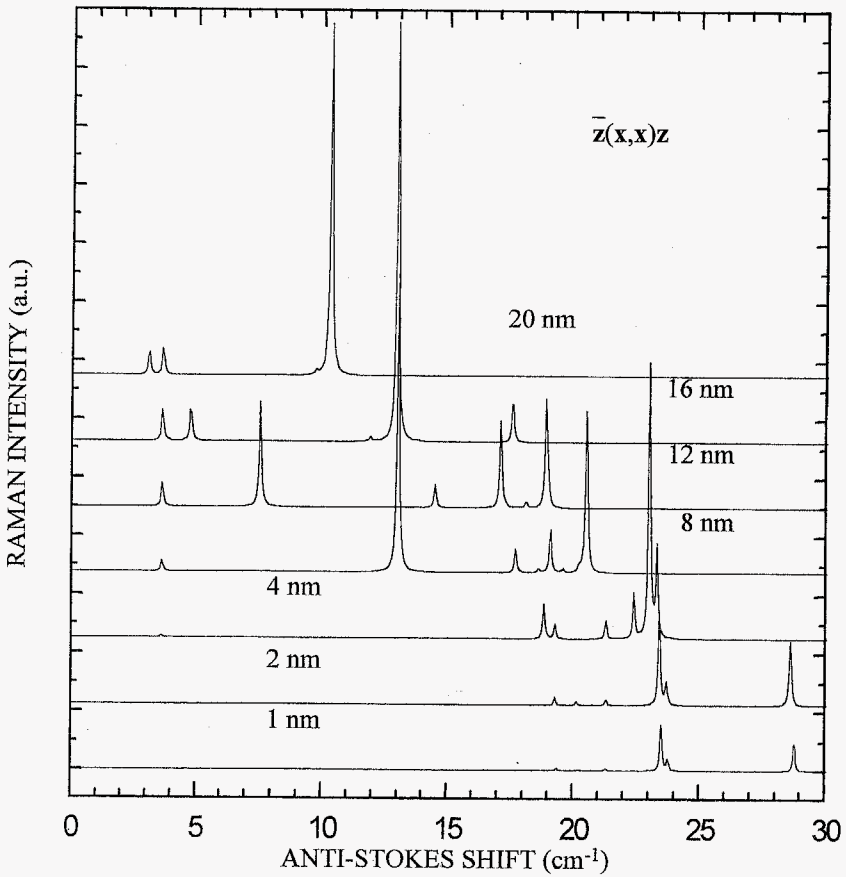


Raman spectra of Ge_xSi_{1-x}/Si QDC



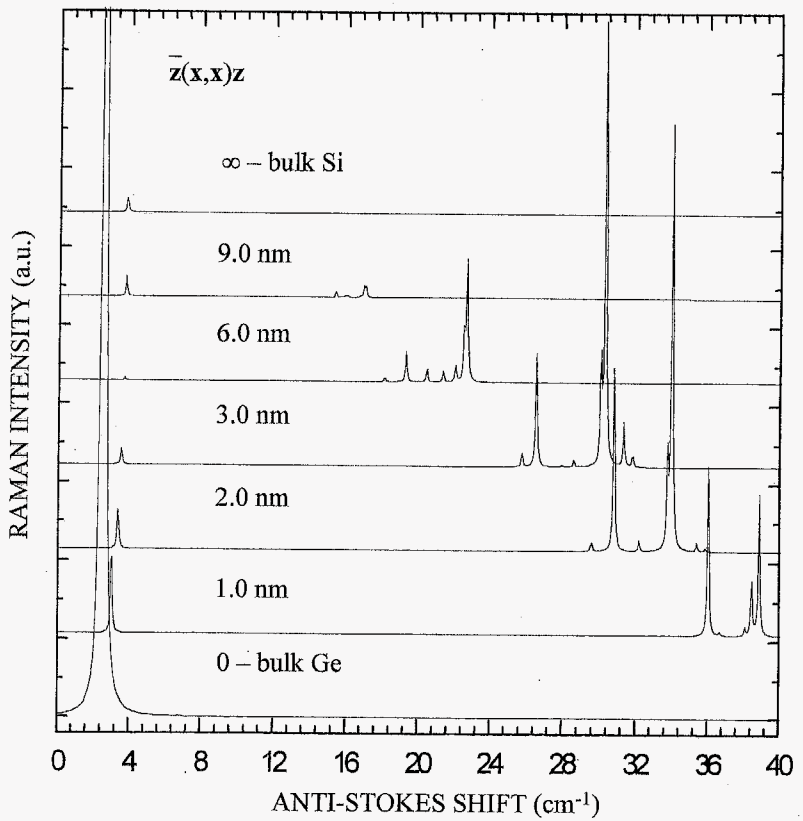
- cubical QDC $L = 3$ nm, $D = 6$ nm
  - x increases
  - Material parameters of quantum dot change
    - (1) phonon spectrum transformation
      - Raman peaks shift
    - (2) breaking of the symmetry of the displacement
      - intensity redistribution

The effect of QDC geometry on the Raman spectra



- Tetragonal Ge/Si QDC
  $L_x = L_y = 3.0$ nm, $D_x = D_y = 9.0$ nm
- $D_z = 2L_z$
- D_z increases
- QBZ shrinks in [[001]] quasi-crystallographic direction
  - acoustical phonon dispersion branches are folded at lower energy
- QDC symmetry changes
  - significant redistribution of the intensity and strong shift of peaks related to some modes.

The effect of interdot distance on QDC Raman spectra



- Cubical Ge/Si QDC L – 3 nm
- H increases
- QBZ shrinks → acoustical phonon dispersion branches are folded at lower energy
- infinite small H → bulk germanium
- infinite large H → bulk silicon
  - only the lowest longitudinal branch is active.
- H ~ L → symmetry breaking is most significant
  - The upper longitudinal and mixed modes are most active

Conclusions

- 3D regimentation of QDs leads to modified phonon dispersion /O.L.L. and A.A.Balandin, PRB **66**, 245319 (2002)./
  - folding of acoustic phonons and
  - quasi-optical phonon branches appearance
- Fine structure in light scattering spectra
- Modified electron-phonon scattering
- Nonlinear thermoelectric properties

  /A.A.Balandin and O.L.L., APL **82**, 415 (2003)./