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

Olga L. Lazarenkova and Alexander A. Balandin
Nano-Device Laboratory
Department of Electrical Engineering
University of California, Riverside

Current affiliation: Nanoelectronic Simulation Applied Cluster Computing Group
Jet Propulsion Laboratory
California Institute of Technology, Pasadena
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\textsubscript{1-x}Eu\textsubscript{x}Te
  - (C) the last PbSe layer of a 60-period PbSe/ Pb\textsubscript{1-x}Eu\textsubscript{x}Te dot crystal.

  The insets show the 2D power spectra of the AFM images.

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

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.

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

K For $x$-component

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

$$+ \frac{\partial}{\partial x} c_{12} \frac{\partial u_y}{\partial y} + \frac{\partial}{\partial x} c_{12} \frac{\partial u_z}{\partial z} + \frac{\partial}{\partial y} c_{44} \frac{\partial u_y}{\partial x} + \frac{\partial}{\partial z} c_{44} \frac{\partial u_z}{\partial x}$$

K For $y$-component replace $x \rightarrow y$, $y \rightarrow z$, $z \rightarrow x$.

K For $z$-component replace $x \rightarrow z$, $y \rightarrow x$, $z \rightarrow y$.

K In subscripts of elastic constants

K 1 $\equiv xx$ - strain along the displacement

K 2 $\equiv yy$ - strain perpendicular to the displacement

K 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 \(\rightarrow 3(N_xN_yN_z)\) 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
  – $L = 3 \text{ nm}, D = 6 \text{ nm}$
  – $c_{11}(\text{Ge}) = 12.40 \cdot 10^{10} \text{N/m}^2$,
  – $c_{12}(\text{Ge}) = 4.13 \cdot 10^{10} \text{N/m}^2$,
  – $c_{44}(\text{Ge}) = 6.83 \cdot 10^{10} \text{N/m}^2$,
  – $c_{11}(\text{Si}) = 16.577 \cdot 10^{10} \text{N/m}^2$,
  – $c_{12}(\text{Si}) = 6.393 \cdot 10^{10} \text{N/m}^2$,
  – $c_{44}(\text{Si}) = 7.962 \cdot 10^{10} \text{N/m}^2$;
  – $\rho(\text{Ge}) = 5.3234 \cdot 10^3 \text{kg/m}^3$,
  – $\rho(\text{Si}) = 2.3290 \cdot 10^3 \text{kg/m}^3$.
• The spectrum is normalized to the vibration intensity in $\text{Ge}_{0.113}\text{Si}_{0.887}$
Electromagnetic wave in QDC

- In effective medium approximation
  \[ D = \hat{\varepsilon} \varepsilon_0 F(r, k, \omega) \exp(-i\omega t), \text{ where } F(r, k, \omega) = A \exp(ik \cdot r) \]
- \( A \) is the light polarization vector: \( (A \cdot k) = 0 \),
- \( \lambda \) is the light wavelength in vacuum.
  \[ |k| = \frac{2\pi \sqrt{\varepsilon}}{\lambda} \]
- Phonon vibrations cause local change of the dielectric susceptibility tensor \( \varepsilon \)
  - in cubic semiconductors
    \[ \varepsilon_{xx} = \varepsilon_{xx}^0 + q_{1111} \frac{\partial U_x}{\partial x} + q_{1122} \frac{\partial U_y}{\partial y} + q_{1122} \frac{\partial U_z}{\partial z} \]
  - \( q_{ijkl} \) are components of the photoelastic tensor
Probability of anti-Stokes Raman scattering in QDC

\[ P_{ii} \propto \left| \langle D_f | D_i \rangle \right|^2 = \varepsilon^2 \varepsilon_0^4 \delta(\omega_f - \omega_i - \Omega) \delta(k_f - k_i - q) \left( \frac{V}{d_x d_y d_z} \right)^2 \int d \mathbf{r} \exp(-i \mathbf{q} \cdot \mathbf{r}). \]

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

\[ |I\rangle, \quad |i\rangle \equiv |1\rangle \quad \text{Lattice vibration (phonon)} \]

\[ |2\rangle, \quad |f\rangle \equiv |2\rangle \quad \text{Scattering by perturbation} \]

No perturbation
Spatial distribution of the light scattering in QDC

- The lowest LA mode
- Ge/Si cubical QDC
  - $L = 3$ nm, $D = 6$ nm
  - $q_{1111}(Ge) = 40.0,$
    $q_{1122}(Ge) = 35.0,$
    $q_{1111}(Si) = 16.0,$
    $q_{1122}(Si) = -2.0;$
    $\varepsilon^0(Si) = 11.9,$
    $\varepsilon^0(Ge) = 16.2.$
Raman spectra of Ge$_x$Si$_{1-x}$/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
  \( L_x = L_y = 3.0 \text{ nm}, \ D_x = D_y = 9.0 \text{ nm} \)
- \( D_z = 2L_z \)
- \( D_z \) 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 \( \rightarrow \) acoustical phonon dispersion branches are folded at lower energy
- infinite small $H \rightarrow$ bulk germanium
- infinite large $H \rightarrow$ bulk silicon
- only the lowest longitudinal branch is active.
- $H \sim L \rightarrow$ 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)/