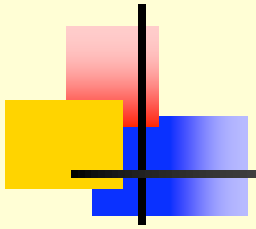
DMR-0205328

ITR: Modeling and Simulations of Quantum Phenomena in Semiconductor Structures of Reduced Dimensions



PI: Mei-Yin Chou (Georgia Tech)

Co-PI's: Uzi Landman (Georgia Tech)

Cyrus Umrigar (Cornell University)

Xiao-Qian Wang (Clark Atlanta University)

Project Summary

- A comprehensive simulation of the electrical, optical, vibrational, structural, and transport properties of various nanostructures, with the focus on their size dependence.

- Issues being examined include stability, growth, electronic structure, vibrational modes, conductance, and nanocontacts.

- The goal is to make use of the computational capabilities provided by today's information technology to perform theoretical modeling of materials that may play a key role in the hardware development for tomorrow's information technology.

Participants

Graduate Students	Postdocs/Research Associates
Anthony Cochran	Silvio a Beccara
Damian Cupid	Robert Barnett
Oladipo Fadiran	Zineb Felfli
Alexis Nduwimana	Wolfgang Geist
Igor Romanovsky	Devrim Guclu
Li Yang	Ryza Musin
Longping Yuan	Andrew Scherbakov
	Xinyuan Zhao

Computational Methods

- First-principles molecular dynamics simulations within density functional theory with pseudopotentials and plane waves

  Stability, growth, energetics, electronic wave functions, vibrational modes, *etc.*

- Quantum Monte Carlo methods (variational and diffusion)

  Energy gap, excitation energies, algorithm development (linear scaling with nonorthogonal Wannier functions, calculation of optical transition strength using DMC to obtain the imaginary-time correlation function, finite magnetic field)

- Many-body perturbation theory

  GW quasiparticle energies, optical excitations including exciton effects (Bethe-Salpeter equation, evaluate the Coulomb scattering matrix in real space using Wannier functions)

Computational Methods (continued)

- First-principles calculation of conductance

  Recursion-transfer-matrix method to solve the coupled differential equation involving reflected and transmitted waves (Hirose and Tsukada) and eigenchannel analysis for the transmission (Brandbyge *et al.*)

  The Green's function method and the self-consistent Lippmann-Schwinger equation with scattering boundary condition (Lang *et al.*)

Project Progress

- Quantum confinement, electronic properties, and vibrational properties of semiconductor nanowires, including Si, Ge, Si-Ge, GaAs, GaN. (Chou and Wang)

- Transport properties, nanowire junctions, nanocontacts (Landman)

- 2D quantum dots at low and high magnetic fields: electron correlation, spin configuration, and various many-body ground states. (Umrigar and Chou)

A Laser Ablation Method for the Synthesis of Crystalline Semiconductor Nanowires, *Morales and Lieber, Science 279, 208 (1998)*

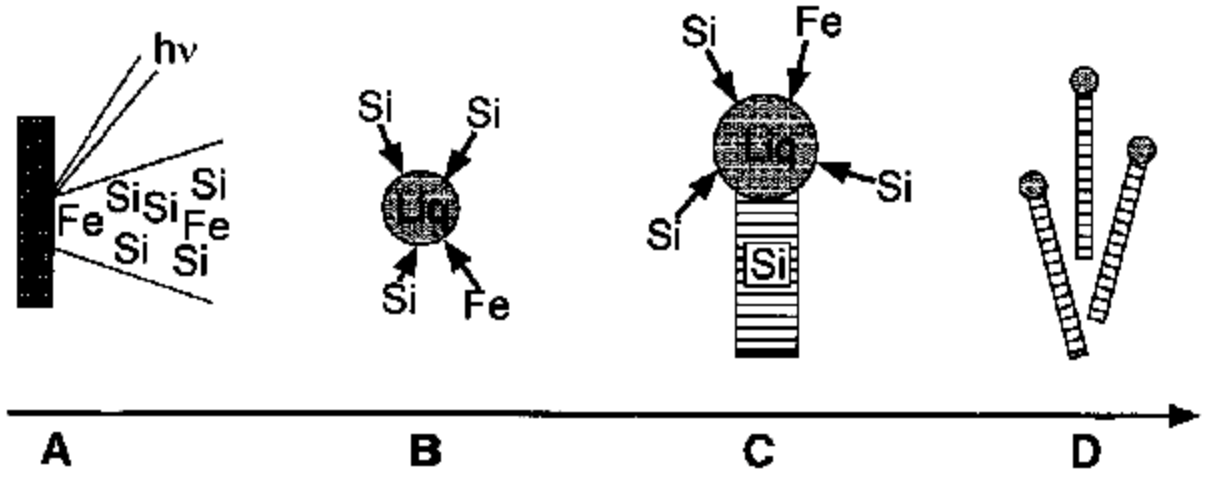
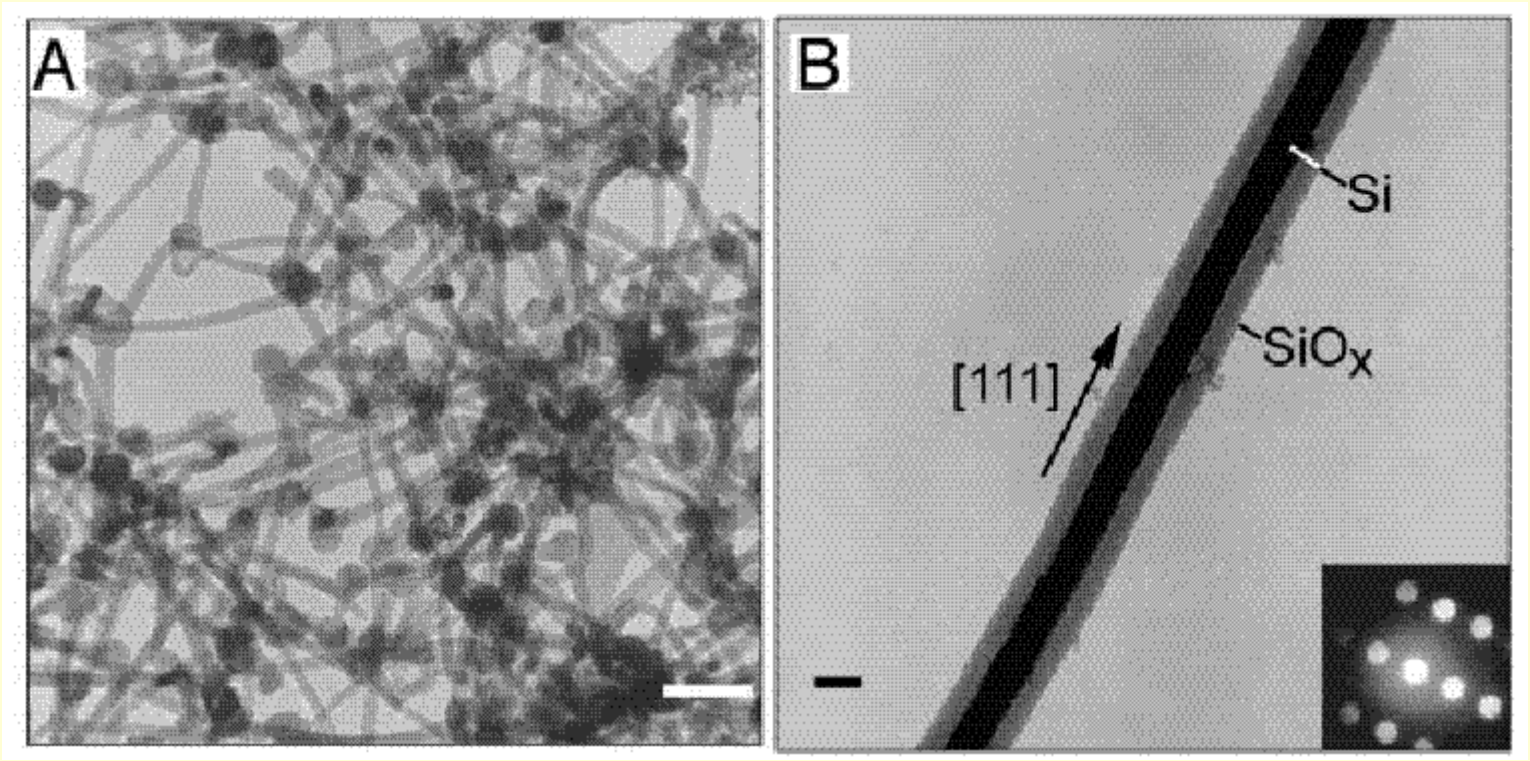


Fig. 3. Proposed nanowire growth model. (**A**) Laser ablation with photons of energy $h\nu$ of the $Si_{1-x}Fe_x$ target creates a dense, hot vapor of Si and Fe species. (**B**) The hot vapor condenses into small clusters as the Si and Fe species cool through collisions with the buffer gas. The furnace temperature (Fig. 1A) is controlled to maintain the Si-Fe nanocluster in a liquid (Liq) state. (**C**) Nanowire growth begins after the liquid becomes supersaturated in Si and continues as long as the Si-Fe nanoclusters remain in a liquid state and Si reactant is available. (**D**) Growth terminates when the nanowire passes out of the hot reaction zone (in the carrier gas flow) onto the cold finger and the Si-Fe nanoclusters solidify.



A Laser Ablation Method for the Synthesis of Crystalline Semiconductor Nanowires

Morales and Lieber, Science 279, 208 (1998)

Room-Temperature Ultraviolet Nanowire Nanolasers

Huang et al., Science 292, 1897 (2001)

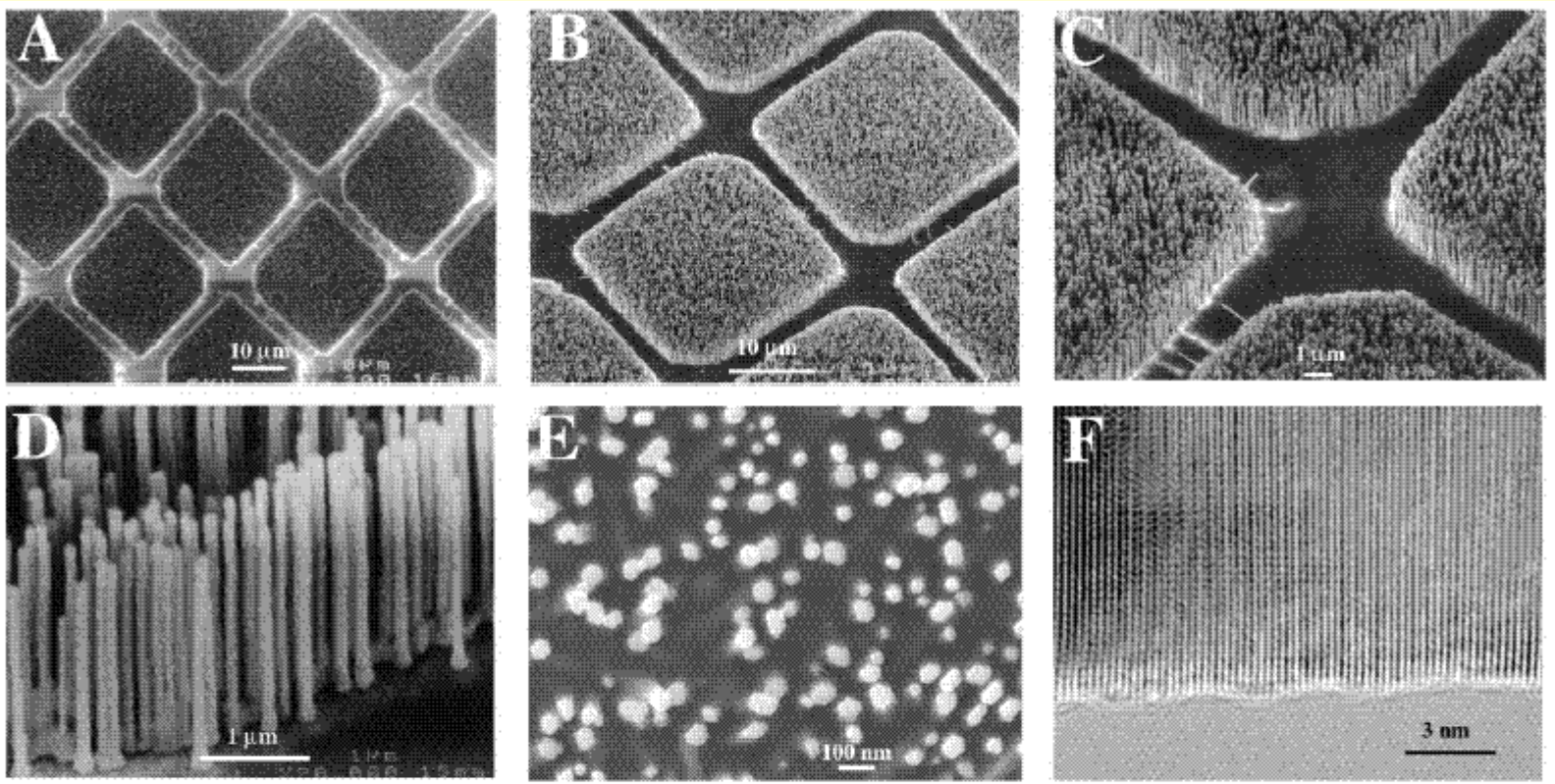
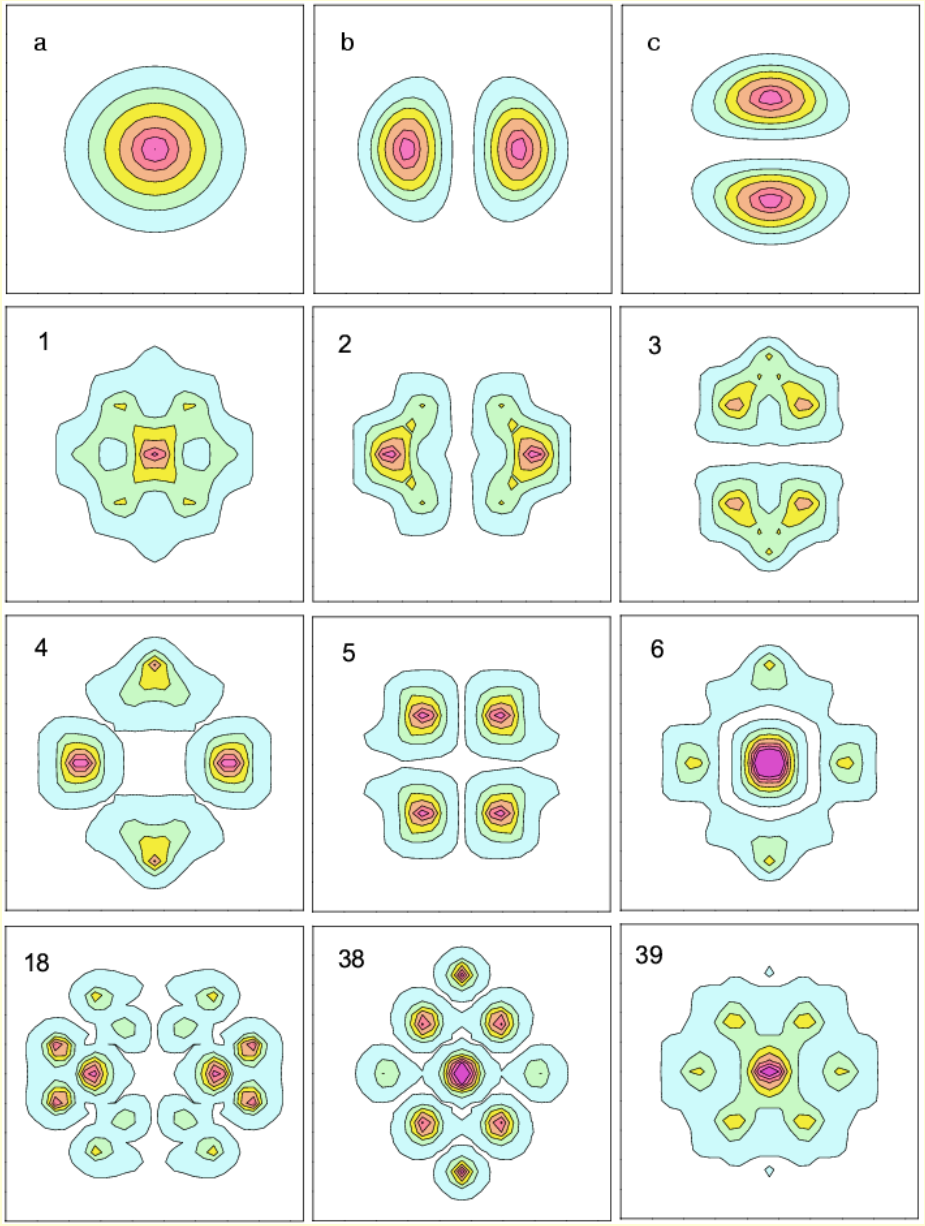


Fig. 1. (**A** through **E**) SEM images of ZnO nanowire arrays grown on sapphire substrates. A top view of the well-faceted hexagonal nanowire tips is shown in (E). (**F**) High-resolution TEM image of an individual ZnO nanowire showing its <0001> growth direction. For the nanowire growth, clean (110) sapphire substrates were coated with a 10 to 35 Å thick layer of Au, with or without using TEM grids as shadow masks (micro contact printing of thiols on Au followed by selective etching has also been used to create the Au pattern). An equal amount of ZnO powder and graphite powder were ground and transferred to an alumina boat. The Au-coated sapphire substrates were typically placed 0.5 to 2.5 cm from the center of the boat. The starting materials and the substrates were then heated up to 880° to 905°C in an Ar flow. Zn vapor is generated by carbothermal reduction of ZnO and transported to the substrates where ZnO nanowires grow. The growth generally took place within 2 to 10 min (*15*).

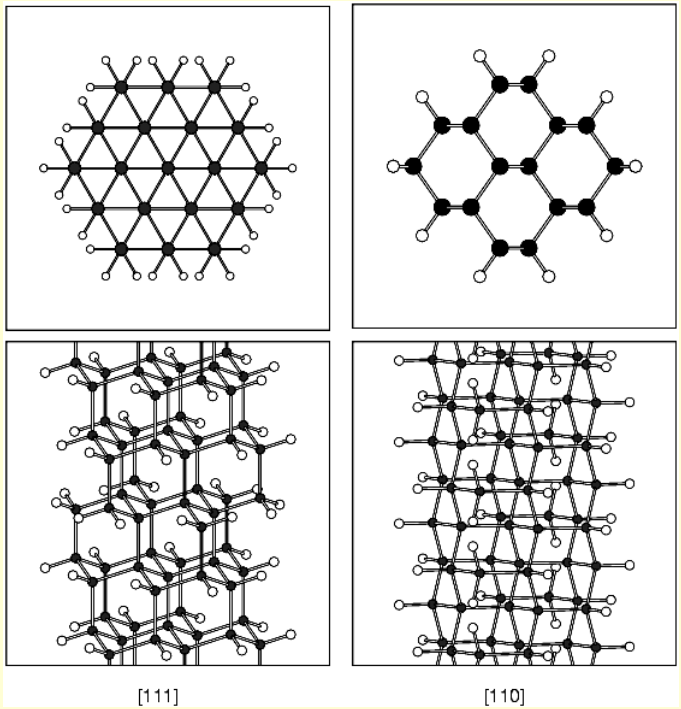
Electron Confinement in Si Nanowires



Band-by-band charge distribution

[110] direction, d=1.2 nm

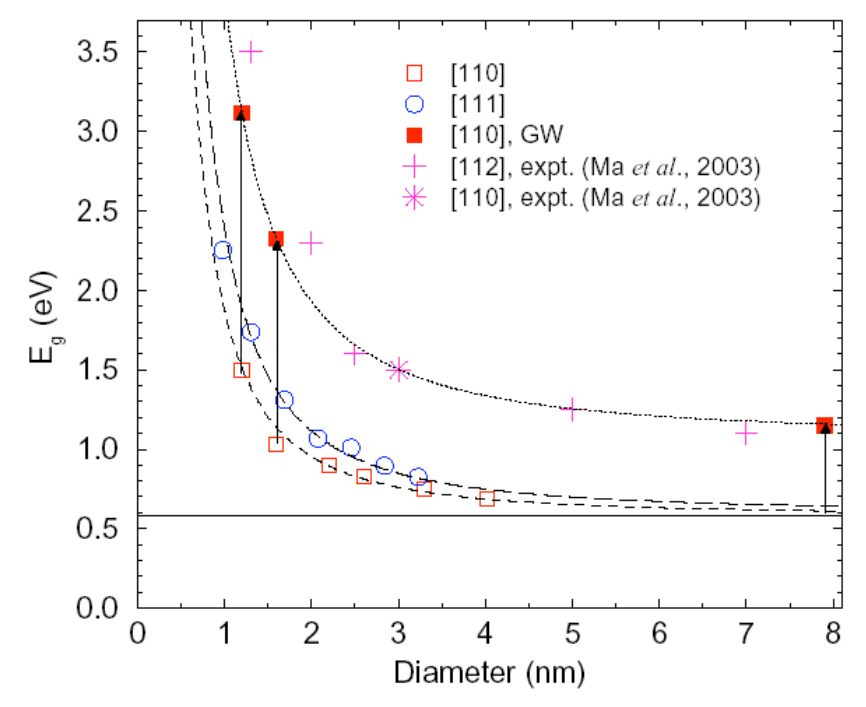
Structure of Si nanowires



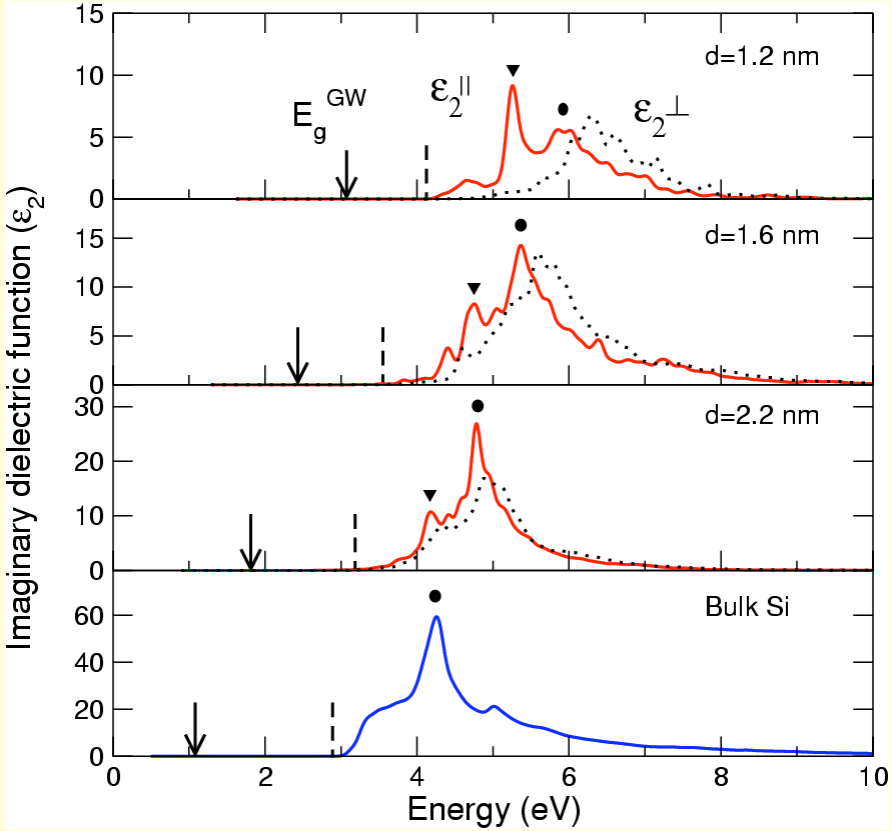
[111] [110]

Electron Structure of Si Nanowires

Direct gap as a function of diameter

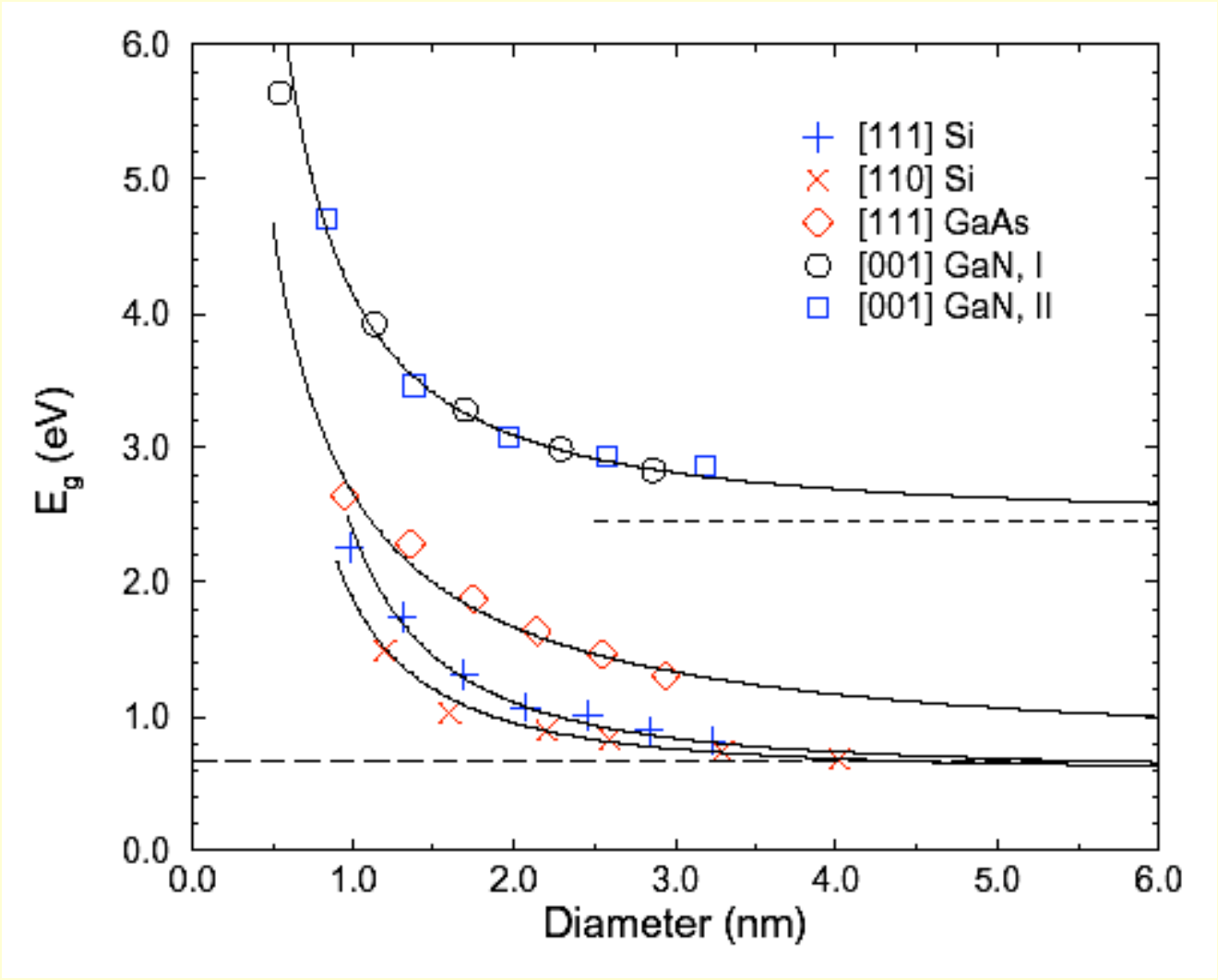


Optical absorption



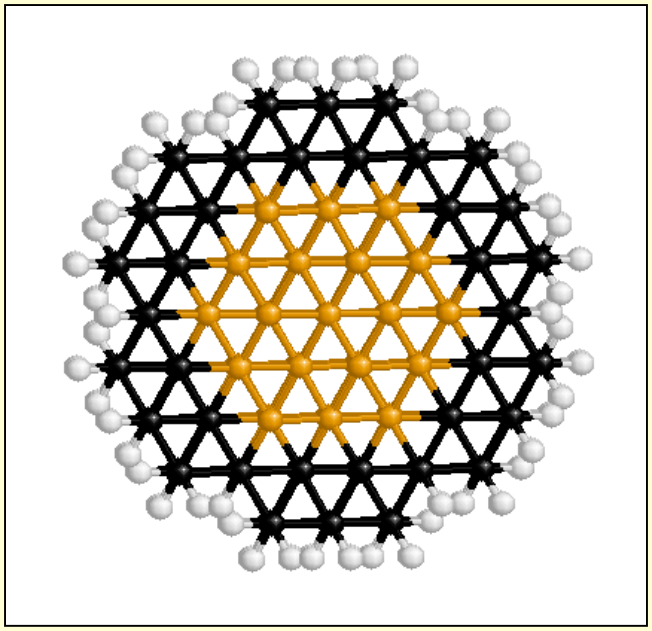
Zhao *et al*. PRL 92, 236805 (2004).

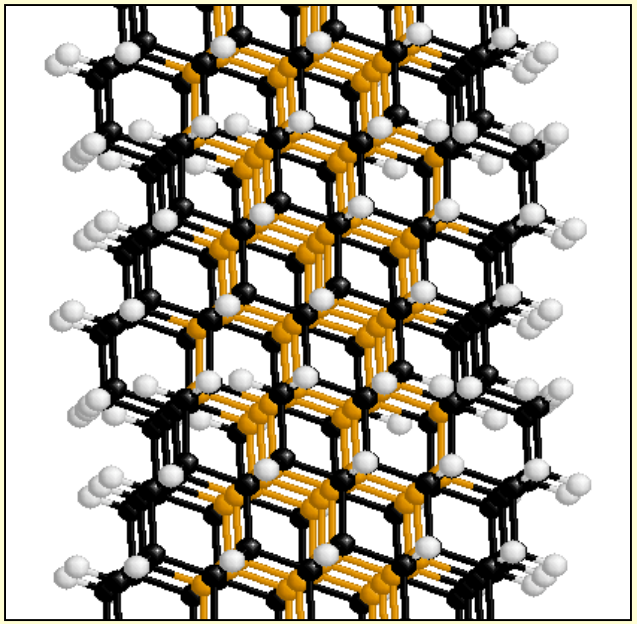
III-V Semiconductor nanowires (Zhao *et al*. poster)



Also, Ge nanowires (Nduwimana *et al*. poster)

Core-Shell Nanowire Heterostrctures



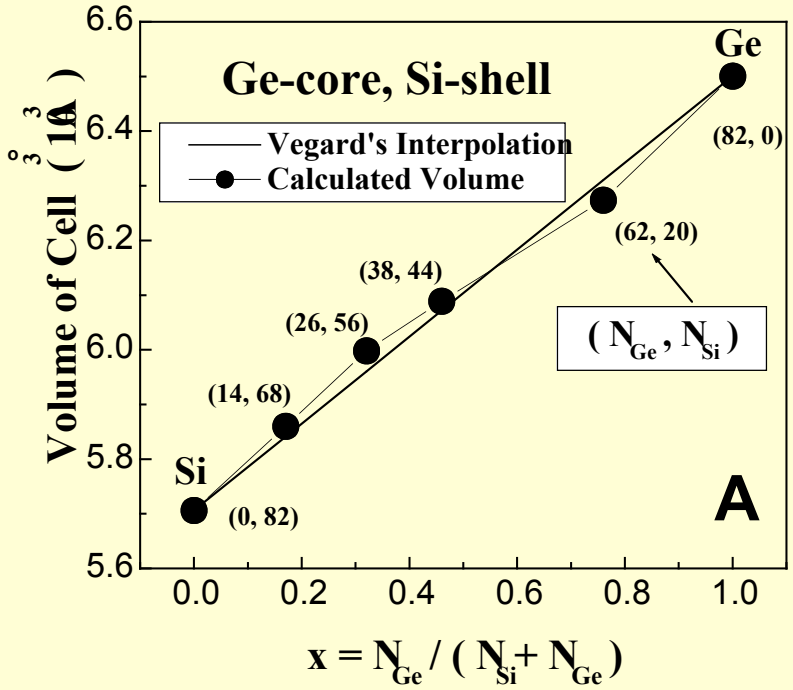


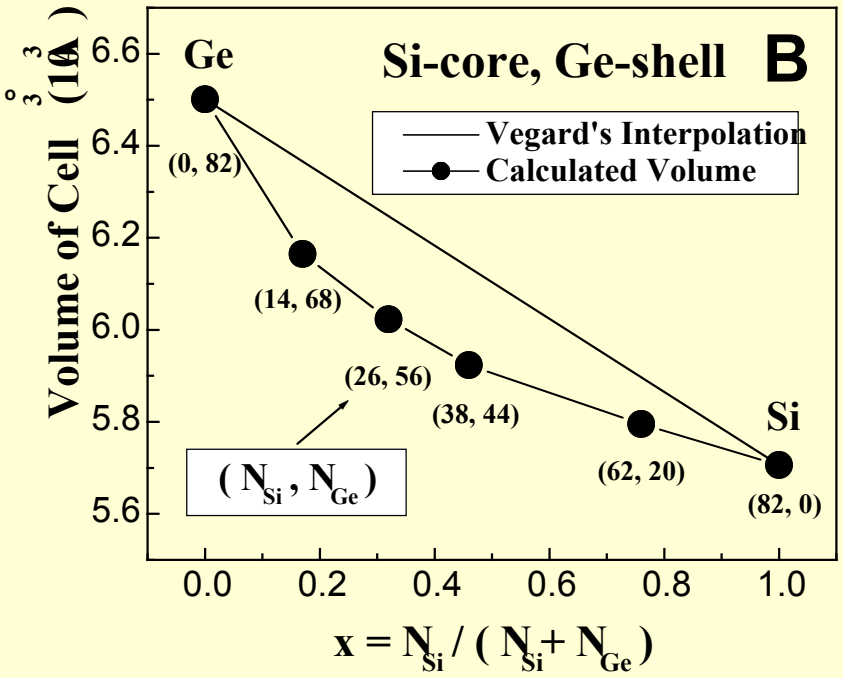
Si-Ge heterostructures: high mobility devices

R. Z. Musin and X. Q. Wang, Clark Atlanta University
Phys. Rev. B (submitted); poster

Si-Ge Core-Shell/Multishell Nanowires

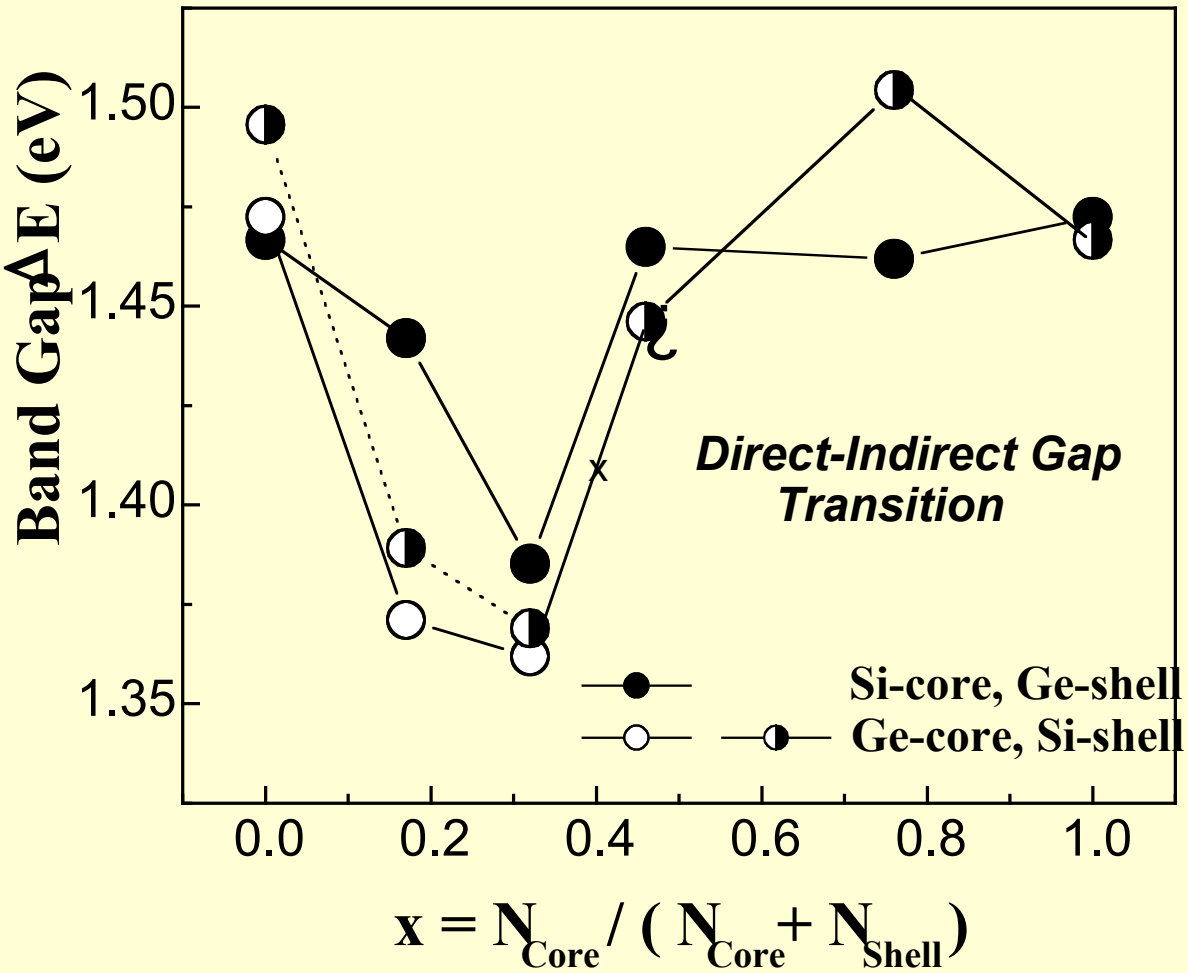
- Si-Ge core-shell and multishell nanowire heterostructures synthesized using chemical vapor deposition method (Lauhon *et al*., Nature 420, 57, 2002)
- Si-Ge heterostructures: high mobility devices
- 4% lattice mismatch: compressively strained Ge and tensile-strained Si
- Ge-core Si-shell structure (amorphous Si shell completely crystallized following thermal annealing)
- Ge deposition on Si nanowire cores
- Geometric and electronic structures of Si-Ge core-shell nanowire heterostructures studied with first-principles calculations
- Insight into the experimentally synthesized core-shell nanowire heterostructures





- Strain relaxation causes changes in geometric and electronic structures
- Negative Deviation from the Vegard's law observed, but not well understood
- Correlations between the geometric and electronic structures found (deviations from the Vegards' law and direct-indirect gap transition for Ge-core Si-shell nanowires)

Band-gap ΔE as a function of composition: bowing



- [111] nanowires: Si direct gap, Ge indirect gap
- Large bowing parameter: deviation from the linear relation for band gaps

"Phonons" in Nanowires

- Thermal properties important for heat conduction and power dissipation

  Confinement effects:

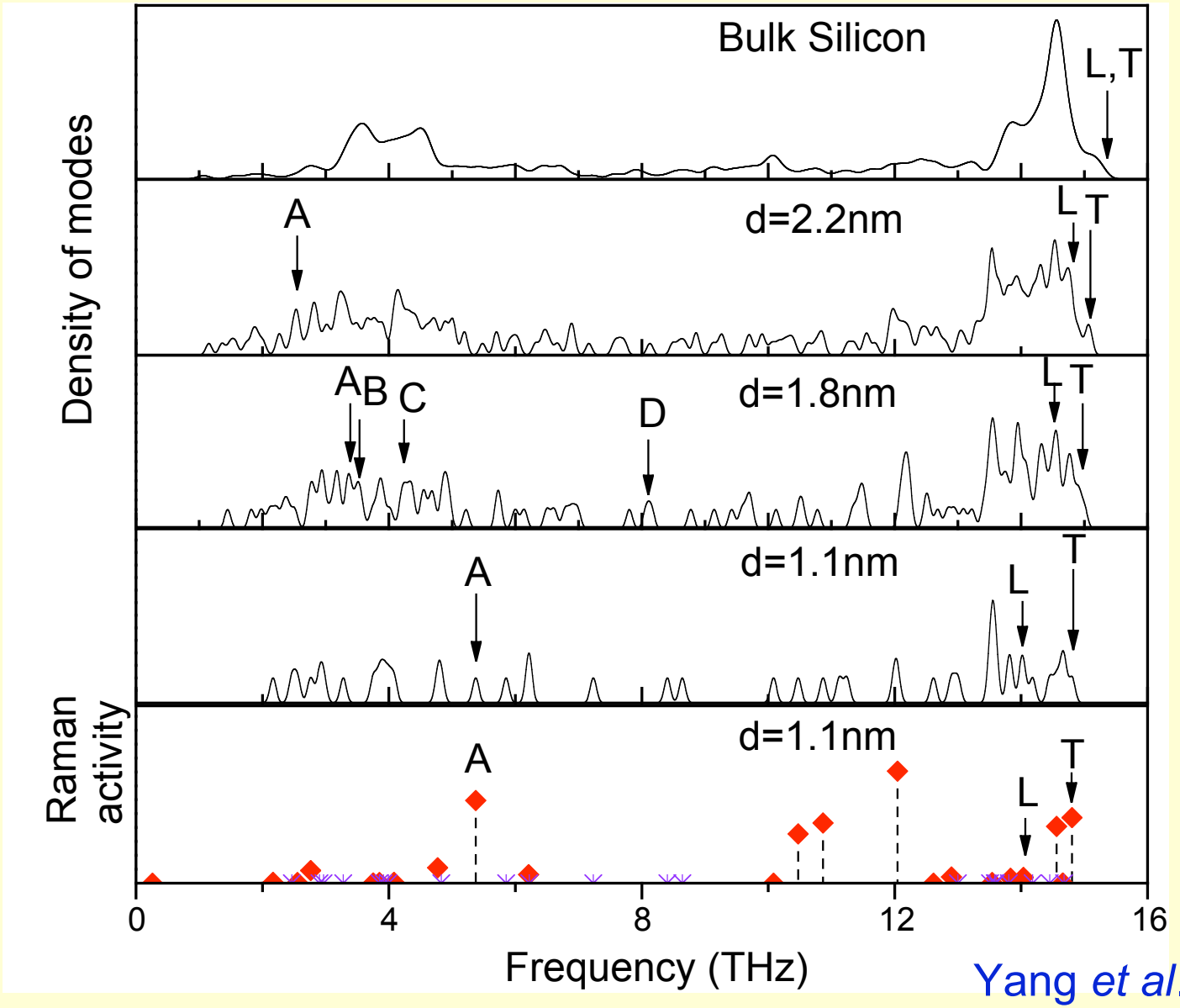
  Broadening and shifting peaks

  Acoustic phonon dispersion and group velocity modified

  Phonon distribution modified by boundary scattering

- Size and shape dependence

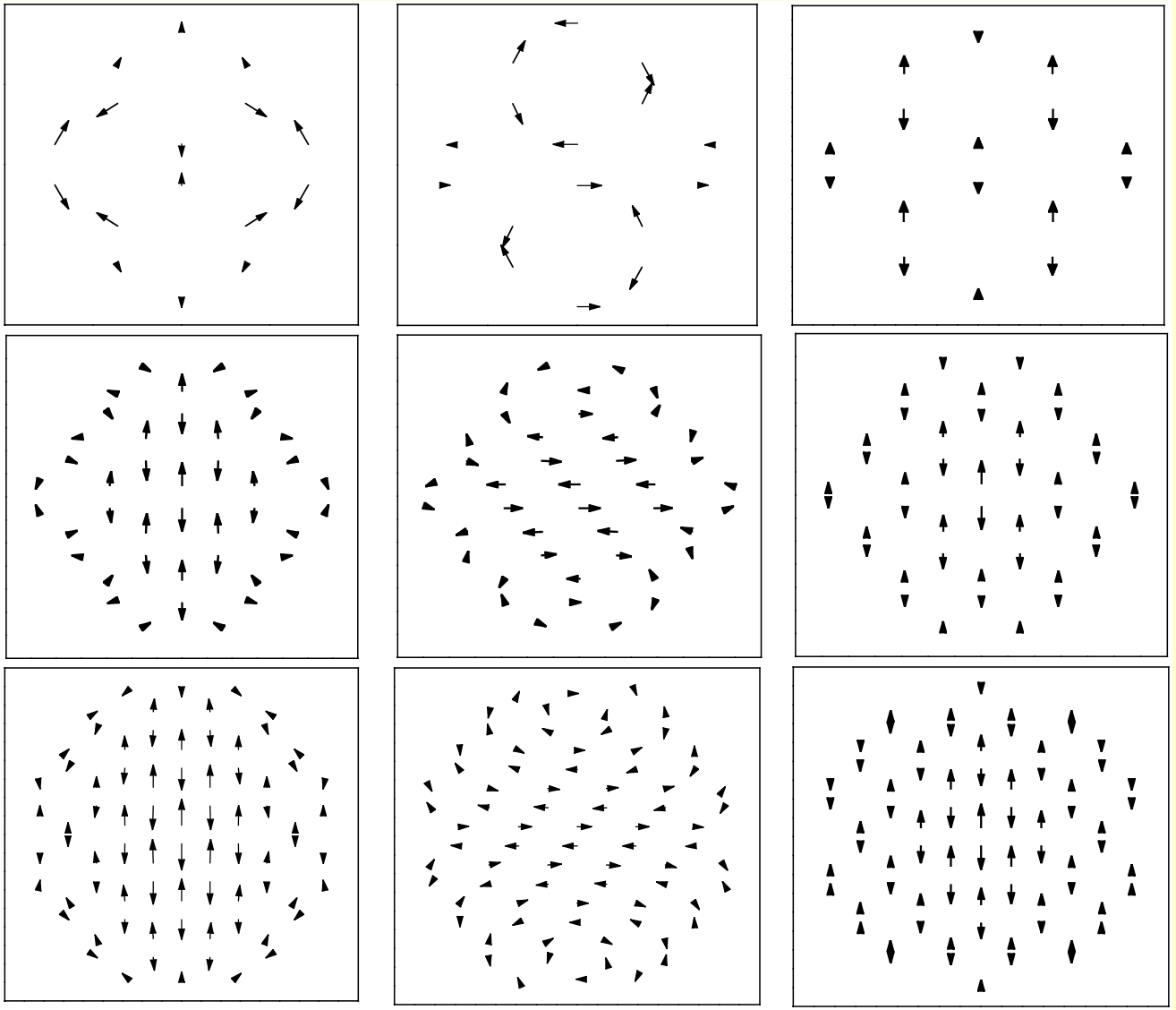
Density of modes at Γ for Si [110] Nanowires



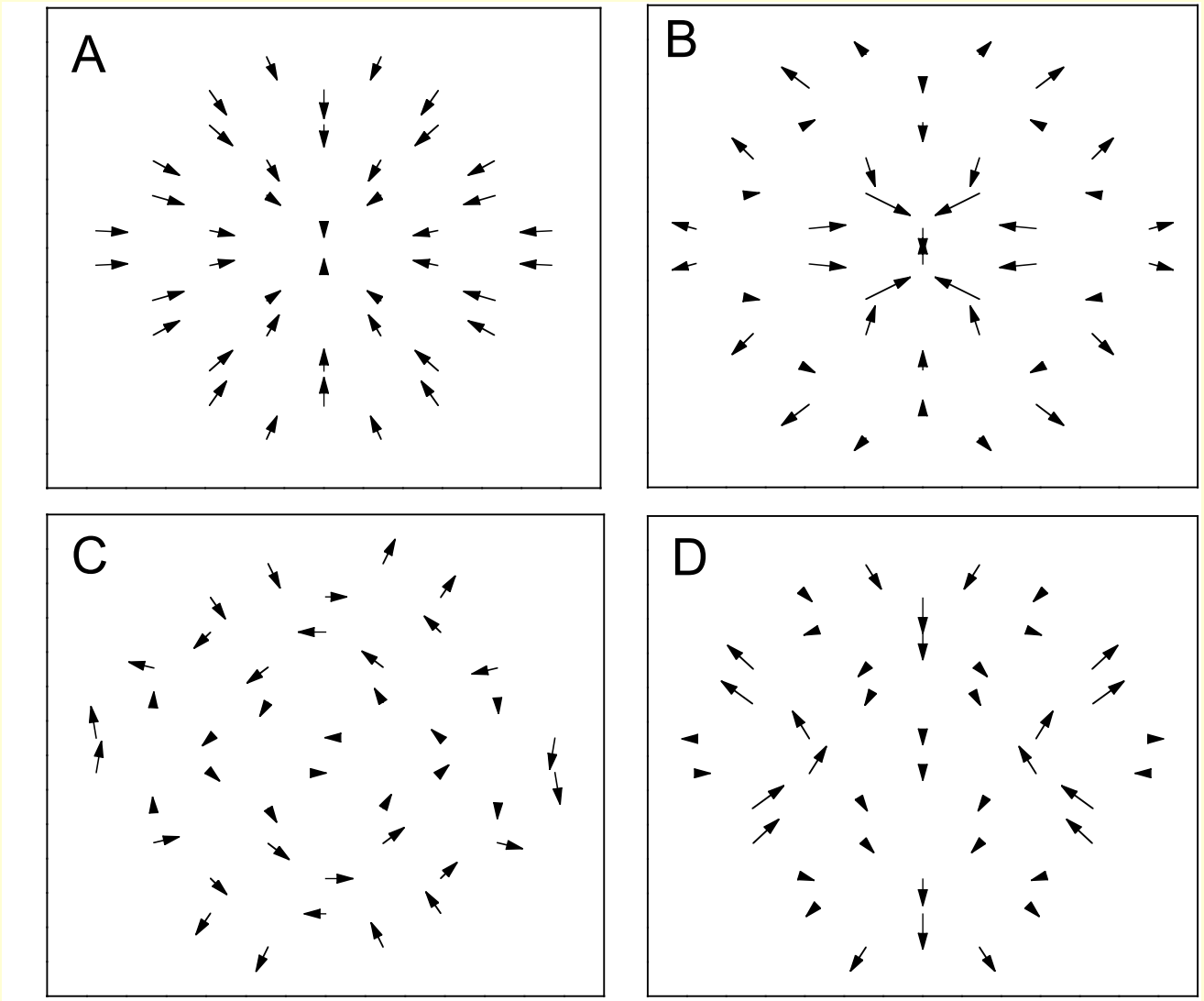
Yang *et al*. poster

Optical modes at Γ

Perpendicular Parallel

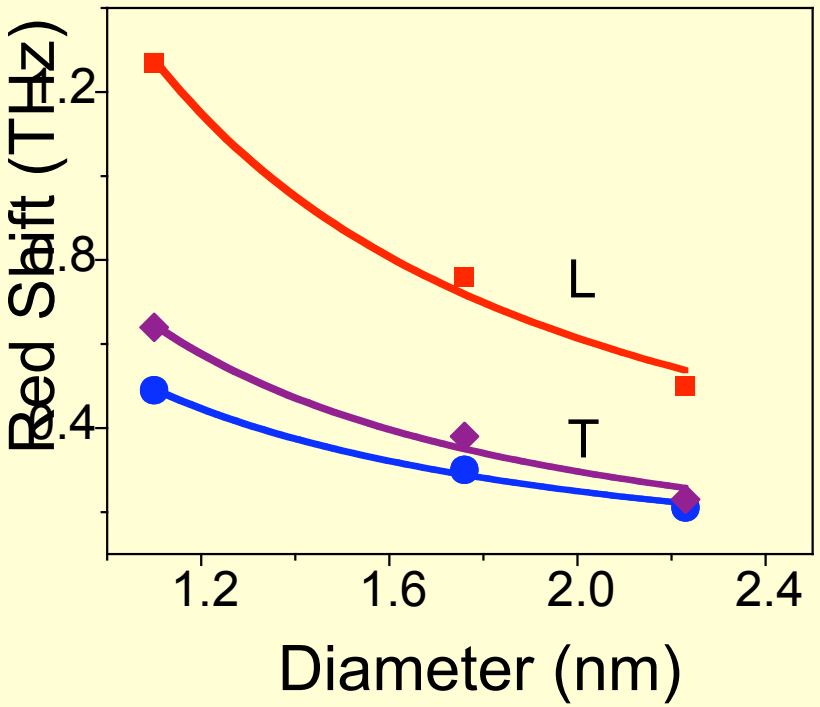


Collective modes at Γ

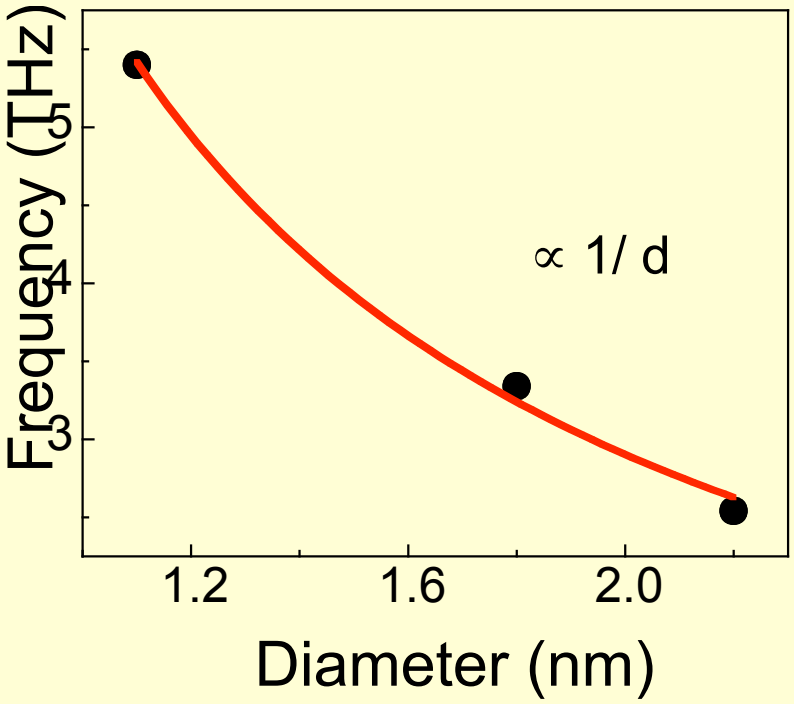


Frequency Shifts in Si Nanowires

Optical Modes at Γ



Breathing Mode at Γ



Yang *et al*. poster

Nanowire "Device" Simulations

Conductance, Contacts, Molecular Junctions, *etc.*
Landman's group, Georgia Tech

- Magnetization Oscillations in Superconducting Ballistic Nanowires:

  A giant magnetic response to applied weak magnetic fields is predicted in the ballistic Josephson junction formed by a superconducting tip and a surface, bridged by a normal-metal nanowire where Andreev states form.

  Krive *et al*. PRL 92, 126802 (2004)

- "Hydrogen welding" and "hydrogen switches" in a mono-atomic gold nanowire

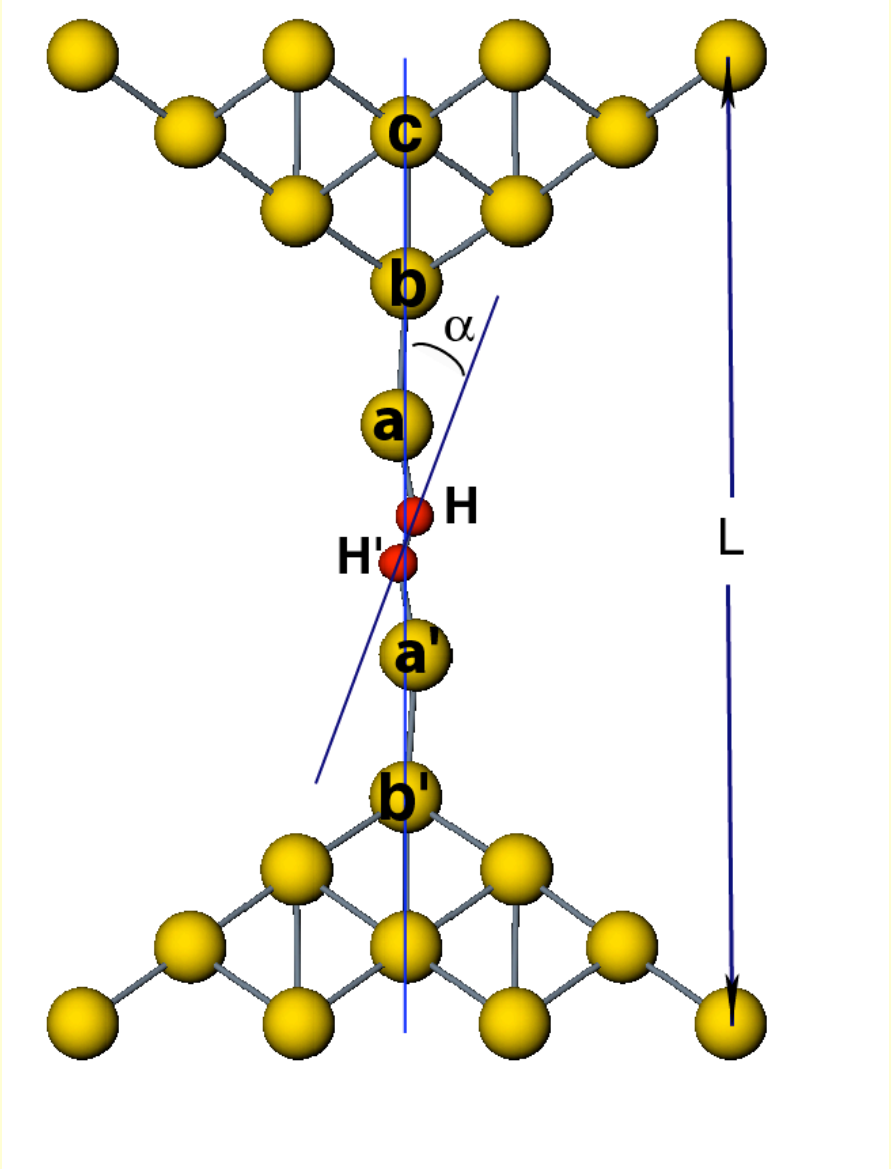
  Ab-initio molecular dynamics simulations; Structural optimization; Electrical conductance (transfer matrix), Vibrational dynamics

  R. N. Barnett, H. Hakkinen, A. G. Scherbakov, and U. Landman

Au wire setup

Recursion-transfer-matrix method to solve the coupled differential equation involving reflected and transmitted waves (Hirose and Tsukada) and eigenchannel analysis for the transmission (Brandbyge *et al.*)

The Green's function method and the self-consistent Lippmann-Schwinger equation with scattering boundary condition (Lang *et al.*)



Bare Au Wires

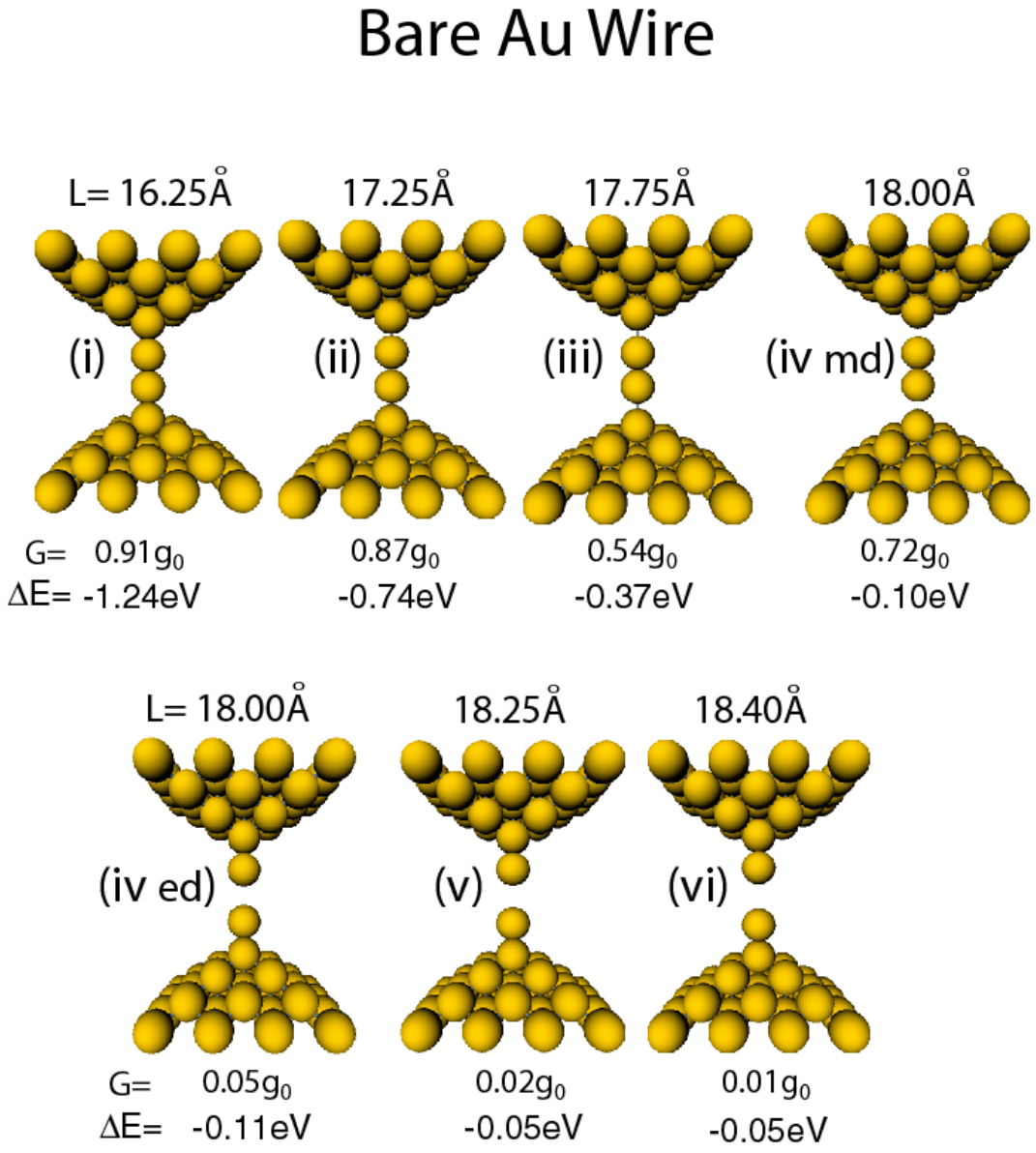


figure 2

Au Wires with Hydrogen

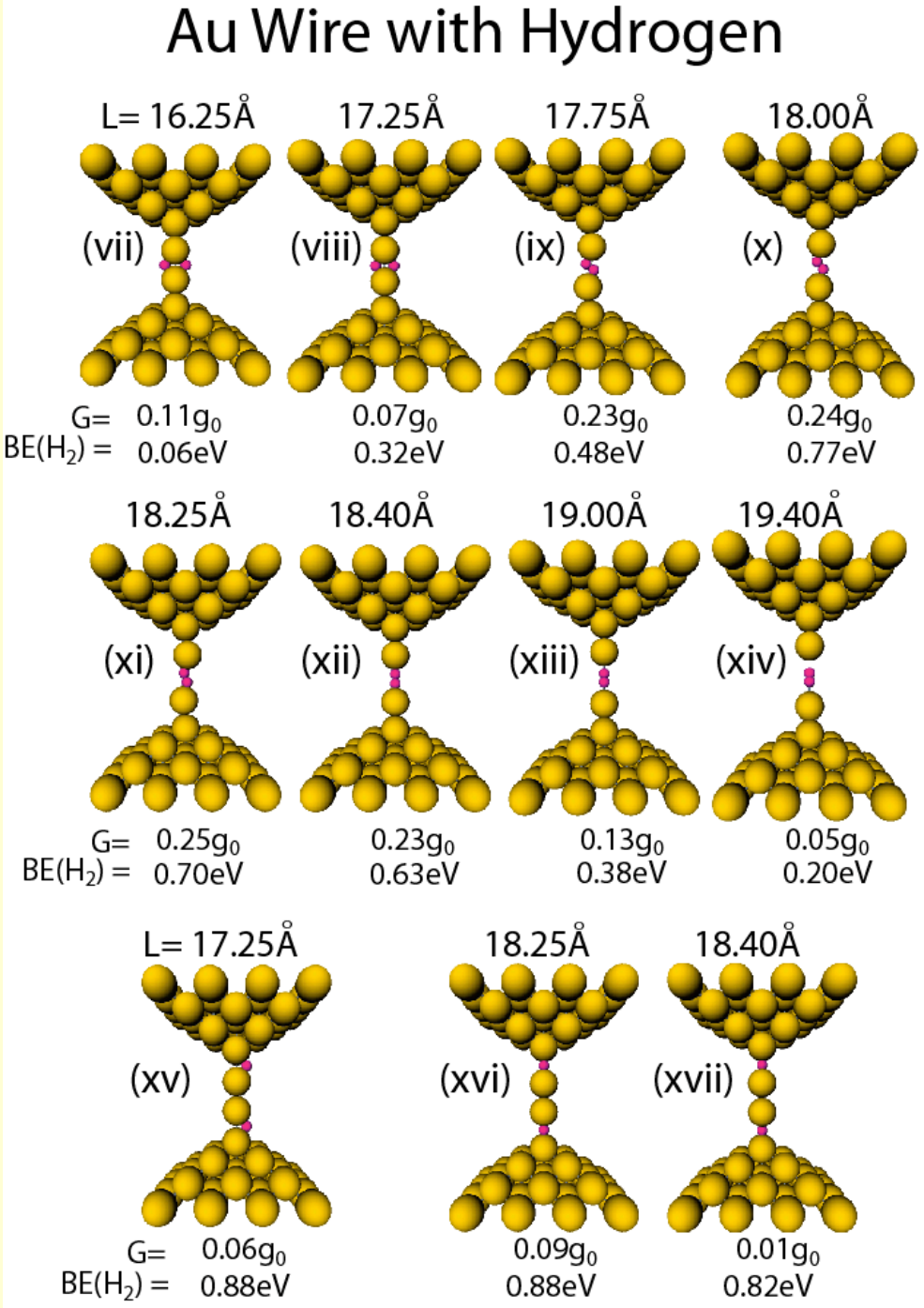


figure 3

vs Length ΔL

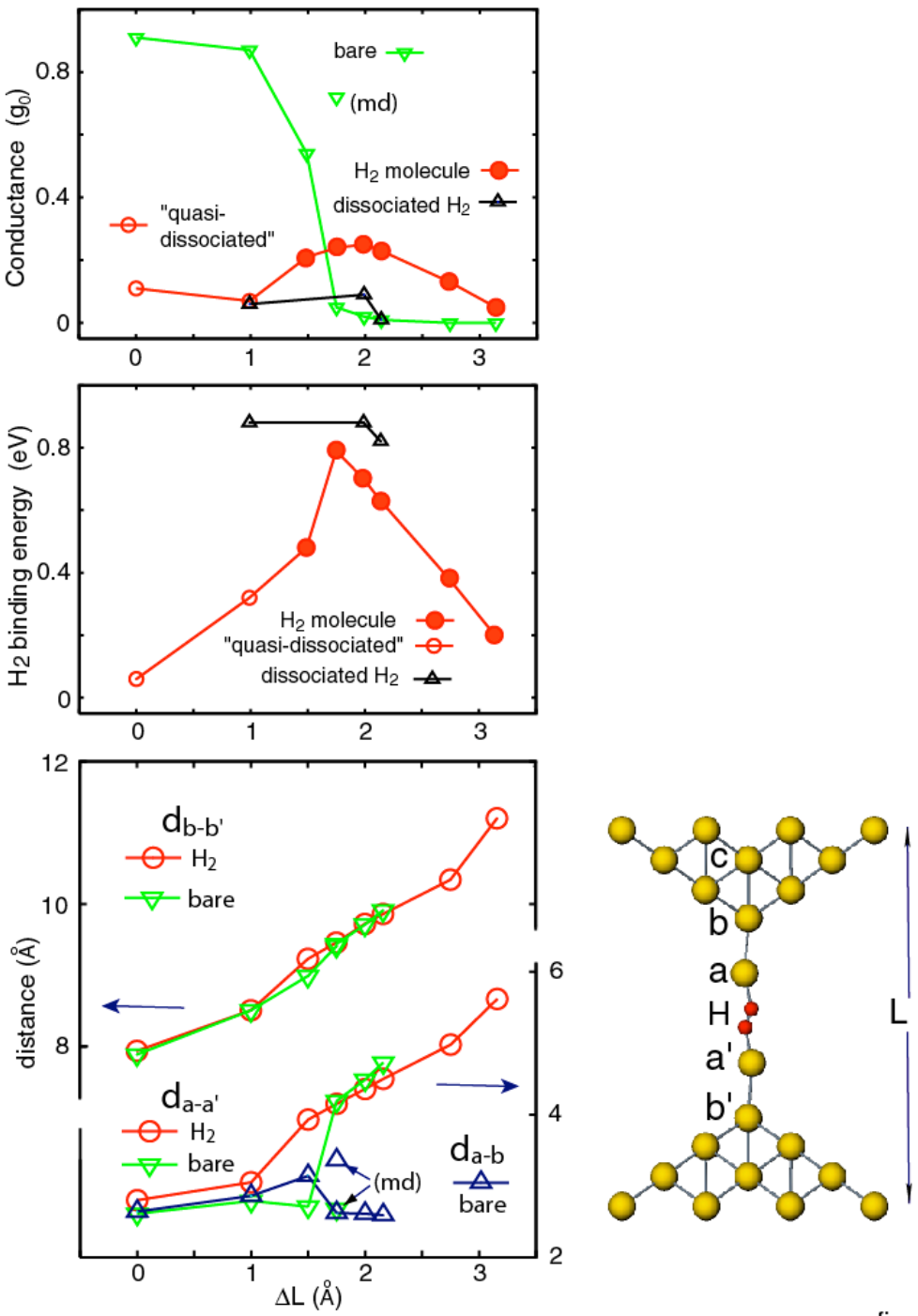


figure 4

Local Density of States

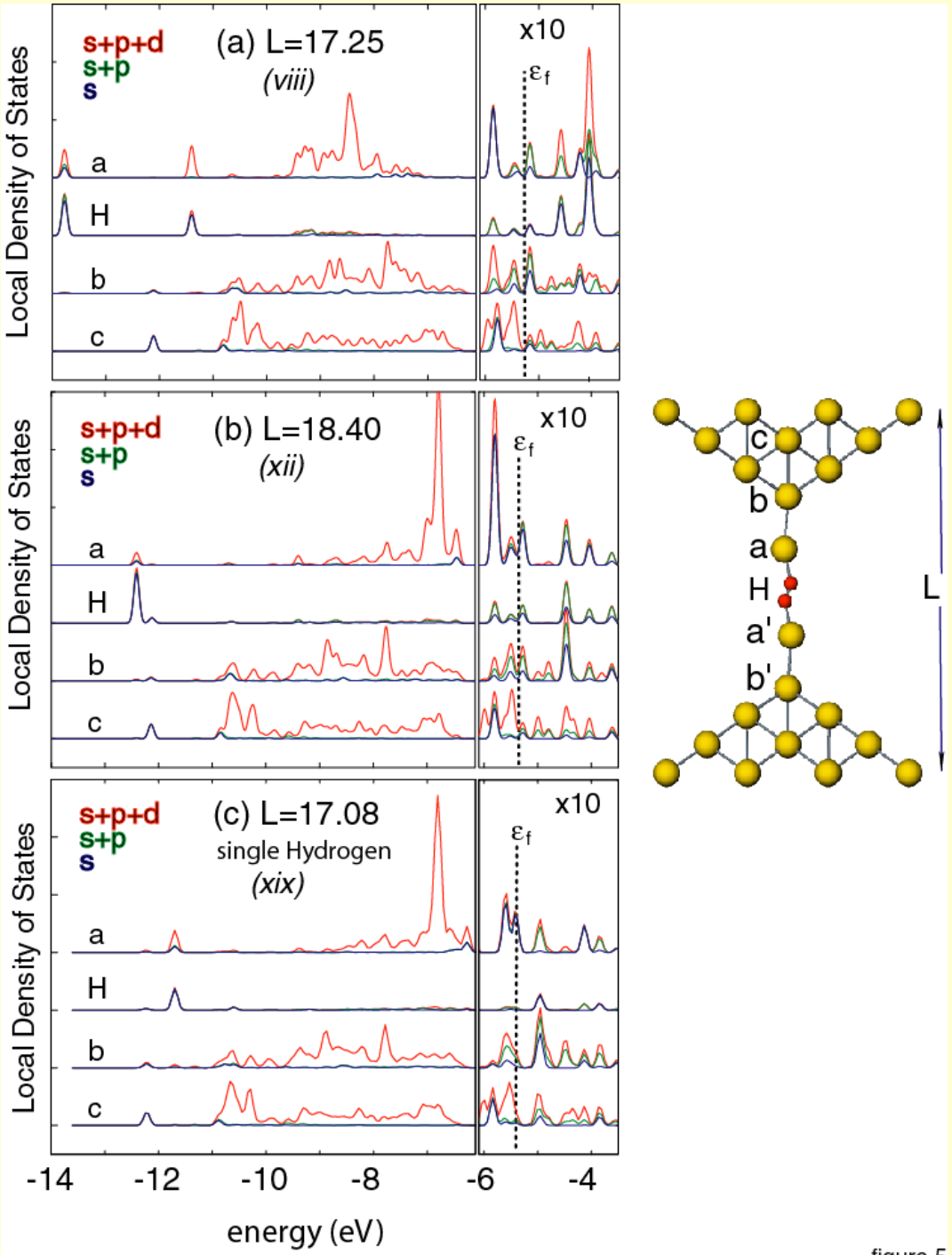


figure 5

wavefunctions

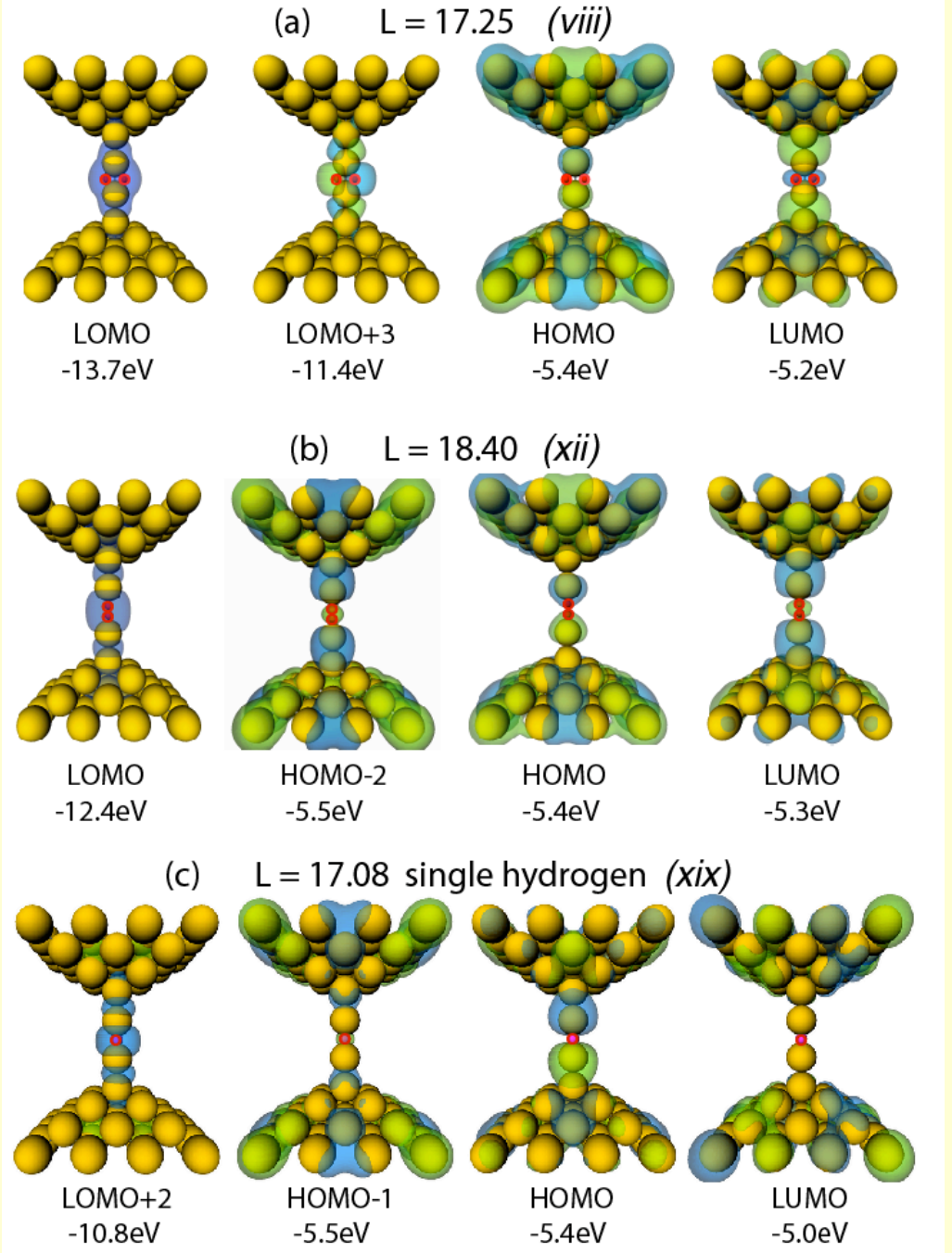


figure 6

Conductance Eigenchannels

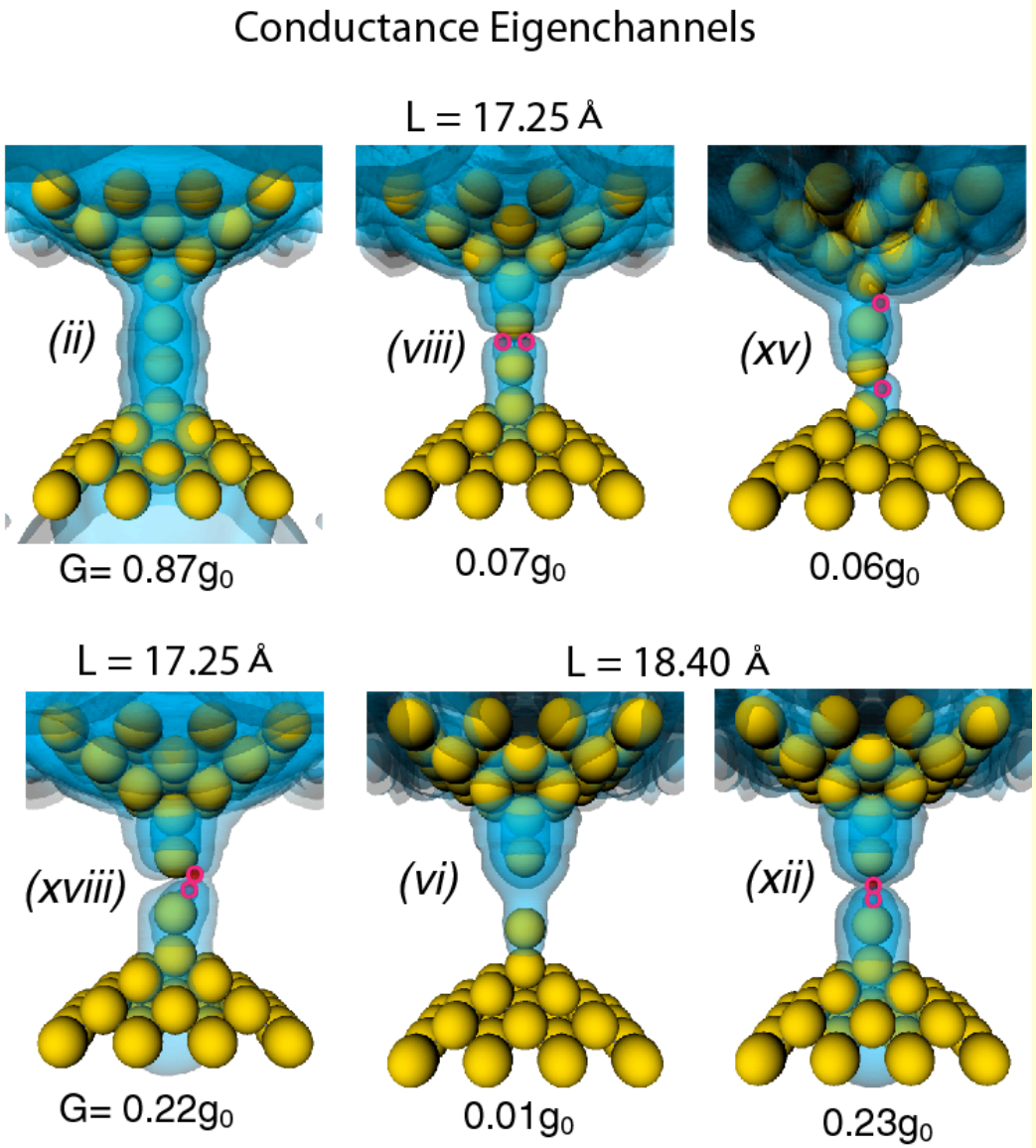


figure 7

H_2 frequencies

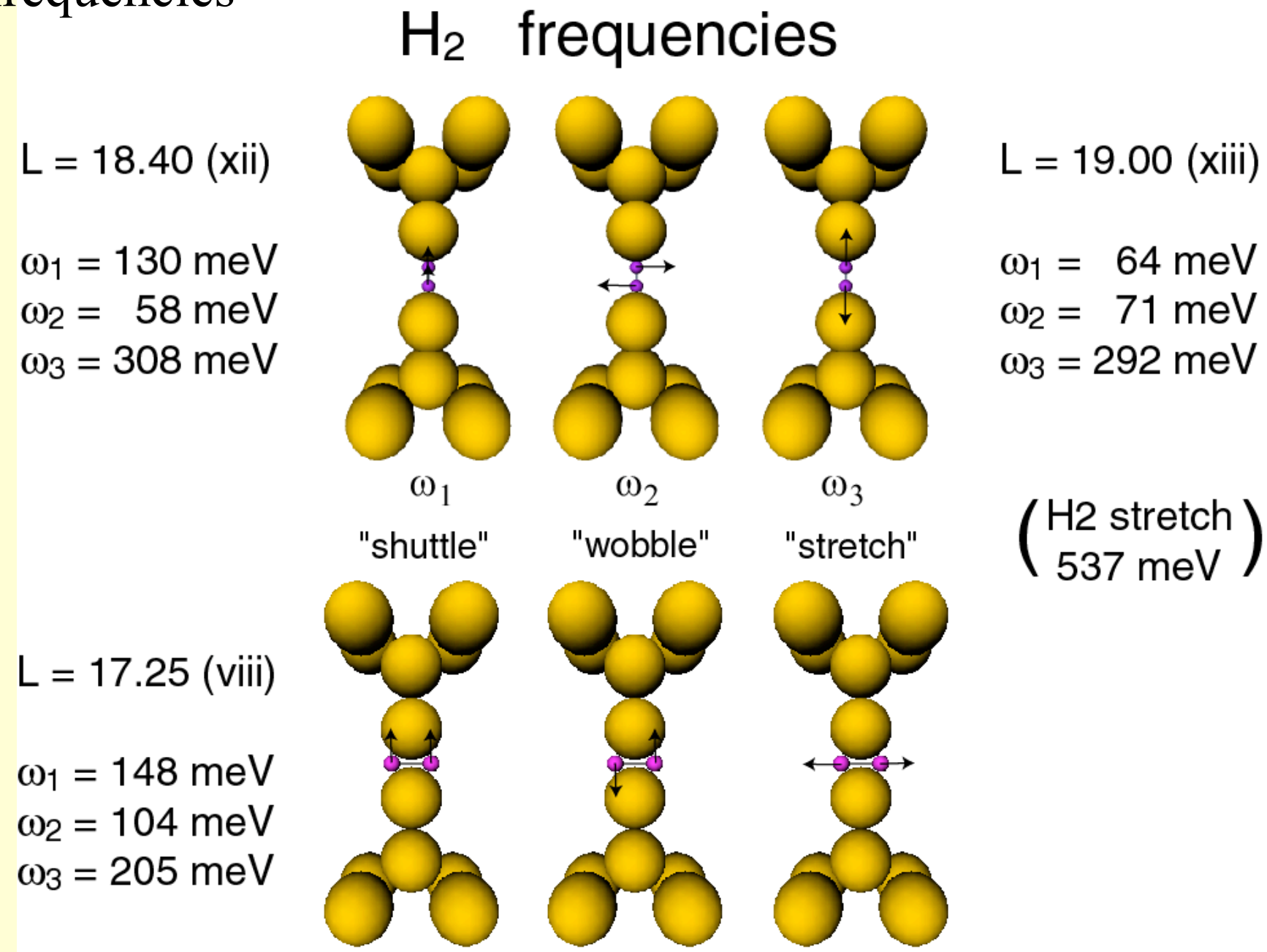
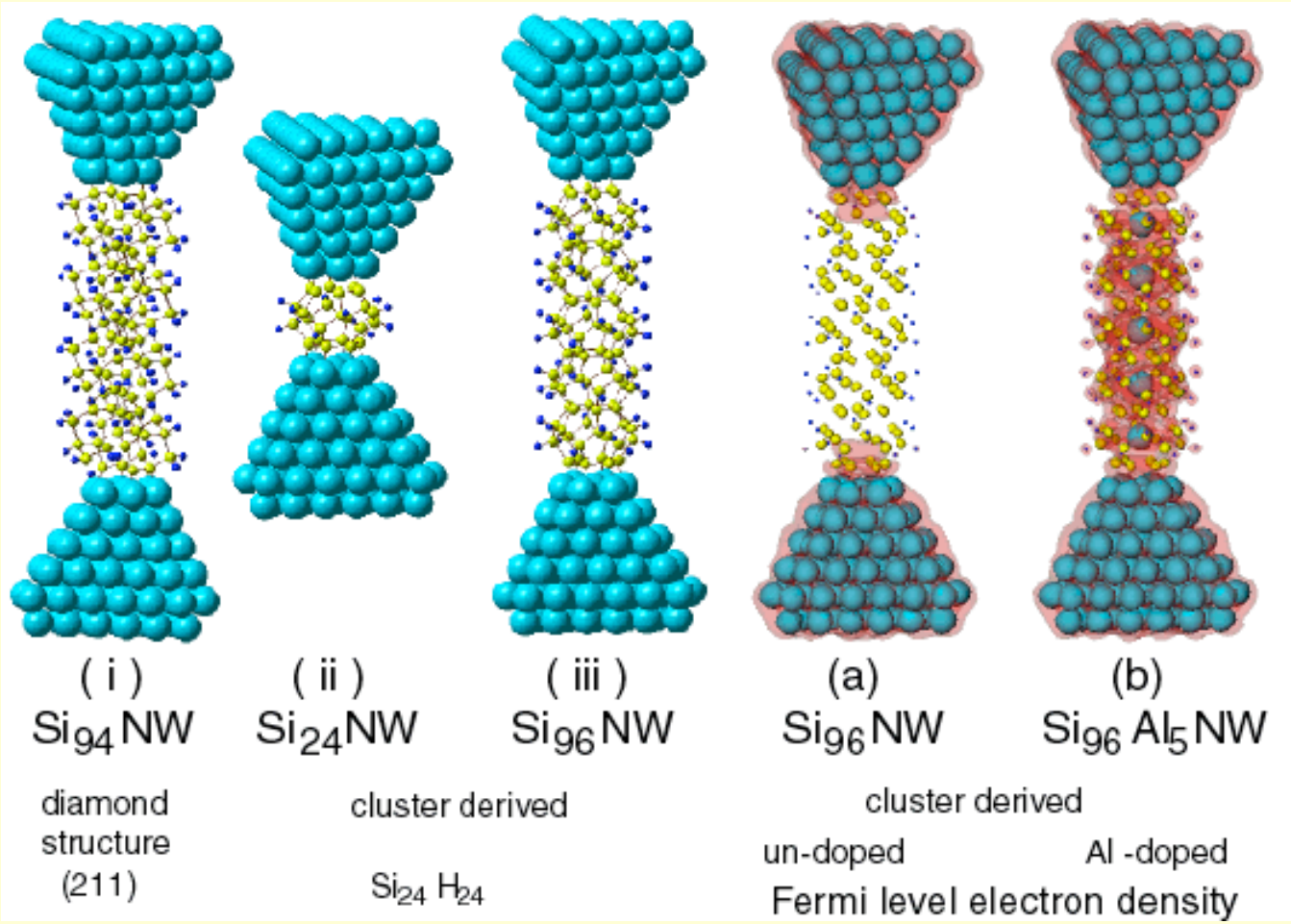
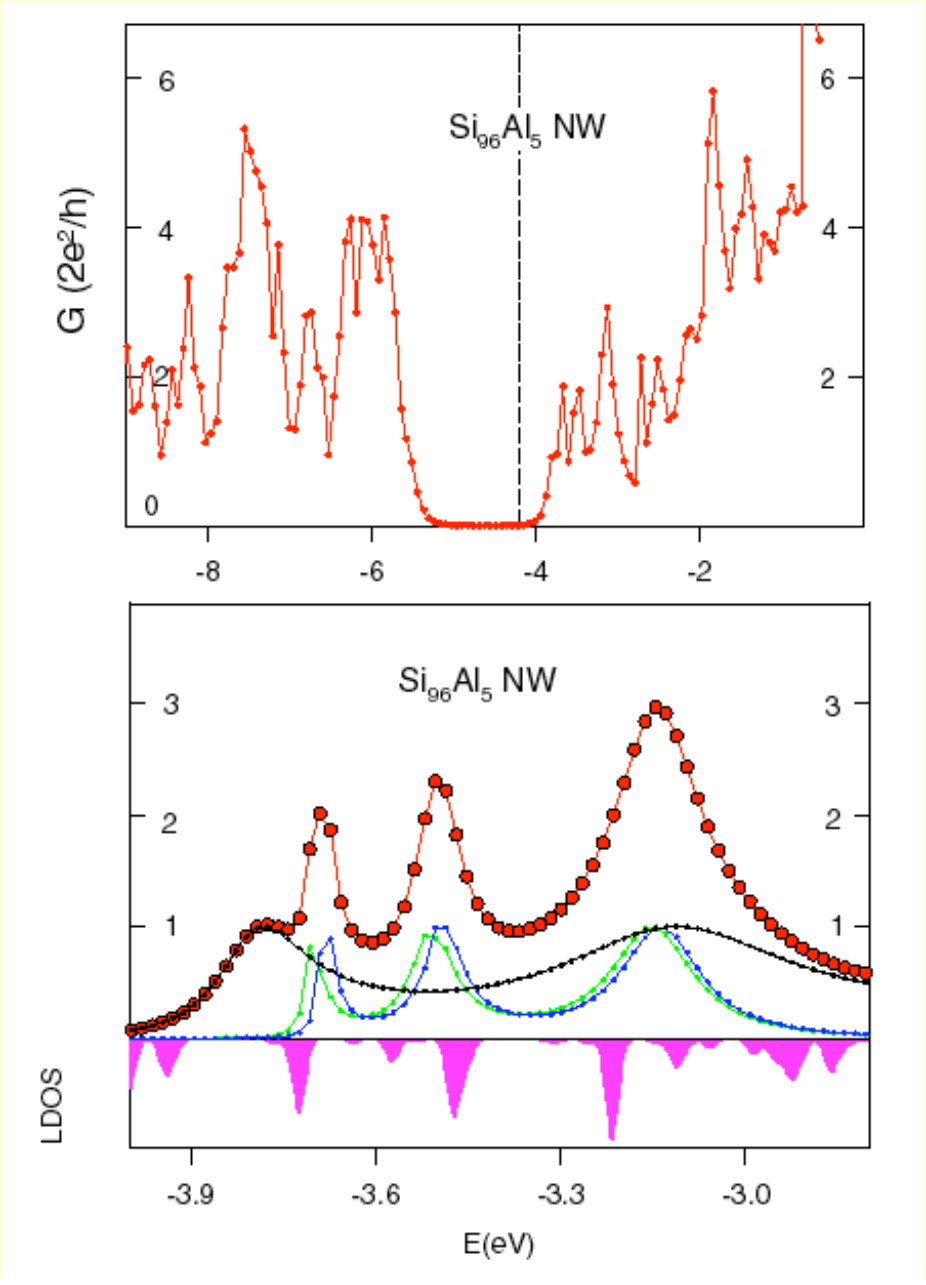


figure 8



Si (yellow) H (dark blue) Al (light blue)

Conductance Spectra



Quantum Monte Carlo study of 2D quantum dots in magnetic fields

D. Güçlü and C. J. Umrigar (Cornell)

W. Geist and M. Y. Chou (Georgia Tech)

- 2D Quantum dots (QDs), also called artificial atoms, can be created by a confinement potential within the quantum well in semiconductor heterostructures.
- Due to the experimental accessibility and control, QDs offer very rich physics which cannot be studied in real atoms. (Coulomb blockade, Kondo effect, quantum computing, *etc.*)
- By applying a magnetic field, it is possible to observe transitions to several many-body ground states with different total angular momentum and spin.

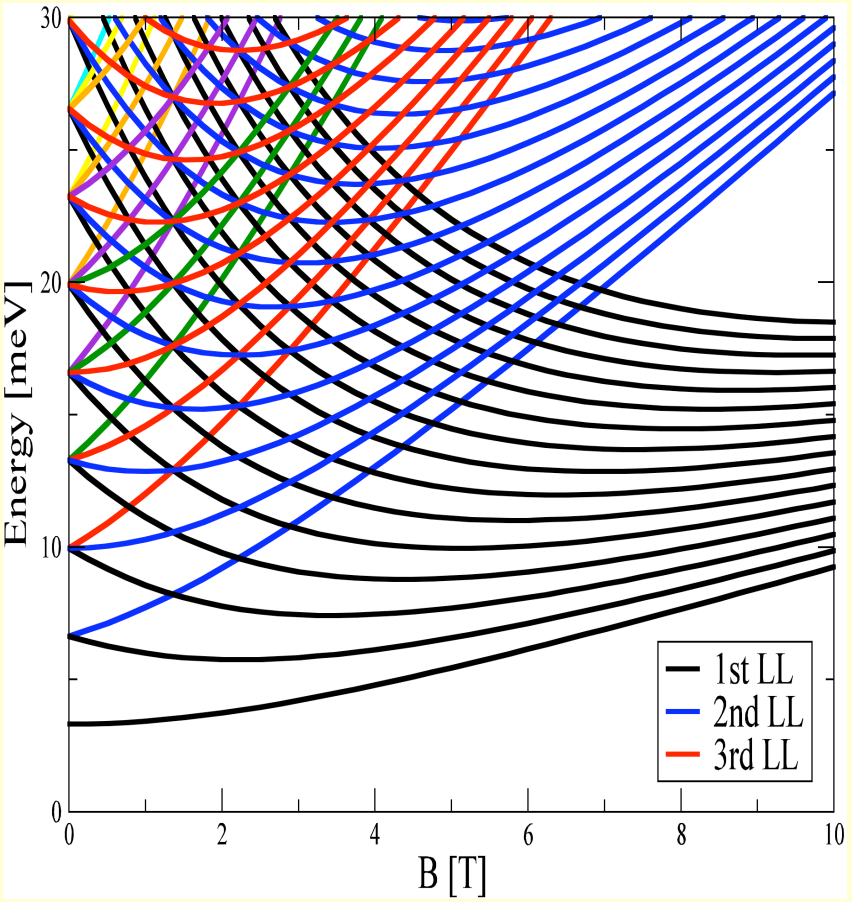
Single electron states

- Non-interacting Hamiltonian:

$$\hat{H}_0 = \frac{1}{2}(\mathbf{p} + \mathbf{A})^2 + \frac{1}{2}\omega_0^2 r^2$$

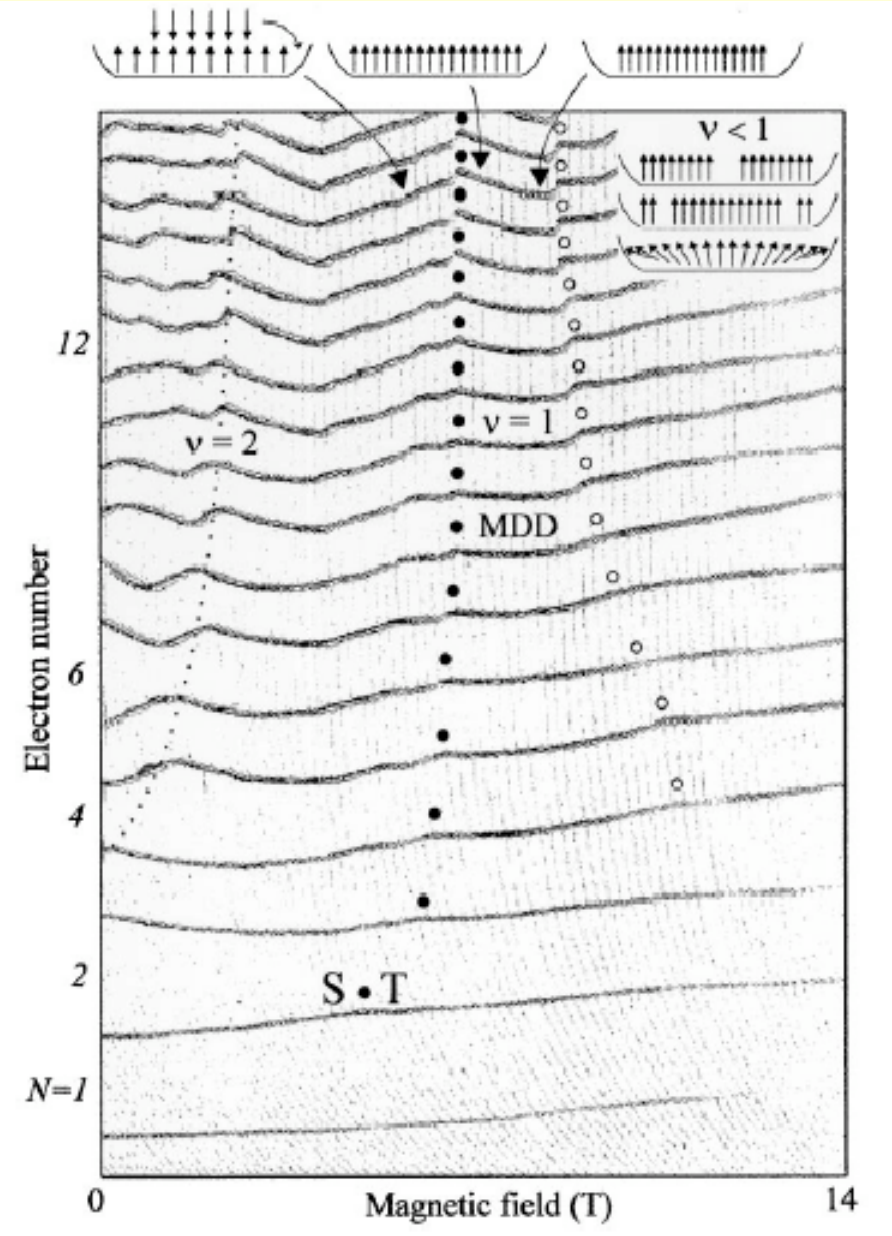
- Fock-Darwin states:

$$\phi_{n,m} \propto e^{-im\theta} r^{|m|} \mathrm{L}_n^{|m|}\left(\frac{r^2}{2}\right) e^{-\frac{r^2}{4}}$$



MDD (maximum-density-droplet) to LDD (lower-density-droplet) Transition

- Physical properties of MDD state ($\nu = 1$ in quantum Hall effect) can be studied by experimental techniques such as *Gated Transport Spectroscopy* Oosterkamp *et al.* PRL 82, 2931 (1999).
- Due to Landau level mixing, theoretical investigation of beyond MDD states (LDD, $\nu < 1$) is difficult, and most of the previous theoretical work is based on lowest-Landau-level approximation.



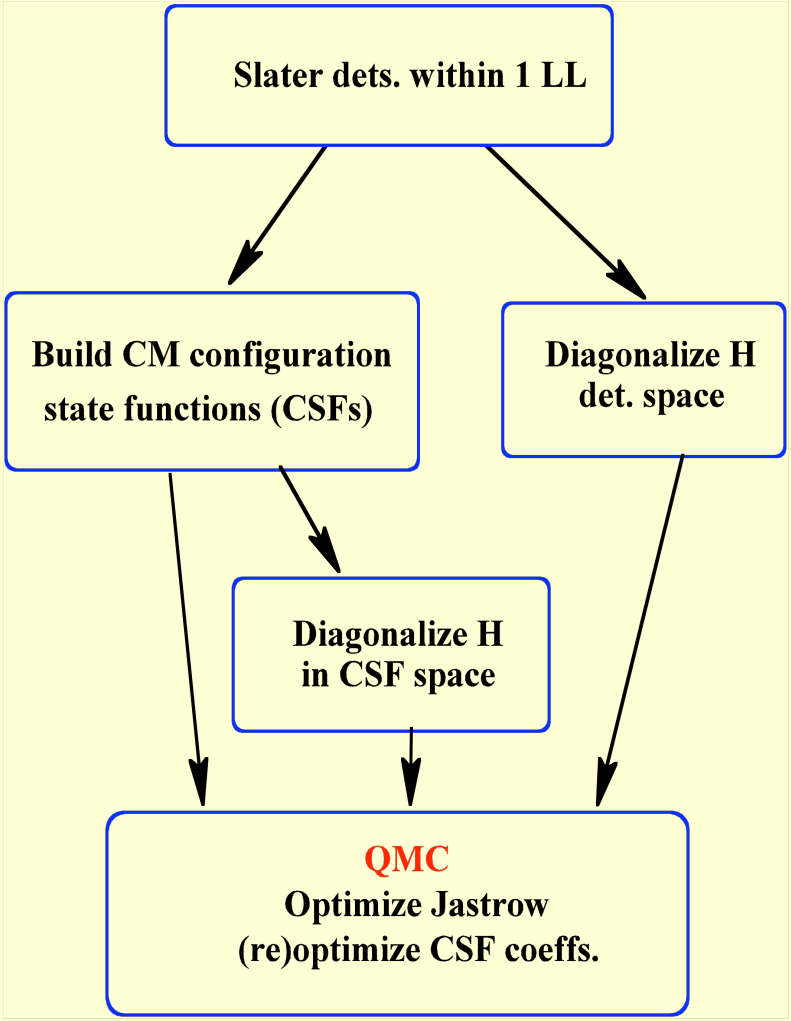
Quantum Monte Carlo

- Jastrow-Slater wavefunctions:

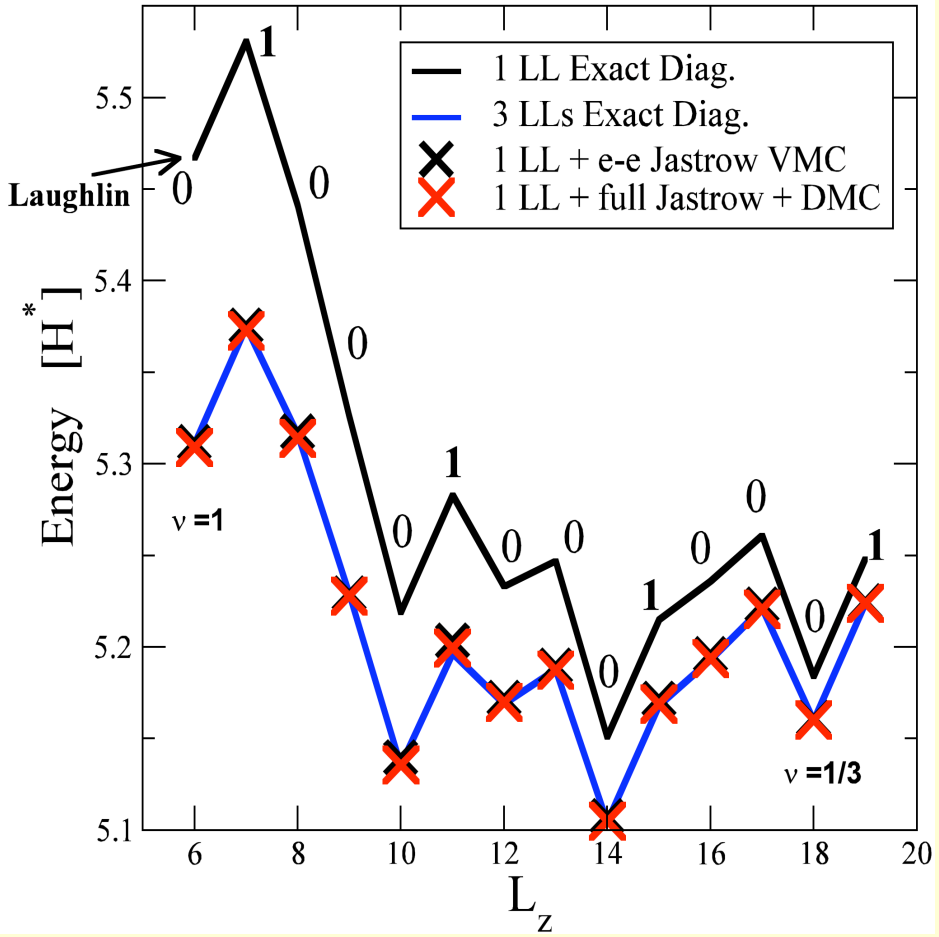
$$\Psi = J_{e-e} J_{e-d} J_{e-e-d} \sum_{1^{st}\,LL} \Psi_{\det}$$

- All 3 approaches give equally good accuracy.

- Determinantal coefficients are independent of system parameters (B, _ ...).



N=4, B=8 T:

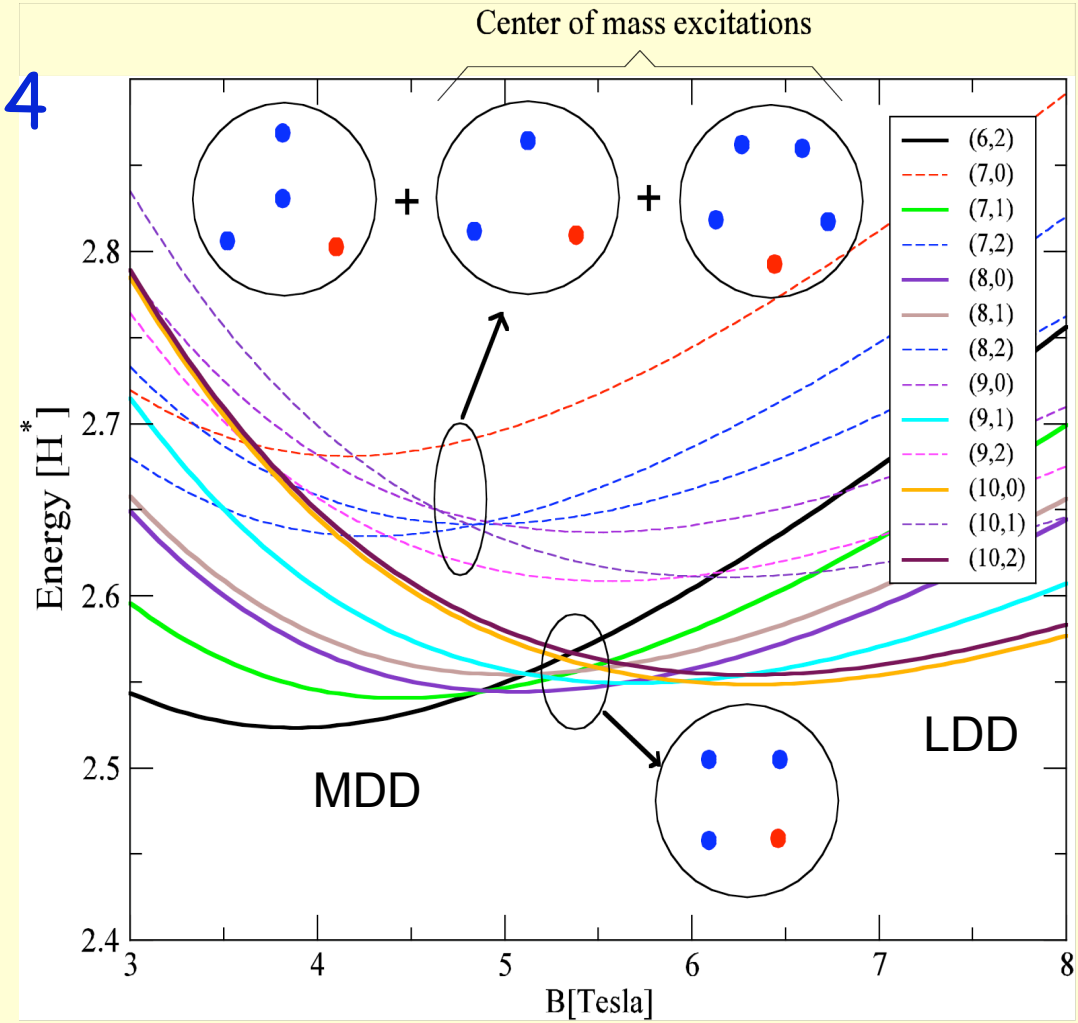


$L_z = 10$	1 LL	2LLs	3LLs
#dets	5	217	1825
#CSFs	2	51	359

- Landau level mixing can be taken into account very accurately and efficiently by multiplying the infinite-field determinants by an optimized Jastrow factor.
- QMC allows us to get extremely accurate results with a very small number of determinants.
- In this spin polarized case, optimization of just the electron-electron Jastrow term allows one to recover almost all the missing energy even in VMC.

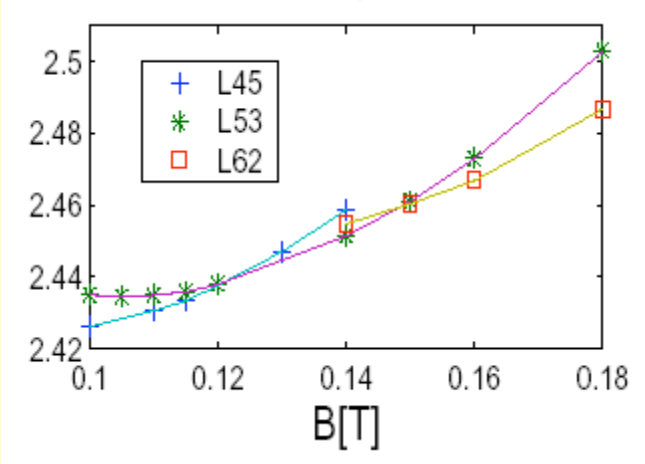
MDD-LDD transition for N=4

- QMC calculations show that MDD-LDD transition has a very rich structure, involving several many-body states characterized by (L,S) in a small “hot region”.
- Strikingly, all the many-body states in the MDD-LDD transition have square symmetry unlike higher energy states.



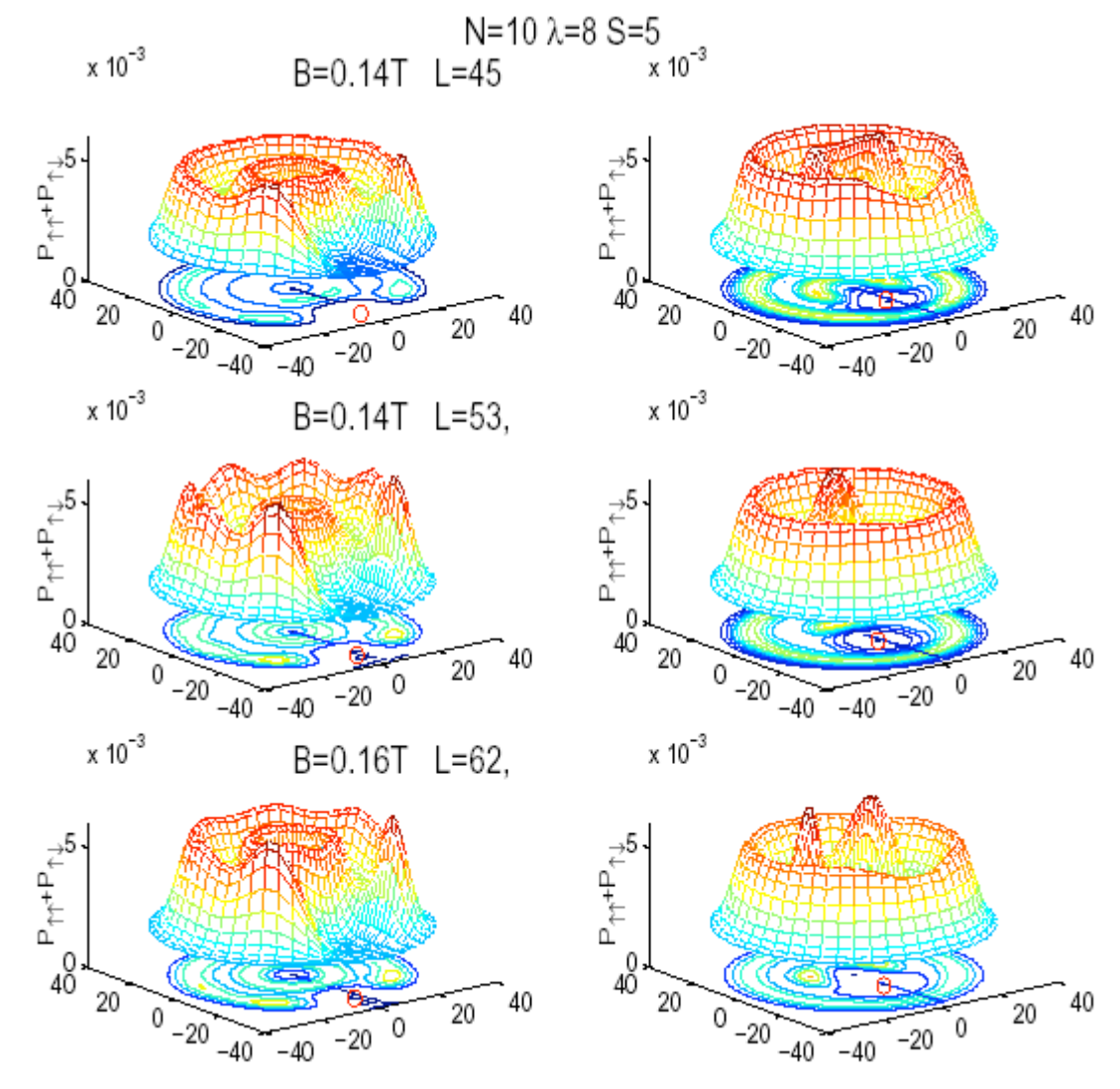
Güçlü *et al*. poster

Energy (H*) for N=10



λ=8 (confinement strength)

Pair correlation function



Education and Outreach

- Train students (undergraduate and graduate) and postdocs in computational techniques for materials simulations
- Involve undergraduate students in materials research through the existing REU program at Georgia Tech
- Partnership between Georgia Tech and Clark Atlanta University (a Historically Black University): regular exchange visits of faculty and students; joint seminars; joint courses; joint workshops
- Information Technology Research Seminars
- Special course “Physics of Small Systems” taught by Landman

- Minority students in the project:
  Alexis Nduwimana (Georgia Tech)
  Damian Cupid (Clark Atlanta)
  Anthony Cochran (Clark Atlanta)
  Carmen Robinson (Clark Atlanta)
  Robert Easley, Jr. (Clark Atlanta)
- Mini-workshop on Quantum Approximate Methods for Novel Materials (Clark Atlanta University, October 2003); all participants are minority students