Transport in nanoscale systems

Antti-Pekka Jauho

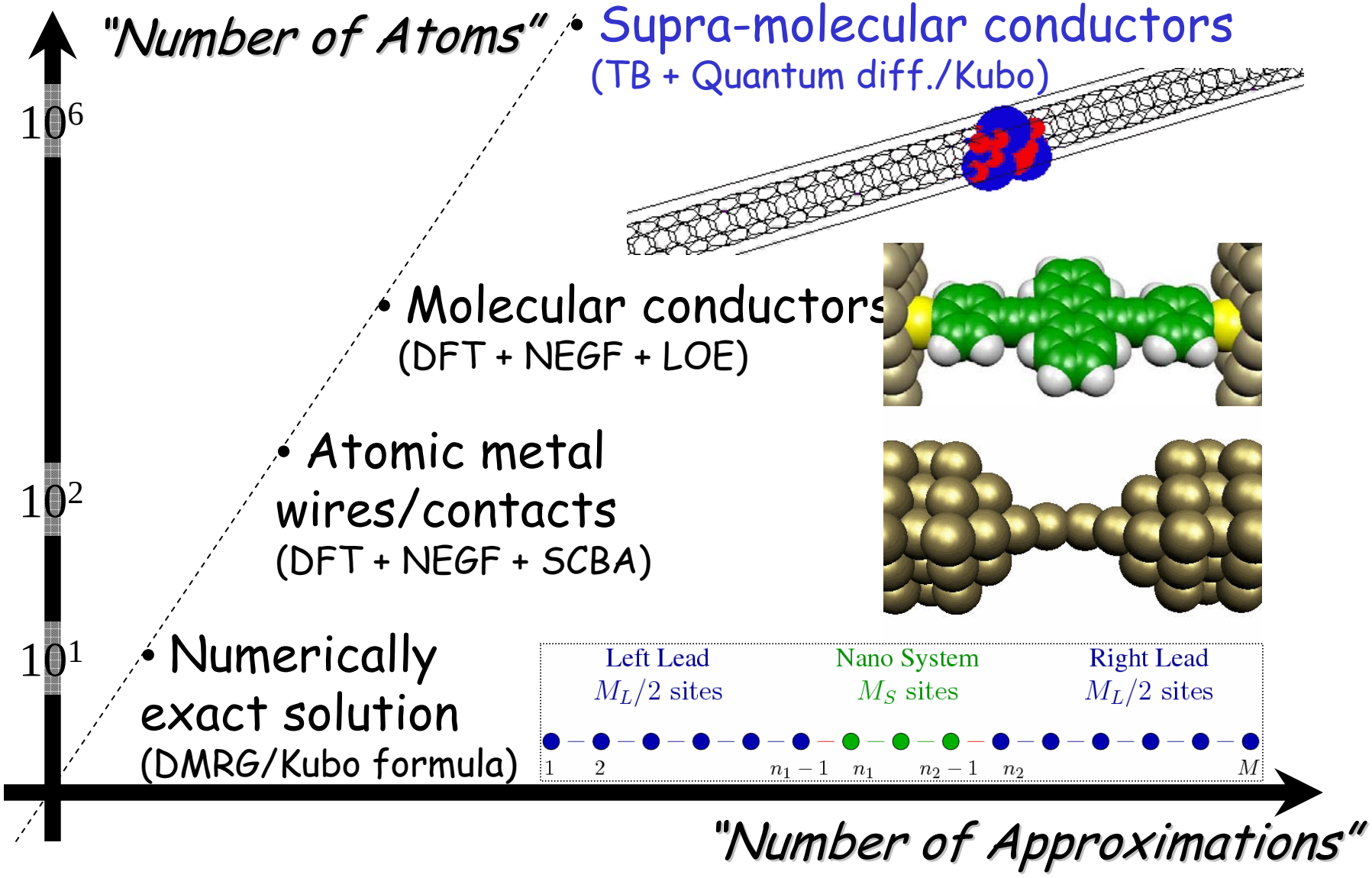
MIC-Department of Micro and Nanotechnology

NanoDTU, Technical University of Denmark

The theoretical scientis attempting to model transport in modern nanoscale systems faces many challenges. The number of atoms requiring a microscopic treatment may vary from a few to several millions. The transport may be coherent, or dominated by interaction effects. No single formalism can capture all the different facets, and in this talk I give a birds-eye-view of the various theoretical tecniques employed in my group.

Regensburg, December 12, 2005

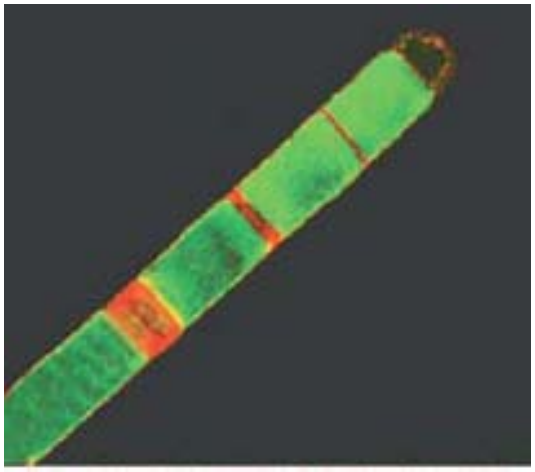
Transport in nanoscale systems



Quantum diffusion and Kubo formula

(Troels Markussen, Mads Brandbyge, APJ)

- Many nanosystems systems contain ~10^5-10^6 atoms
- Imperfect systems: impurities, defects etc.
- First principle methods not usable due to $O(N^3)$ scaling
- Need for approximate methods:
  - Influence of impurities in bulk and on the surface
  - Defects
  - Barcodes



Theory

- Tight-binding description (parameters are evaluated from first principles:

$$\hat{H} = \sum_i \left[\epsilon_i \, | \, i \, \rangle \langle \, i \, | + \sum_{j=n.n.} \left(t_{ij} \, | \, j \, \rangle \langle \, i \, | + t_{ij}^* \, | \, i \, \rangle \langle \, j \, | \right) \right]$$

- Kubo-Greenwood formula:

$$\sigma_{dc}(E) = 2\,\hbar\, e^2\, \pi\, Tr \left[\hat{V}_x \, \delta(E - \hat{H}) \, \hat{V}_x \, \delta(E - \hat{H}) \right]$$

- Following [1] this can be rewritten as:

$$\sigma_{dc}(E) = 2\, e^2\, \pi \lim_{t \to \infty} Tr \left[\frac{1}{t} (\hat{X}(t) - \hat{X}) \delta(E - \hat{H}) \, (\hat{X}(t) - \hat{X}) \right]$$

(the trace is taken over some complete set)

[1]: S. Roche and D. Mayou, Phys. Rev. Lett. **79**, 2519 (1997)

Theory

- Tracing over the 'site' basis, using $\hat{X}|i(0)\rangle = 0$ the conductance for a 1-D system of length L is calculated as:

$$G(E,L) = \frac{\sigma_{dc}(E)}{L} = 2\,e^2\,\pi\,n(E)\,\frac{D_E(\tau)}{L}$$

$$D_E(\tau) = \langle X^2(\tau)\rangle_E \frac{1}{\tau} = \frac{\sum_i \langle i(\tau)|\hat{X}\,\delta(E-\hat{H})\,\hat{X}|i(\tau)\rangle}{n(E)} \cdot \frac{1}{\tau}$$

$$L = \frac{\langle i(\tau)|\hat{X}\delta(E-\hat{H})\hat{X}|\,i(\tau)\rangle}{n(E)}$$

- Main task: Time evolution of $|i(t)\rangle = e^{-i\hat{H}t/\hbar}|i(0)\rangle$

Time propagation

- The time-evolution operator, U(t), is expanded in the orthogonal basis of Chebyshev polynomials:

$$U(\hat{H},t)|\psi_0\rangle = \sum_{n=0}^{\infty} c_n(t)\, T_n(\hat{H})\, |\psi_0\rangle = \sum_{n=0}^{\infty} c_n(t)|\Phi_n\rangle$$

- Chebyshev states:

$$\begin{aligned}
|\Phi_0\rangle &= T_0(\hat{H})|\psi_0\rangle = |\psi_0\rangle \\
|\Phi_1\rangle &= T_1(\hat{H})|\psi_0\rangle = \hat{H}|\psi_0\rangle \\
|\Phi_2\rangle &= T_2(\hat{H})|\psi_0\rangle = 2\hat{H}T_1(\hat{H})|\psi_0\rangle - T_0(\hat{H})|\psi_0\rangle \\
&= 2\hat{H}|\Phi_1\rangle - |\Phi_0\rangle \\
&\vdots \\
|\Phi_{n+1}\rangle &= T_{n+1}(\hat{H})|\psi_0\rangle = 2\hat{H}|\Phi_n\rangle - |\Phi_{n-1}\rangle
\end{aligned}$$

Convergence

- The coefficients are:

$$c_n(t) = \int_{\mathbb{R}} e^{-ixt}\, T_n(x)\, w(x)\, \mathrm{d}x \qquad w(t) = \frac{1}{\sqrt{1-t^2}}, \quad t \in [-1,1]$$
$$= 2\, i^n\, J_n\left(\frac{W\, t}{2\hbar}\right) e^{i\, a\, t/\hbar}$$

$$W = E_{max} - E_{min}\,, \qquad a = (E_{max} + E_{min})/2.$$

- For large n the Bessel function behaves as

$$J_n(x) \approx \frac{1}{\Gamma(n+1)} \left(\frac{x}{2}\right)^n$$

- For sufficiently large n the coefficients thus tend to zero :

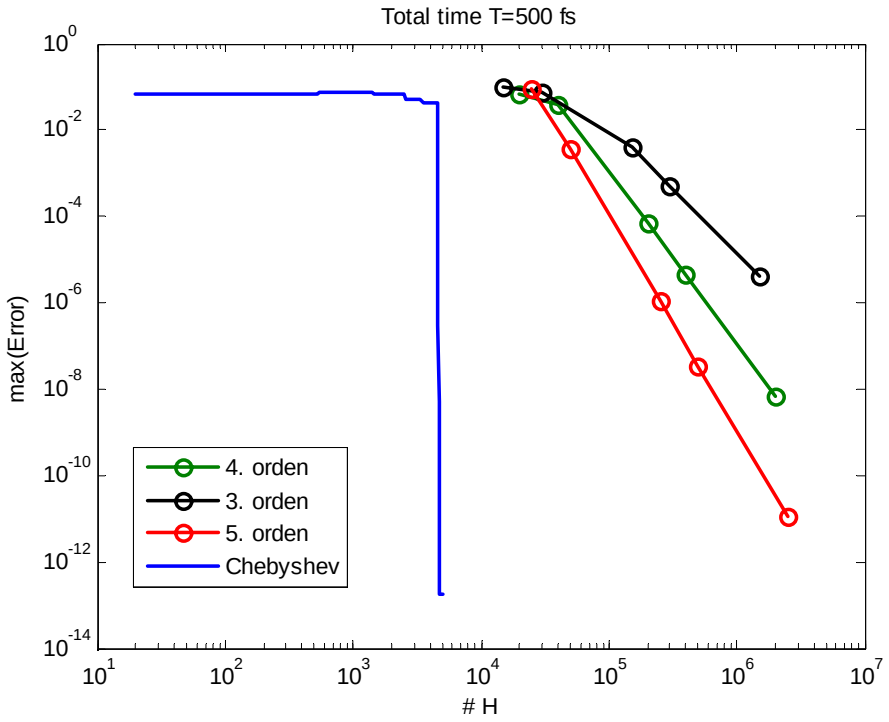
$$|c_n(t)| \propto \left(\frac{eWt}{4\hbar n}\right)^n \to 0, \quad for \quad n \to \infty$$

One-dimensional Chain

- Hamiltonian $H = \sum_{i}^{M} t\left(|i\rangle\langle i-1| + |i\rangle\langle i+1|\right)$
- Comparison between Taylor and Chebyshev

$$Error = \max\{||\psi\rangle_i - |\tilde{\psi}\rangle_i|\}$$

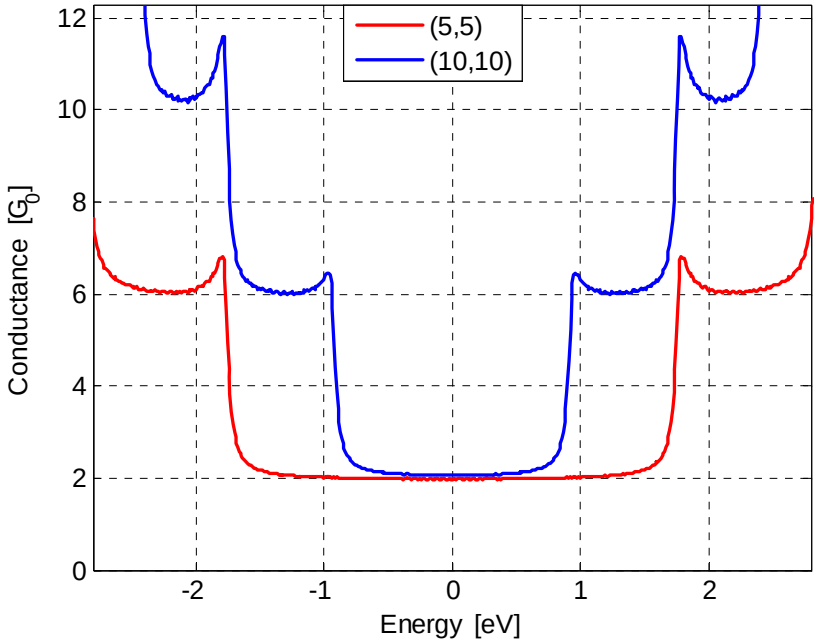
- Data:
  - t=-3 eV
  - M=2000
  - T=500 fs

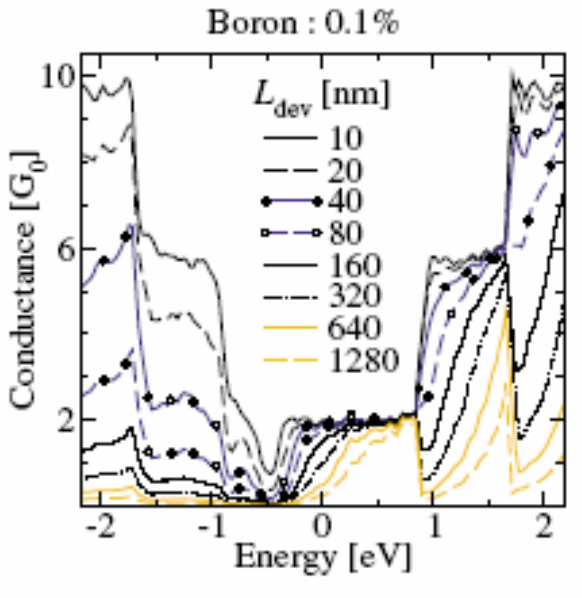


Results: Conductance of a CNT

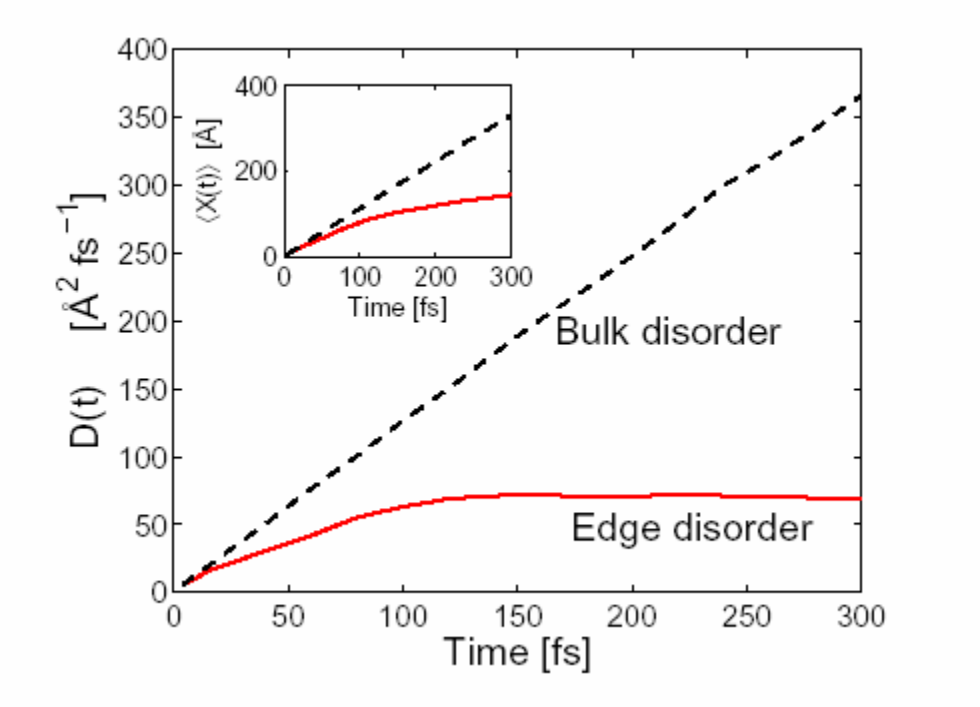
- Perfect CNT – conductance is independent of length

$$G(E,L) = \frac{\sigma_{dc}(E)}{L} = 2\,e^2\,\pi\,n(E)\,\frac{D_E(\tau)}{L}$$





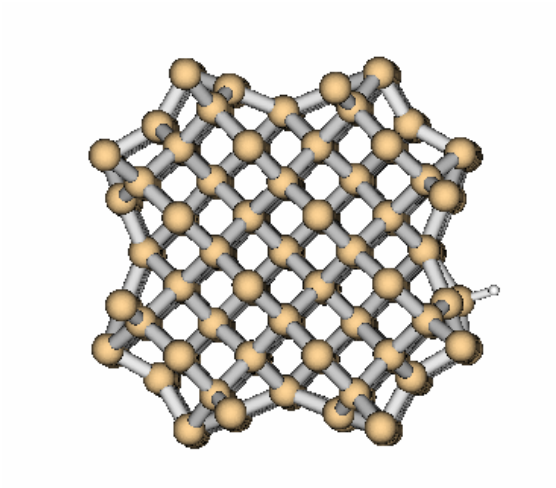
Results: Disordered Si-nanowire

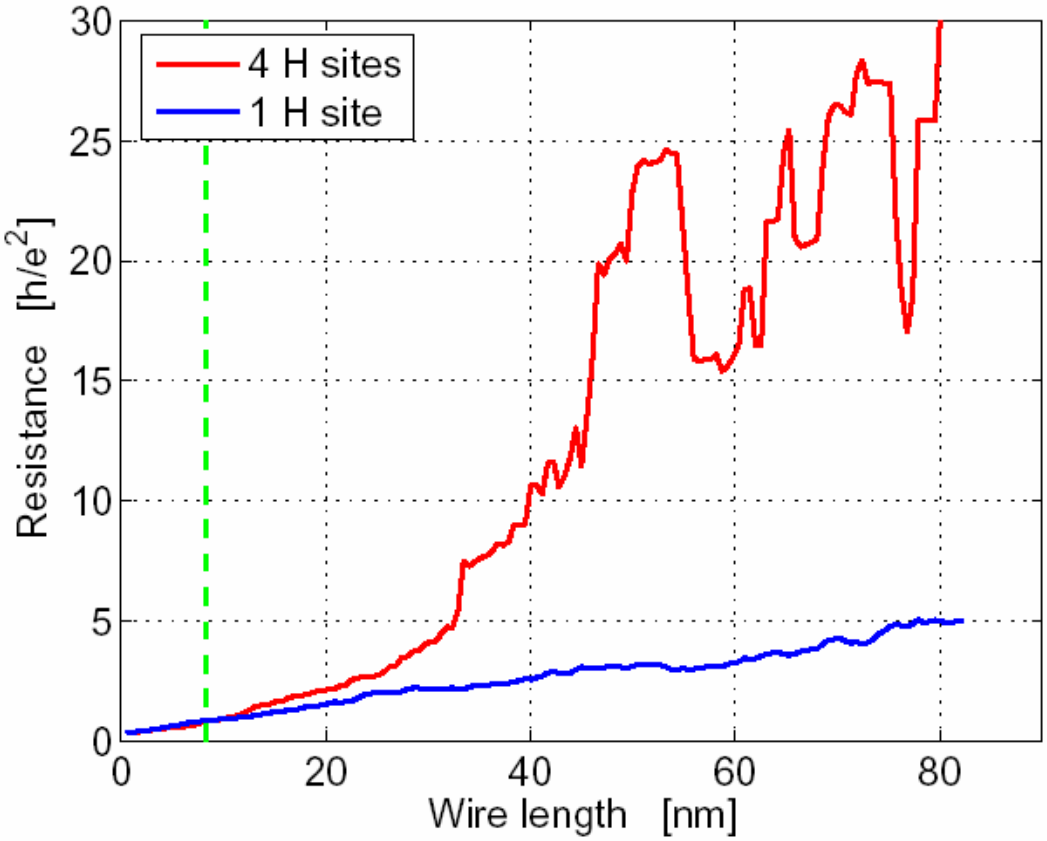


Bulk disorder: little effect (ballistic transport)

Edge disorder: Ohmic behavior

Results: Si-nanowire with H-adatoms

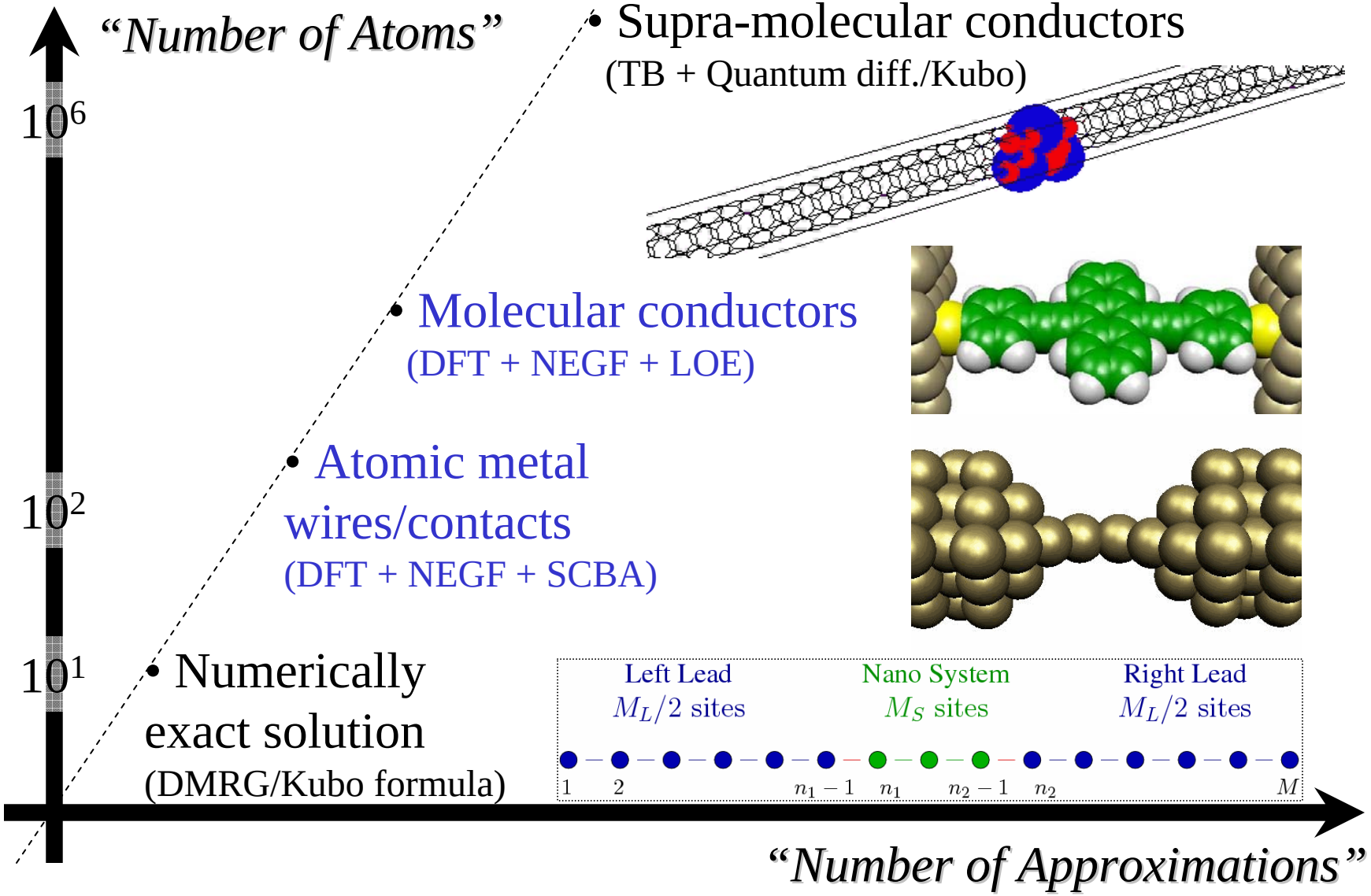




Blue curve: Ohmic behavior

Red curve: Localization

Molecular electronics/transport



Modeling of inelastic transport in molecular electronics from first principles; The DFT-NEGF paradigm

(Thomas Frederiksen, Mads Brandbyge, Magnus Paulsson, APJ)

In recent years several groups have proposed computational algorithms which combine the density-functional theory (DFT),and nonequilibrium Green functions (NEGF). We present the basic ideas, and their inherent limitations, and discuss several applications, and relevance to experimental work. An important very recent extension concerns the inclusion of inelastic effects in to the DFT-NEGF paradigm. Inelastic effects are crucial in many practical applications, for example in understanding the stability and heating occuring in nanoelectronic components. We describe our recent calculations on atomic scale nanowires, for which the theoretical results can be compared to high-quality experiments.

Plan for this subsection

- Review of the basic equations and their limitations
- Uses and misuses of the Meir-Wingreen formula
- Density functional method for nonequilibrium electron transport
- Inelastic scattering and local heating in atomic gold wires
- Can it be done cheaper - LOE

1. The basic equations

Philosophy:

- small system coupled to ideal, large contacts
- in infinite past, the subsystems are separated with respective chemical potentials (Caroli et al. 1970)
- the couplings are turned on adiabatically
- Keldysh contour enters because final state is not known
- nonperturbative, self-consistent calculation of the system parameters may be necessary
- This is not unique – can think of other ways of approaching the problem

1. The basic equations (examples of Hamiltonians)

$$H = \sum_{k,\alpha\in L/R} \epsilon_{k,\alpha} c^{\dagger}_{k,\alpha} c_{k,\alpha} + \sum_{k,\alpha\in L/R;n} \left[V_{k\alpha;n} c^{\dagger}_{k,\alpha} d_n + \text{h.c.} \right] + H_{\text{cen}} \left[\{d_n\}, \{d^{\dagger}_n\} \right]$$

$$H_{\text{cen}} = \sum_n \epsilon_n d^{\dagger}_n d_n + H_{\text{int}}$$

$$H^{\text{el-ph}}_{\text{int}} = \sum_{m\sigma} d^{\dagger}_{m,\sigma} d_{m,\sigma} \sum_{\mathbf{q}} M_{m,\mathbf{q}} \left[a^{\dagger}_{\mathbf{q}} + a_{\mathbf{q}} \right]$$

$$H^{A}_{\text{int}} = U \sum_m d^{\dagger}_{m,\uparrow} d_{m,\uparrow} d^{\dagger}_{m,\downarrow} d_{m,\downarrow}$$

1. The basic equations (calculation of current)

The current operator:

$$I_L = -\frac{ie}{\hbar}\sum_{k,n}\left[-V_{kL;n}c_{kL}^{\dagger}d_n + V_{kL;n}^{*}d_n^{\dagger}c_{kL}\right]$$

Current leaving the left contact (requires **noninteracting (mean-field)** contacts):

$$\langle I_L\rangle = J_L(t) = -\frac{2e}{\hbar}\int_{-\infty}^{t}dt_1\int\frac{d\epsilon}{2\pi}\mathrm{Im}\mathrm{Tr}\Big\{e^{-i\epsilon(t_1-t)}\mathbf{\Gamma}^{L}(\epsilon,t_1,t) \times\left[\mathbf{G}^{<}(t,t_1)+f_L^0(\epsilon)\mathbf{G}^{r}(t,t_1)\right]\Big\}.$$

Static limit ➥ Meir-Wingreen formula

$$J = \frac{ie}{2\hbar}\int\frac{d\epsilon}{2\pi}\mathrm{Tr}\Big\{\left[\mathbf{\Gamma}^{L}(\epsilon)-\mathbf{\Gamma}^{R}(\epsilon)\right]\mathbf{G}^{<}(\epsilon) + \left[f_L^0(\epsilon)\mathbf{\Gamma}^{L}(\epsilon)-f_R^0(\epsilon)\mathbf{\Gamma}^{R}(\epsilon)\right]\left[\mathbf{G}^{r}(\epsilon)-\mathbf{G}^{a}(\epsilon)\right]\Big\}$$

1. The basic equations (comments)

- this is just a paradigm – one still needs to evaluate (in one way or another!)

$$G^{<}_{nm}(t,t_1) = i\langle d^{\dagger}_m(t_1) d_n(t)\rangle$$
$$G^{r}_{nm}(t,t_1) = -i\theta(t-t_1)\langle [d_n(t), d^{\dagger}_m(t_1)]\rangle$$

- displacement currents **not** included ➧ require separate treatment
- interpretation – write MW formula in an alternative form:

$$J_L = \frac{e}{h}\int d\epsilon \mathrm{Tr}\left\{\mathbf{\Sigma}^{L,<}(\epsilon)\mathbf{G}^{>}(\epsilon) - \mathbf{\Sigma}^{L,>}(\epsilon)\mathbf{G}^{<}(\epsilon)\right\}$$

- first term: current from left contact to central region
- second term: current from central region to left contact

1. The basic equations (comments cont'd)

- In general, one needs to solve the coupled Keldysh-Dyson equations:

$$G^{<} = G^{r}\Sigma^{<}G^{a},$$
$$G^{r} = G_{0}^{r} + G_{0}^{r}\Sigma^{r}G^{r}$$

Limitations:

- physical criteria for selecting what is contact, and what is central region (i.e, where are the interactions allowed to operate)?
- charge neutrality of the system?
- energy relaxation in the noninteracting leads (no such Hamiltonians present in the formulation)?
- adiabatic turning-on of the contacts – not usually realized in experiments! (alternative formulation has been given by Stefanucci et al. PRB 2004), following early work of Cini – too early to draw strong conclusions)

2. Uses and misuses of the Meir-Wingreen formula

(i) Mean-field theory (DFT):

$$J_L = \frac{e}{h} \int d\epsilon T_{\rm tot}(\epsilon)[f_L^0(\epsilon) - f_R^0(\epsilon)],$$

$$T_{\rm tot}(\epsilon) = {\rm Tr}\left\{\mathbf{\Gamma}^L(\epsilon)\mathbf{G}^r(\epsilon)\mathbf{\Gamma}^R(\epsilon)\mathbf{G}^a(\epsilon)\right\}$$

- an excellent formula (see, e.g., Datta's book) – but not the whole truth
- forms the basics for a huge number of calculations, even on industrial level

2. Uses and misuses of the Meir-Wingreen formula (cont'd)

(ii) Conservation laws: write

$$\mathbf{\Sigma}_{\rm tot} = \mathbf{\Sigma}_{\rm int} + \sum_{\alpha \in L/R} \mathbf{\Sigma}^{\alpha}$$

Then (using Keldysh equation)

$$\mathrm{Tr}\left\{\mathbf{\Sigma}^{<}_{\rm tot}\mathbf{G}^{>} - \mathbf{\Sigma}^{>}_{\rm tot}\mathbf{G}^{<}\right\} \equiv 0$$

and current conservation $\sum_{\alpha} J^{\alpha} = 0$ leads to

$$\int d\epsilon \mathrm{Tr}\left\{\mathbf{\Sigma}^{<}_{\rm int}(\epsilon)\mathbf{G}^{>}(\epsilon) - \mathbf{\Sigma}^{>}_{\rm int}(\epsilon)\mathbf{G}^{<}(\epsilon)\right\} = 0$$

which is a useful check on numerics (N.B. This formula is familiar for people working with kinetic theory: integrated collision term must vanish!)

2. Uses and misuses of the Meir-Wingreen formula (cont'd)

(ii) Analytics in the wide-band limit.

$$J = \frac{e}{h}\frac{\Gamma^L\Gamma^R}{\Gamma^R+\Gamma^L}\int d\epsilon[f_L(\epsilon)-f_R(\epsilon)]A(\epsilon) \qquad A(\epsilon) = -2\mathrm{Im}G^r(\epsilon)$$

In atomic limit, the GF can be computed for several important cases.

A single level coupled to phonons:

$$G^r(t) = -i\theta(t)\exp[-it(\epsilon_0-\Delta)-\Phi(t)] \qquad \Delta = \sum\frac{M_q^2}{\omega_q}$$

$$\Phi(t) = \sum_q \frac{M_q^2}{\omega_q^2}[N_q(1-e^{i\omega_q t})+N_q(1-e^{-i\omega_q t})]$$

An isolated Anderson impurity:

$$G^\sigma(\epsilon) = \frac{\langle n_{\bar\sigma}\rangle}{\epsilon-\epsilon_\sigma-U}+\frac{1-\langle n_{\bar\sigma}\rangle}{\epsilon-\epsilon_\sigma}$$

Humble advice: do not broaden these GF's by a phenomenological width (in a well-meant but ill-conceived attempt to simulate coupling to leads): lots of interesting physics is inadvertently lost!

3. DFT for nonequilibrium electron transport

- Standard methods for electronic structure calculations assume a finite, or a periodic geometry
- Standard methods assume that the electronic system is in equilibrium
- Molecular electronics requires something entirely different:

  (a) small, translationally noninvariant subsystem

  (b) coupling to semi-infinite leads

  (c) nonequilibrium state in the subsystem

To use DFT in nonequilibrium, one must assume that the Kohn-Sham orbitals can be used to calculate the current. (Possible extensions: use TDDFT, or current-density-functional theory)

If this approach is OK, one can use NEGF to construct a nonequlibrium electron density, which can be fed back in the DFT loop. The price is an extra iterative subloop, but the convergence can be tested, and improved, by appropriate choice of exchange-correlation functional, and/or basis set.

3. DFT for nonequilibrium electron transport (summary of TranSIESTA method)

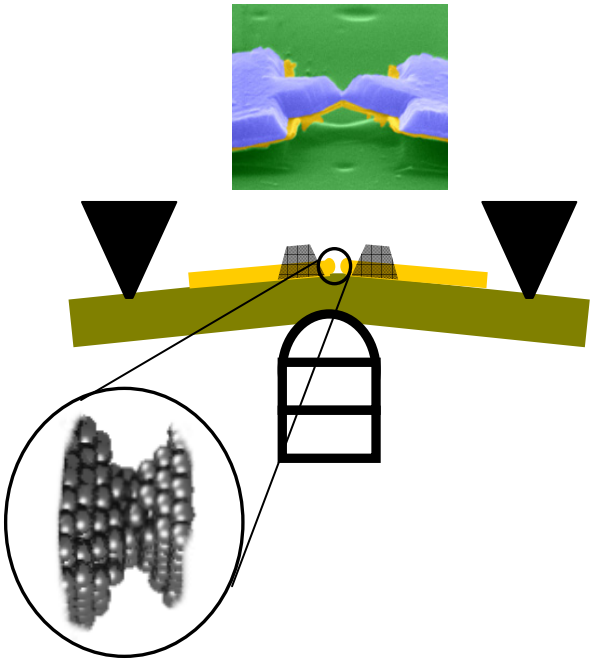
$$n(x) = -iG^{<}(x = x', t = t') = \int \frac{d\epsilon}{2\pi i} G^{<}(x = x', \epsilon)$$

$$G^{<} = G^{r}\Sigma^{<}G^{a} \qquad \Sigma^{<} = i(\Gamma^{L} f_{L} + \Gamma^{R} f_{R})$$

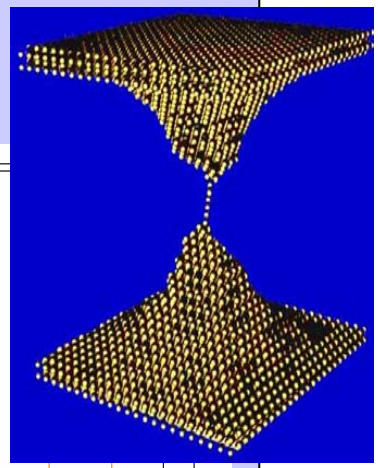
$$\mathbf{G}^{r}(E) = [E\mathbf{I} + i\eta - \mathbf{H}]^{-1} \qquad \mathbf{H} = \begin{pmatrix} \mathbf{H}_L + \mathbf{\Sigma}_L & \mathbf{V}_L & 0 \\ \mathbf{V}_L^{\dagger} & \mathbf{H}_C & \mathbf{V}_R \\ 0 & \mathbf{V}_R^{\dagger} & \mathbf{H}_R + \mathbf{\Sigma}_R \end{pmatrix}$$

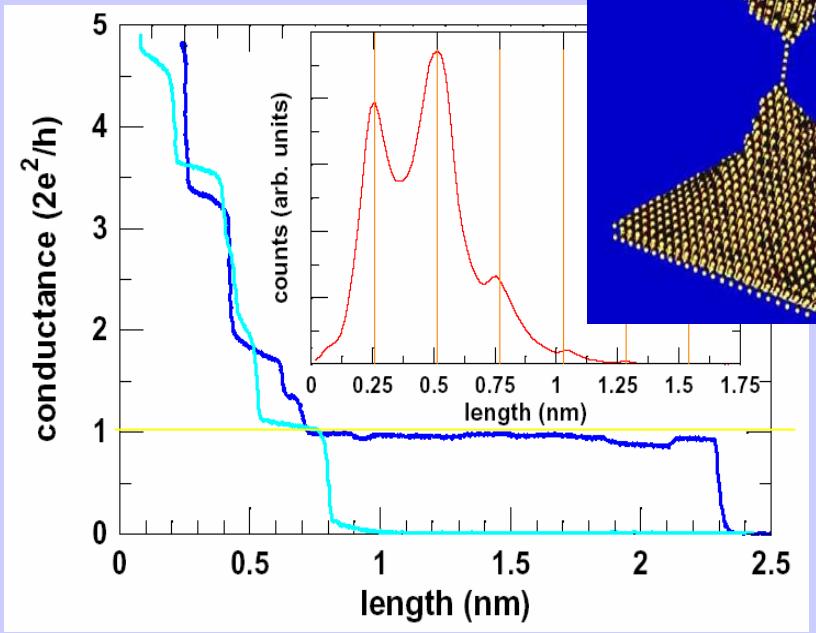
$$\text{initial}\, n(x) \Rightarrow \text{SIESTA} \Rightarrow \psi_{\text{KS}}(x) \Rightarrow \text{NEGF} \Rightarrow \text{new}\, n(x)$$

4. Inelastic scattering and local heating in atomic gold wires



- Pulling an atomic Au wire





A. Yanson *et al.*, Nature 395, 783 (1998)

C. Untiedt *et al.* Phys. Rev. B **66**, 085418 (2002)

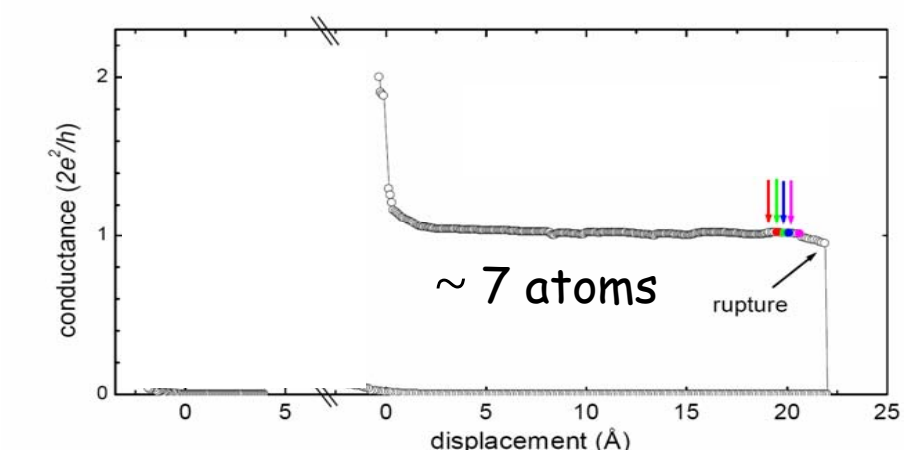
Inelastic phonon signal

VOLUME 88, NUMBER 21 PHYSICAL REVIEW LETTERS 27 MAY 2002

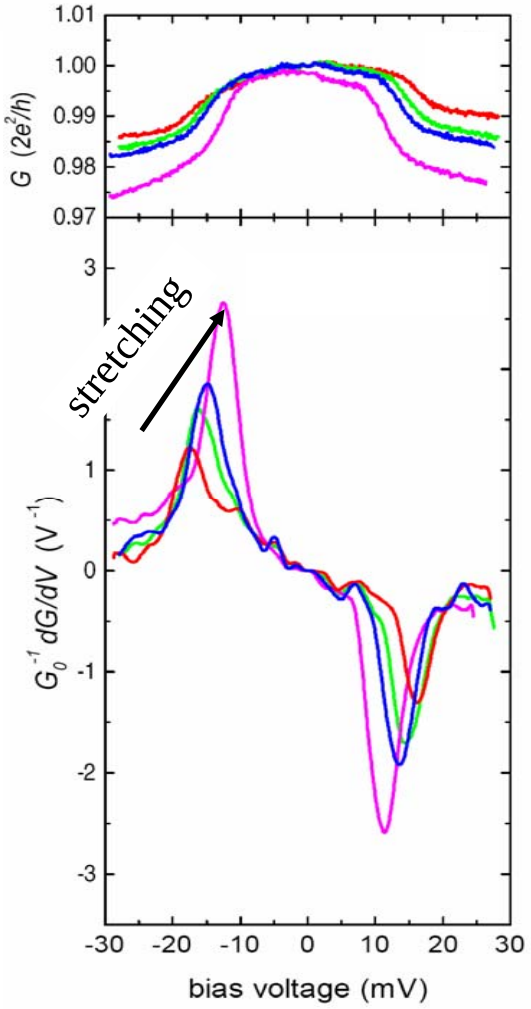
Onset of Energy Dissipation in Ballistic Atomic Wires

Nicolás Agraït,* Carlos Untiedt,† Gabino Rubio-Bollinger, and Sebastián Vieira

Gold single-atom wire



$T = 4\ K$

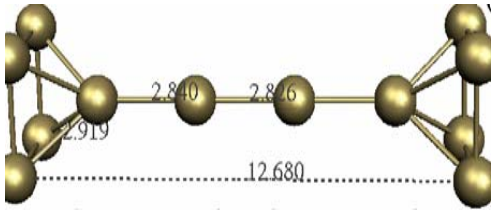


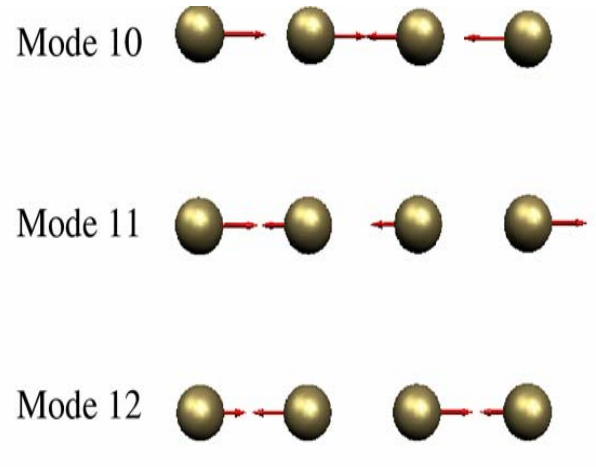
- Mode selective (only one main peak seen)
- Conductance drop of 1-1.5% dep. on length
- Drop increase with stretch
- Streching 1Å gives 7meV frequency shift

Phonon interaction

- Born-Oppenheimer approximation
- Harmonic approximation
- Free motion of HO's

$$\mathbf{H}_{\text{int}} = \mathbf{H}_{\text{e}}(\{d_n^\dagger\};\{d_n\}) + \sum_\lambda \Omega_\lambda b_\lambda^\dagger b_\lambda + \sum_{n,n'} \mathbf{M}_{n,n'}^\lambda d_n^\dagger d_{n'}(b_\lambda^\dagger + b_\lambda)$$





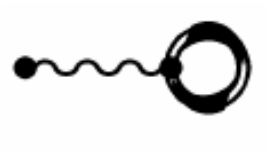
9.4 meV

12.8 meV

13.4 meV

Self-consistent Born approximation

Assuming free propagation for the phonons



$$\Sigma^{H,r}_{SCBA} = 2_{\text{spin}} \sum_\lambda i \frac{2}{\Omega_\lambda} \mathbf{M}_\lambda \int \frac{d\omega}{2\pi} \text{Tr}[\mathbf{G}^<(\omega)\mathbf{M}_\lambda]$$

$$\Sigma^{H,<}_{SCBA} = 0$$

Hilbert transforms

Delta functions

$$\Sigma^{F,r}_{SCBA}(\omega) = i \sum_\lambda \int \frac{d\omega'}{2\pi} \mathbf{M}_\lambda \left[D^r_0(\lambda, \omega-\omega')[\mathbf{G}^<(\omega') + \mathbf{G}^r(\omega')] + D^<_0(\lambda,\omega-\omega')\mathbf{G}^r(\omega') \right] \mathbf{M}_\lambda$$

$$\Sigma^{F,<}_{SCBA}(\omega) = i \sum_\lambda \int \frac{d\omega'}{2\pi} \mathbf{M}_\lambda D^<_0(\lambda,\omega-\omega')\mathbf{G}^<(\omega')\mathbf{M}_\lambda$$

$$= \sum_\lambda \mathbf{M}_\lambda \left[(N_\lambda + 1)\mathbf{G}^<(\omega+\Omega_\lambda) + N_\lambda \mathbf{G}^<(\omega-\Omega_\lambda) \right] \mathbf{M}_\lambda$$

Current conservation?

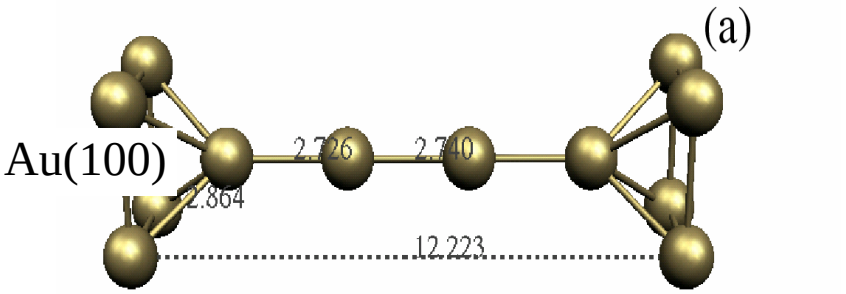
$$
\begin{aligned}
\sum_{\alpha=1}^{M} J^{\alpha} &= \sum_{\alpha} \frac{1}{\hbar} \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} \mathrm{Tr}\left[\mathbf{\Sigma}^{\alpha,<}(\omega)\mathbf{G}^{>}(\omega) - \mathbf{\Sigma}^{\alpha,>}(\omega)\mathbf{G}^{<}(\omega)\right] \\
&= \frac{1}{\hbar} \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} \mathrm{Tr}\left[\{\mathbf{\Sigma}^{<}_{\mathrm{tot}}(\omega) - \mathbf{\Sigma}^{<}_{\mathrm{int}}(\omega)\}\mathbf{G}^{>}(\omega)\right. \\
&\qquad\qquad \left. -\{\mathbf{\Sigma}^{>}_{\mathrm{tot}}(\omega) - \mathbf{\Sigma}^{>}_{\mathrm{int}}(\omega)\}\mathbf{G}^{<}(\omega)\right] \\
&= \frac{1}{\hbar} \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} \mathrm{Tr}\left[-\mathbf{\Sigma}^{<}_{\mathrm{int}}(\omega)\mathbf{G}^{>}(\omega) + \mathbf{\Sigma}^{>}_{\mathrm{int}}(\omega)\mathbf{G}^{<}(\omega)\right] = 0?
\end{aligned}
$$

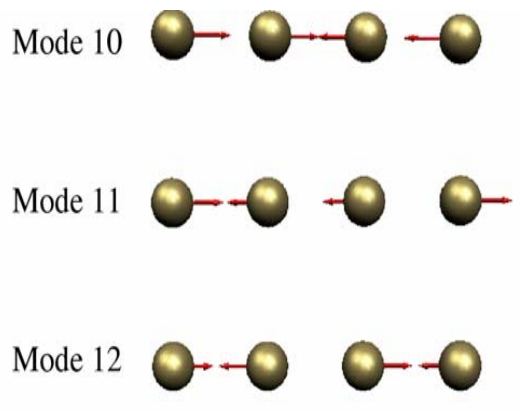
$$
\mathbf{\Sigma}^{\lessgtr}_{\mathrm{SCBA}}(\omega) = i\hbar \sum_{\lambda} \mathbf{M}^{\lambda}\{(N_{\lambda}+1)\mathbf{G}^{\lessgtr}(\omega \pm \Omega_{\lambda}) + N_{\lambda}\mathbf{G}^{\lessgtr}(\omega \mp \Omega_{\lambda})\}\mathbf{M}^{\lambda}
$$

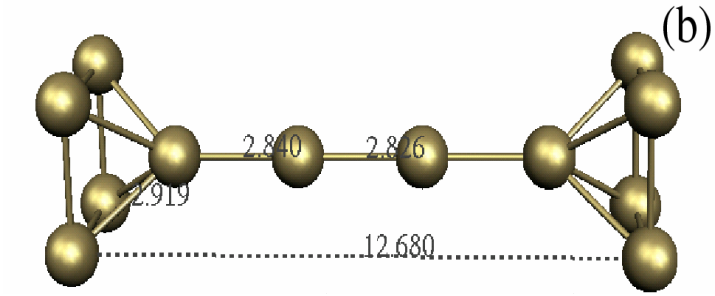
…SCBA conserves current!

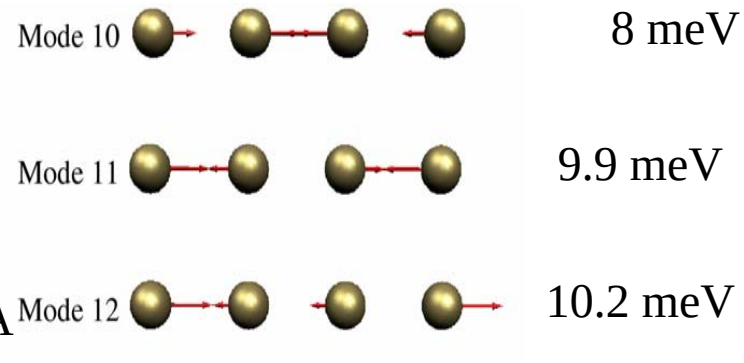
4-atom gold chain

We consider 2 structures and their modes:









✓ Modes/Geometry: Plane-wave basis, GGA

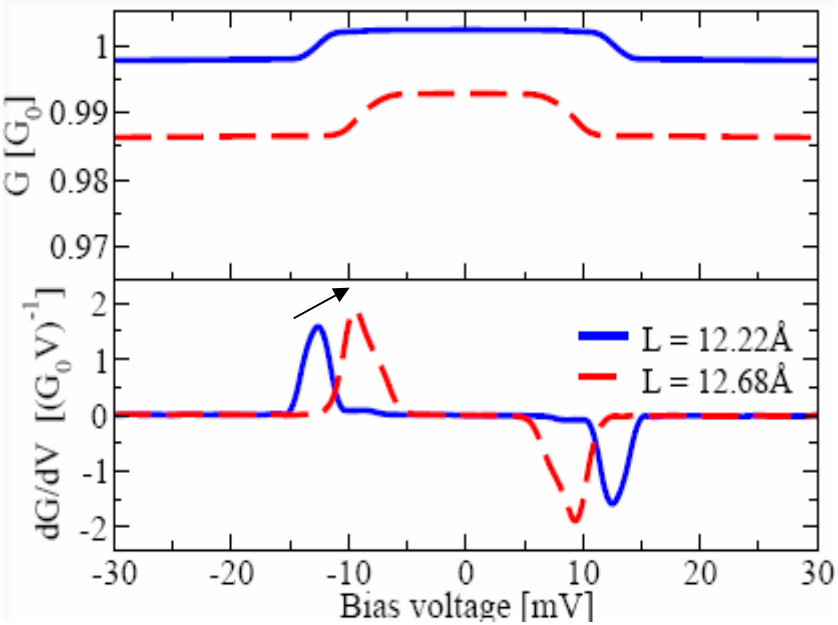
✓ Transport: Transiesta (AO-basis set), Au(100) electrodes

el-ph coupling by finite difference (Head-Gordon & Tully, JCP 96, 3939 (1992))

✓ e-ph self-energy: SCBA

$N_{ph} = n_B(4K) \sim 0$

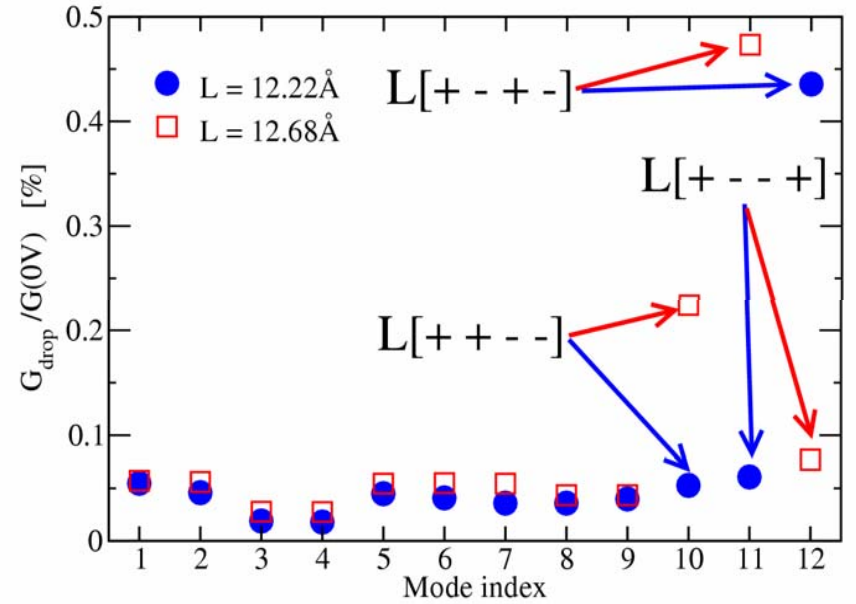
Assume damping by some external reservoir: $N_{ph} = n_B(4K) \sim 0$

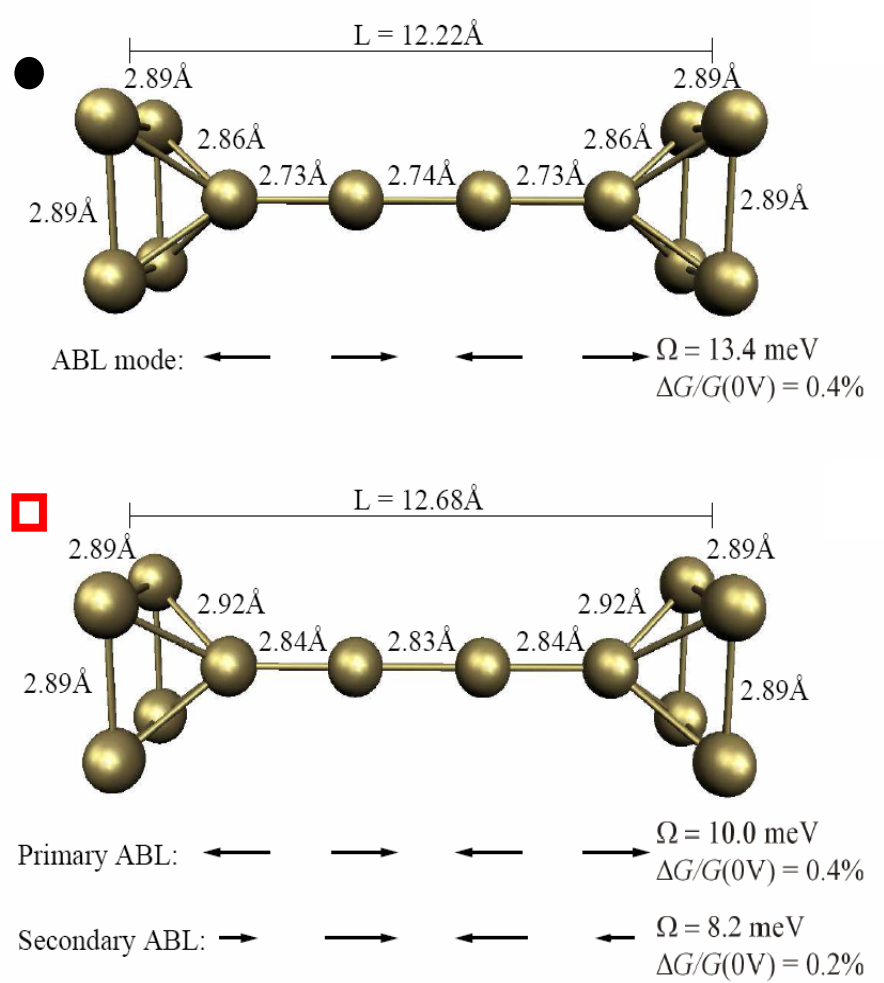


- All modes included in self-energy
- Few modes contribute
- Drop ~ 0.5%
- Mode softening

Frederiksen *et al.*, Phys. Rev. Lett. 93, 256601 (2004).

Mode selectivity ?



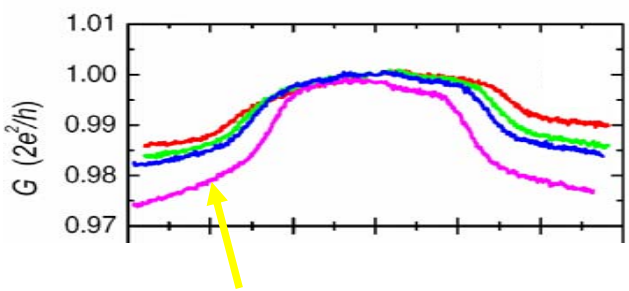


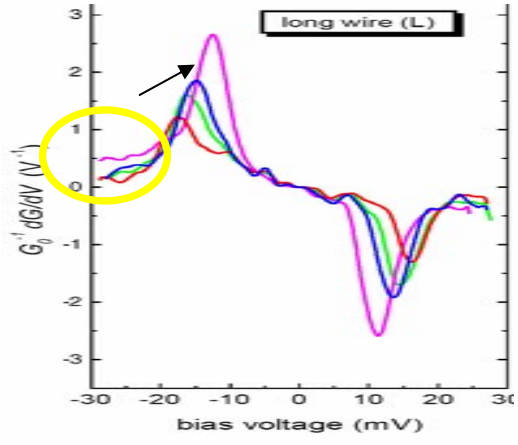
- **No:** two modes for the strained chain
- No transversal modes participate
- **Yes:** one mode for unstrained chain (Alternating Bond Mode)

Frederiksen *et al.*, Phys. Rev. Lett. 93, 256601 (2004).

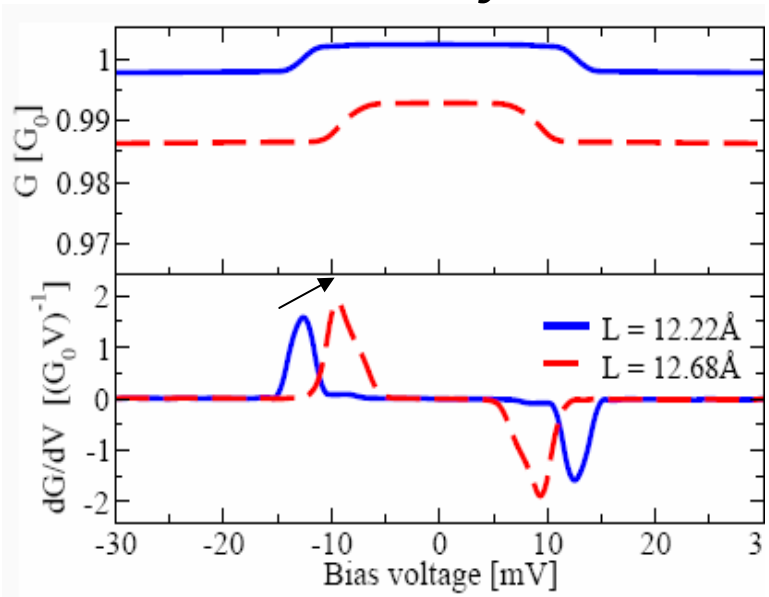
$N_{ph} = n_B(4K) \sim 0$

Experiment:





Theory:



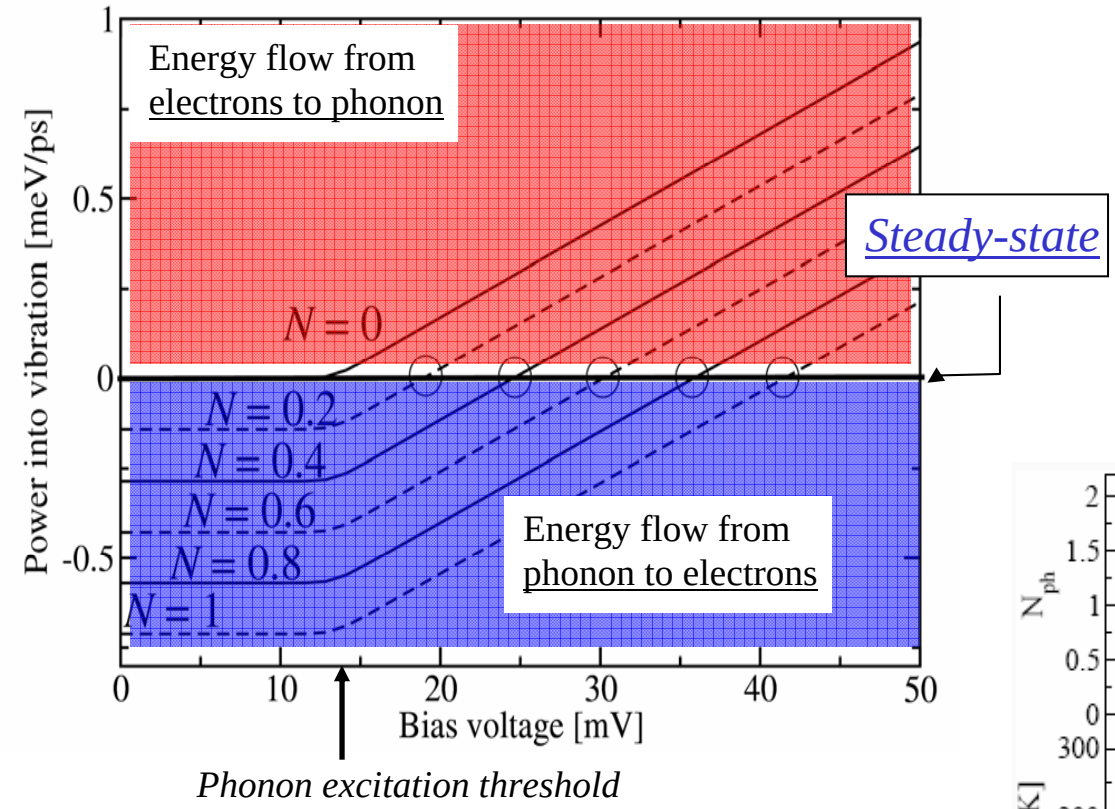
- All modes included
- Few modes contribute
- Drop ~ 0.5%
- Mode softening
- Zero slope beyond threshold

Frederiksen *et al.*, Phys. Rev. Lett. 93, 256601 (2004).

Energy flow

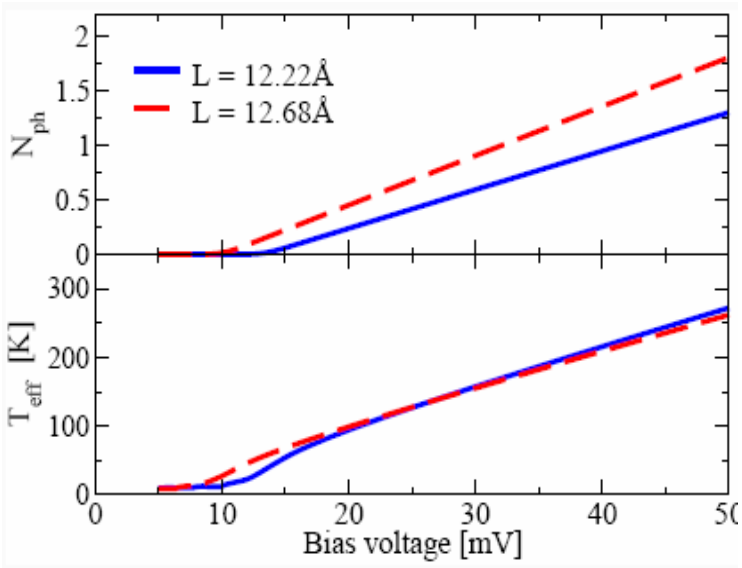
$$P^{\alpha} = \int \frac{d\omega}{2\pi} \omega \mathrm{Tr}[\boldsymbol{\Sigma}^{\alpha,<}(\omega)\mathbf{G}^{>}(\omega) - \boldsymbol{\Sigma}^{\alpha,>}(\omega)\mathbf{G}^{<}(\omega)]$$

Including only the most important mode:



Phonon excitation threshold

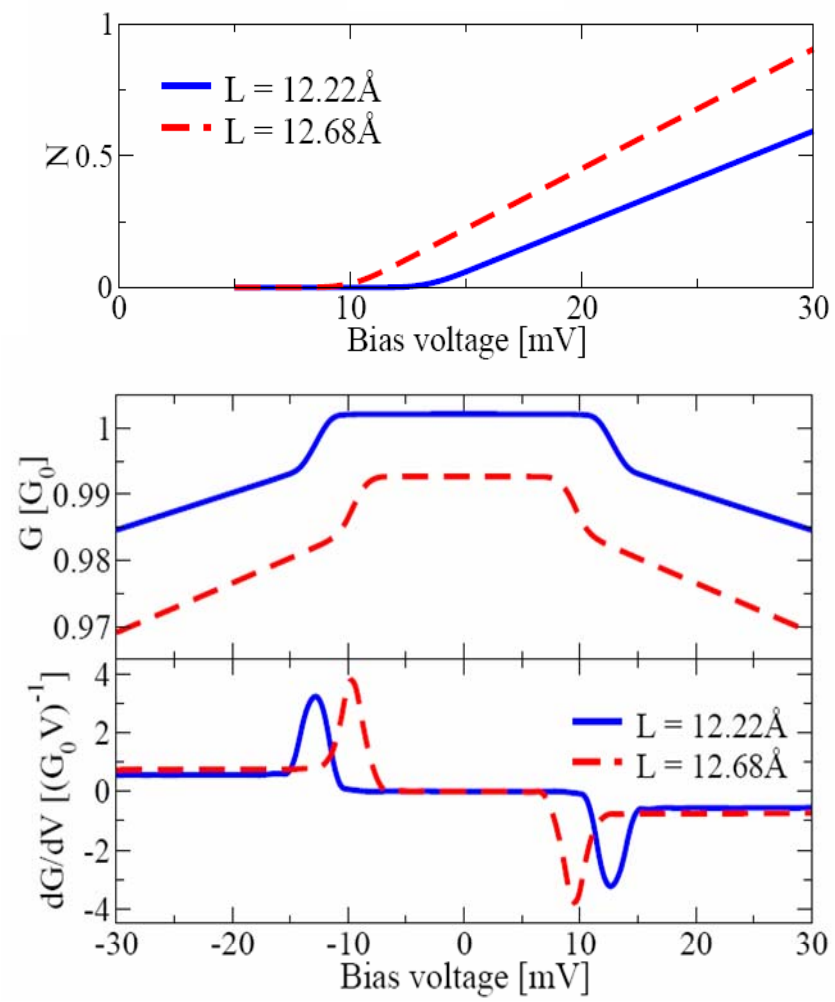
- Assuming phonons only damped by the coupling to electrons
- Steady state number of phonons:



Frederiksen *et al.*, Phys. Rev. Lett. 93, 256601 (2004).

Conductance incl. heating:

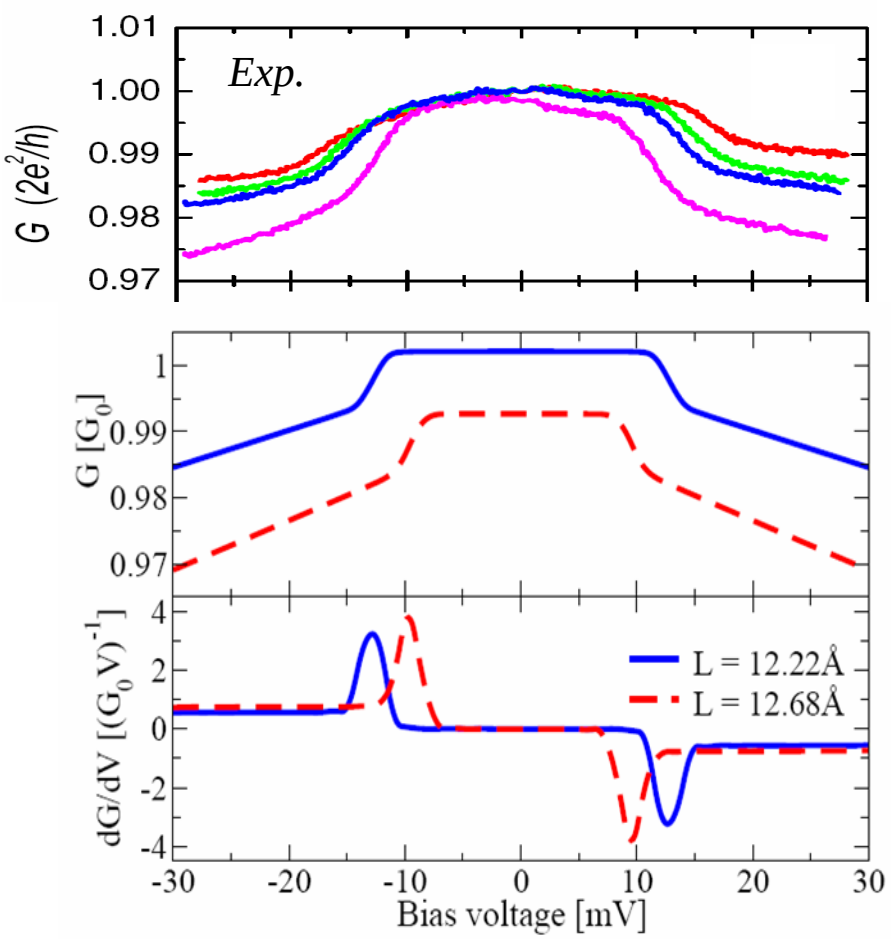
Theory



- Larger drop (≙1%)
- Slope of conductance beyond threshold

Frederiksen *et al.*, Phys. Rev. Lett. 93, 256601 (2004).

Comparing theory and experiment



- Conductance drop:
  Exp. $\sim 1 - 1.5\%$, Th. $\sim 1\%$
- Mode softening:
  $\Delta\Omega/\Delta L = -7$meV/Å same as exp.
- Increase of phonon signal with strain.
- Conductance slope due to heating:
  slightly larger than exp.
- Peak broadening:
  FWHM $\approx 5k_BT \approx 2$meV for 1 mode,
  FWHM ≈ 4meV for 2 modes.
  $\gamma_{\text{e-h}} \sim 30\mu$eV $\ll \Omega$

  FWHM(exp) ≈ 5meV

Frederiksen *et al.*, Phys. Rev. Lett. 93, 256601 (2004).

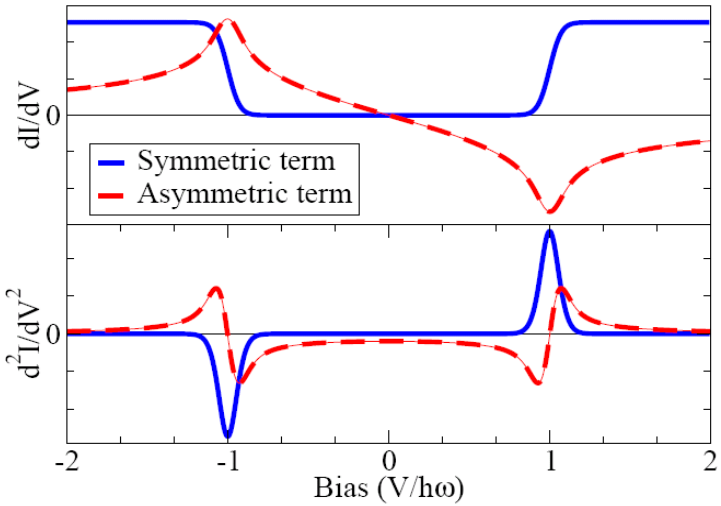
Full SCBA is computationally too expensive for more complicated structures. Therefore: try lowest order expansion in the phonon coupling:

(Magnus Paulsson, Thomas Frederiksen, Mads Brandbyge PRB RC Nov 2005)

$$I_\alpha = \frac{-e}{\hbar}\int_{-\infty}^{\infty}\frac{\mathrm{d}\omega}{2\pi}\mathrm{Tr}[\mathbf{\Sigma}_\alpha^{<}(\omega)\mathbf{G}^{>}(\omega) - \mathbf{\Sigma}_\alpha^{>}(\omega)\mathbf{G}^{<}(\omega)]$$

- Approximations:
  - Lowest order expansion in M
  - Energy independent DOS
- Analytical integration over energy
  - Lengthy derivation
  - Mathematica!

The expression for the current "simplifies" in the lowest order expansion – all GF's are now without phonon-coupling.



$$I^{(1)} = \frac{e}{\pi\hbar}\left(eV\,\mathrm{Tr}\left[G\Gamma_2 G^\dagger\Gamma_1\right] + \sum_\alpha I_\alpha^{(1)}\right)$$

$$I_\alpha^{(1)} = \left(2eVn_\alpha + \frac{\hbar\omega_\alpha - eV}{e^{\frac{\hbar\omega_\alpha - eV}{kT}} - 1} - \frac{\hbar\omega_\alpha + eV}{e^{\frac{\hbar\omega_\alpha + eV}{kT}} - 1}\right) \times$$

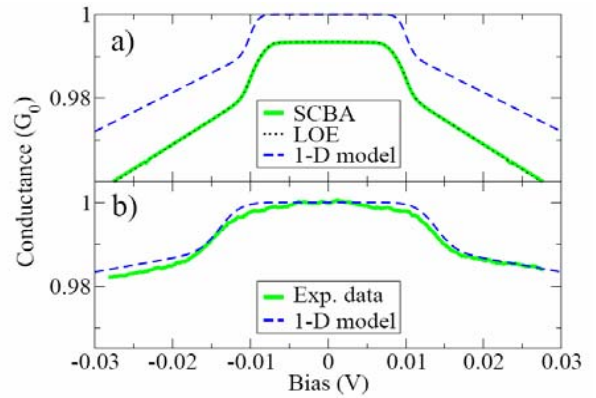
$$\mathrm{Tr}\left[G^\dagger\Gamma_1 G\left(M_\alpha G\Gamma_2 G^\dagger M_\alpha + \frac{i}{2}\left\{\Gamma_2 G^\dagger M_\alpha A M_\alpha - M_\alpha A M_\alpha G\Gamma_2\right\}\right)\right] +$$

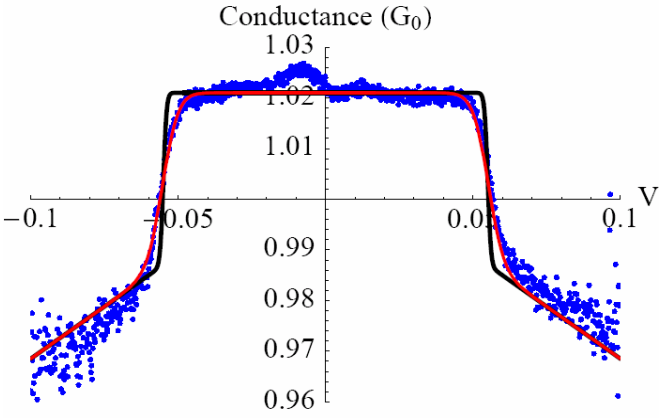
$$\frac{1}{2}\int_{-\infty}^{\infty} \mathrm{dE}\left(f(E) - f(E+eV)\right)\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\mathrm{dx}\,\frac{f(x+\hbar\omega_\alpha) - f(x-\hbar\omega_\alpha)}{x-E} \times$$

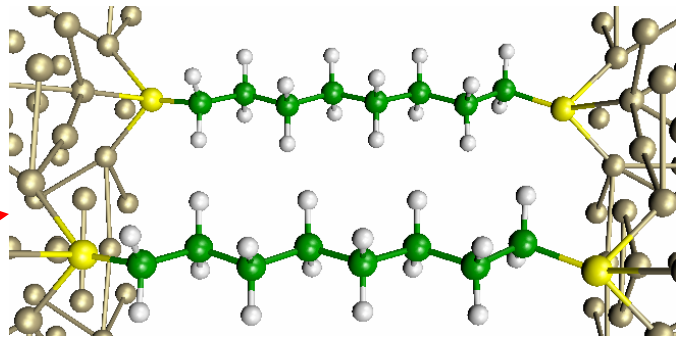
$$\mathrm{Tr}\left[G^\dagger\Gamma_1 G\left(\Gamma_2 G^\dagger M_\alpha G\left\{\Gamma_2 - \Gamma_1\right\}G^\dagger M_\alpha + M_\alpha G\left\{\Gamma_2 - \Gamma_1\right\}G^\dagger M_\alpha G\Gamma_2\right)\right]$$

(M.Paulsson, *et al.*, cond-mat/0505473)

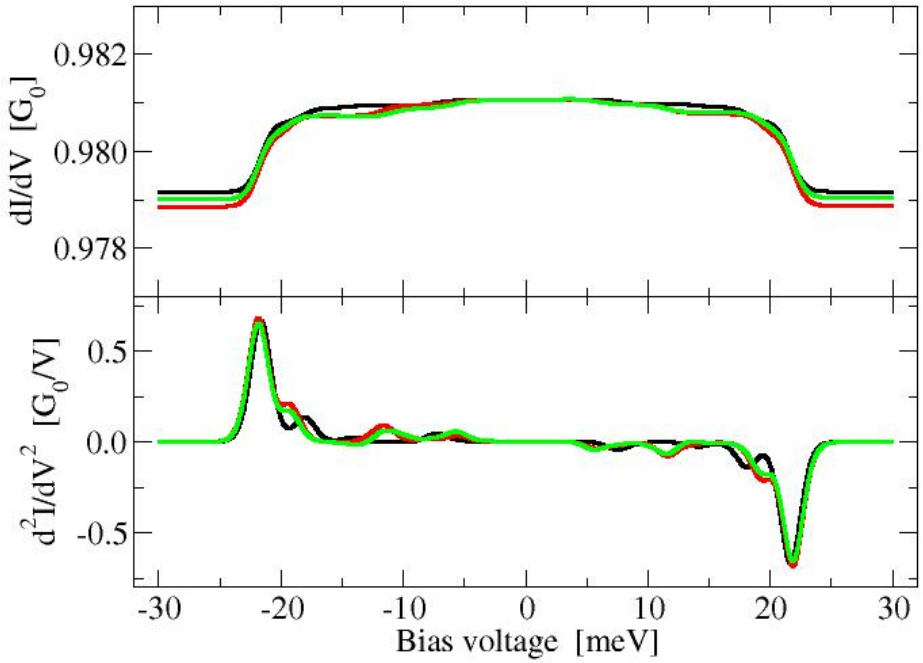
- SCBA
  - Possible but difficult
  - Scattering in Gold chains described by DFT
- Lowest order expansion
  - Computationally simple
  - Accurate
  - Intuitive explanation of exp.
- Large systems with DFT!



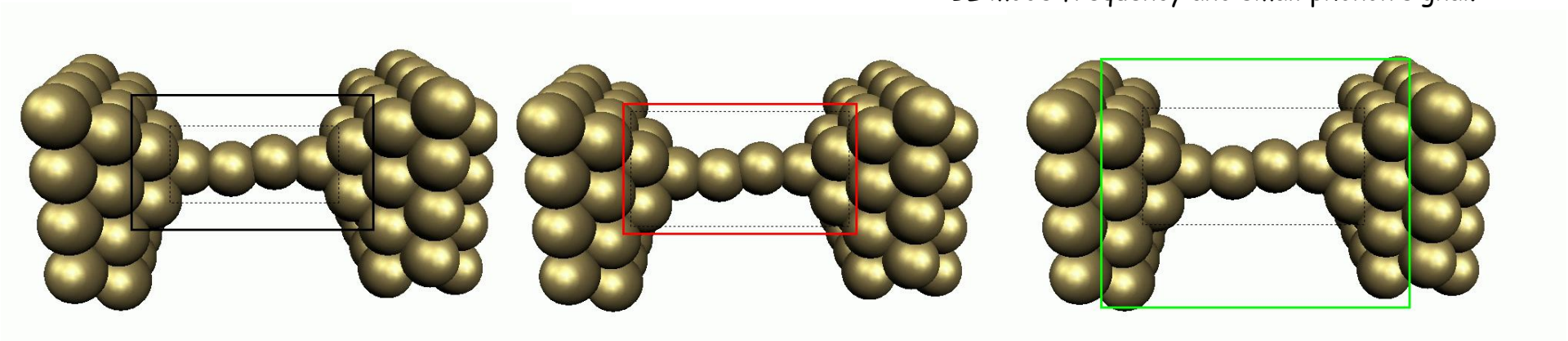




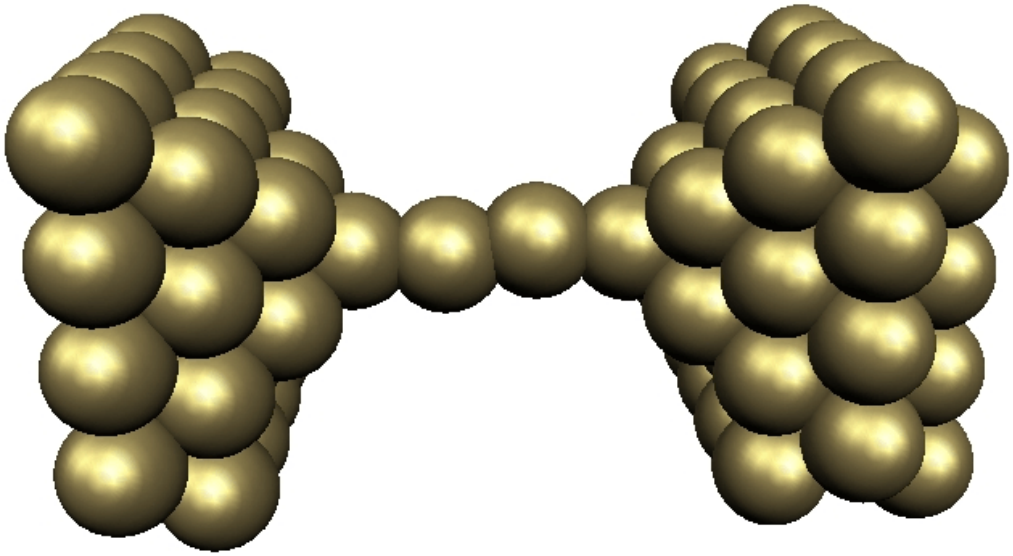
Does the device size matter?



- 4-atom **Au-wire** (sligtly **zig-zag**)
- Pyramidal bases, (100) surfaces
- 4x4 atoms in transverse plane of unit cell
- Single-Zeta plus polarization SIESTA basis (**9 orbitals per atom**)
- Wire, base, and first electrode layer are relaxed
- Lowest order expansion calculations **(LOE)**
- **No heating** in this calculation!
- Vibrational modes with energy >5meV included (low freq modes not accurate)
- Black dotted box indicates the vibrating atoms
- Colored (full line) boxes denote the device subspace for the transport calculation
- The low tension of the wire leads to high ABL mode frequency and small phonon signal.

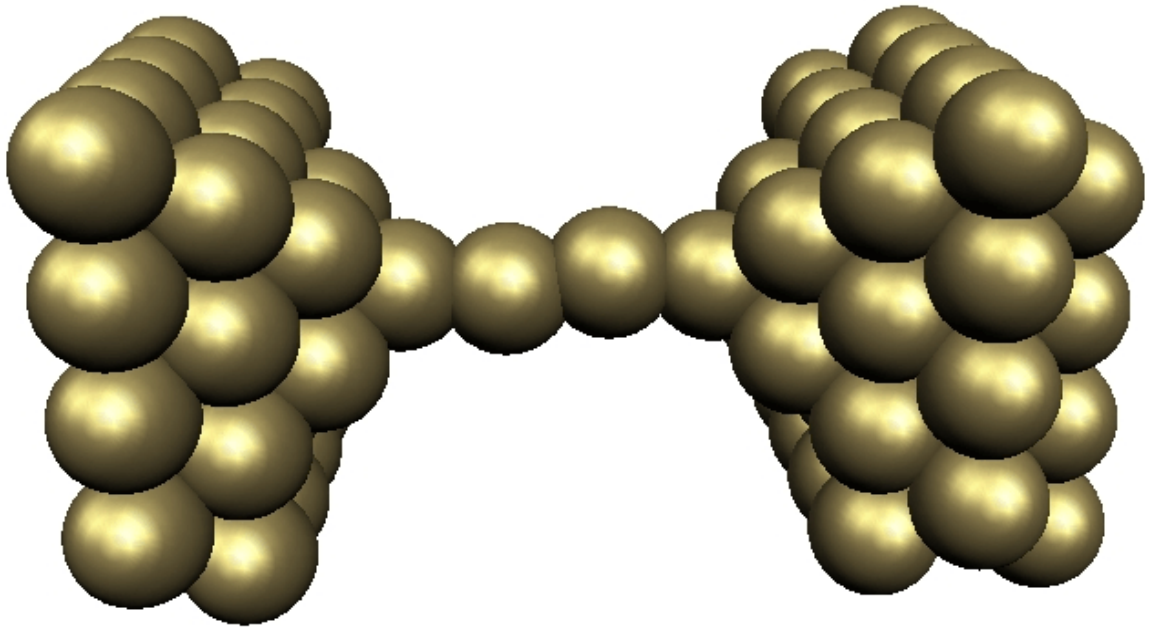


The important ABL mode from red/green vibrating region (21.9meV)



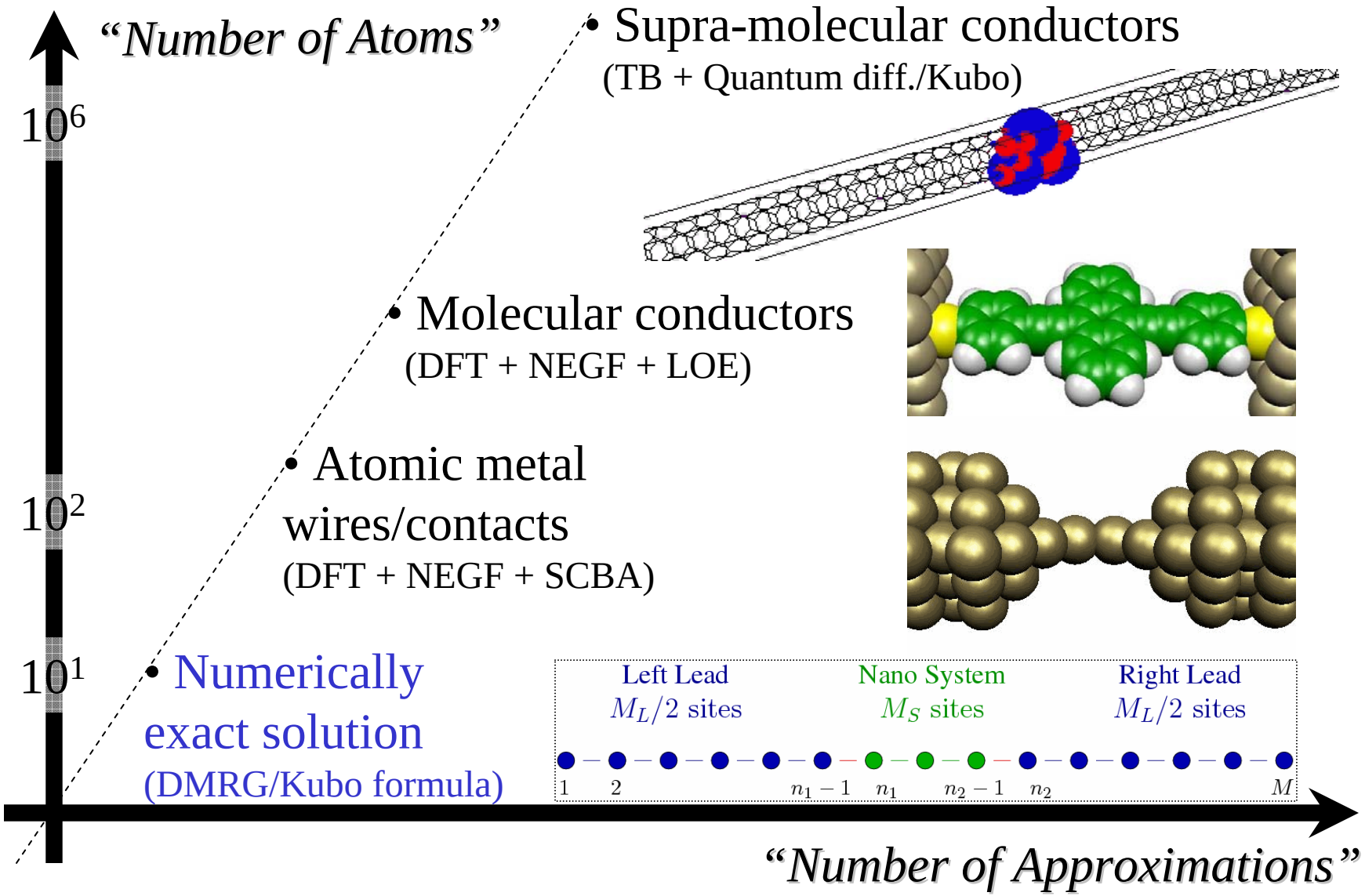
Notice that the pyramidal bases move, but only VERY LITTLE!

The important ABL mode from black vibrating region (21.6meV)



Notice that the pyramidal bases DO NOT move!

Molecular electronics/transport



Density-Matrix Renormalization Group for transport – Kubo formalism

(Dan Bohr, Peter Schmitteckert, Peter Wölfle, APJ)

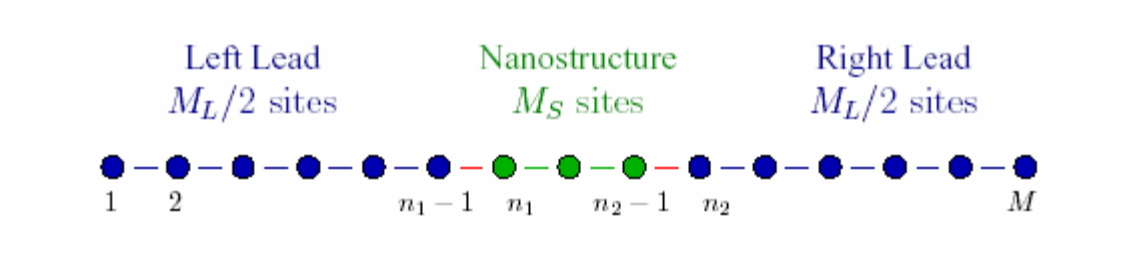
Vision:

DMRG is a (numerically) exact method for calculating correlation functions for (mostly one-dimensional) strongly correlated systems. **We want to develop a transport theory based on DMRG.**

Bonuses: Due to its "exactness", DMRG will be a very stringent benchmark for approximate theories.

Minuses: DMRG is very numerical, and it is not always straightforward to interpret the results physically. Also, DMRG becomes very costly if one goes beyond 1-d, implying possibly only a restricted domain for applications.

The model:



$$
\begin{aligned}
\hat{H}_0 &= \hat{H}_{\rm NS} + \hat{H}_{\rm L} + \hat{H}_{\rm C}, \\
\hat{H}_{\rm NS} &= \sum_{j=n_1}^{n_2-1} U_g c_j^\dagger c_j + \sum_{j=n_1+1}^{n_2-1} \left(- t_{\rm Dot}(c_j^\dagger c_{j-1} + c_{j-1}^\dagger c_j) + V c_j^\dagger c_j c_{j-1}^\dagger c_{j-1} \right) \\
\hat{H}_{\rm L} &= -t \sum_{i=2}^{n_1-1} (c_i^\dagger c_{i-1} + c_{i-1}^\dagger c_i) - t \sum_{i=n_2+1}^{M} (c_i^\dagger c_{i-1} + c_{i-1}^\dagger c_i), \\
\hat{H}_{\rm C} &= -t_L (c_{n_1}^\dagger c_{n_1-1} + c_{n_1-1}^\dagger c_{n_1}) - t_R (c_{n_2}^\dagger c_{n_2-1} + c_{n_2-1}^\dagger c_{n_2}) \\
&\quad + \gamma_V V (c_{n_1}^\dagger c_{n_1} c_{n_1-1}^\dagger c_{n_1-1} + c_{n_2}^\dagger c_{n_2} c_{n_2-1}^\dagger c_{n_2-1}).
\end{aligned}
$$

Kubo formula (linear response):

$$\begin{aligned}
\hat{H} &= \hat{H}_0 + \delta\hat{H}, \\
\langle \tilde{J}_n(t) \rangle &= \bar{J} - i \int_{-\infty}^{t} dt' \langle \psi_0 | [\tilde{J}_n(t), \delta\tilde{H}(t')] | \psi_0 \rangle, \\
\tilde{J}_n(t) &= -it_n \left[\tilde{c}_n^\dagger(t)\tilde{c}_{n-1}(t) - \tilde{c}_{n-1}^\dagger(t)\tilde{c}_n(t) \right],
\end{aligned}$$

Two different correlation functions can be used to calculate the conductivity:

$$\begin{aligned}
g_{J_j N} &= -\frac{e^2}{h} \langle \psi_0 | \hat{J}_{n_j} \frac{4\pi i\eta}{(\hat{H}_0 - E_0)^2 + \eta^2} \hat{N} | \psi_0 \rangle, \\
g_{JJ} &= \frac{e^2}{h} \langle \psi_0 | \hat{J}_{n_1} \frac{8\pi\eta(\hat{H}_0 - E_0)}{\left[(\hat{H}_0 - E_0)^2 + \eta^2\right]^2} \hat{J}_{n_2} | \psi_0 \rangle
\end{aligned}$$

Since these are ground state correlators, (corrector vector) DMRG is directly (at least in principle) applicable for their evaluation.

Use damped boundary conditions to reduce finite-size effects (here for right lead):

$$[-t,\cdots,-t,\underbrace{-t,-t,\cdots,-t}_{M_D}] \rightarrow [-t,\cdots,-t,\underbrace{-td,-td^2,\cdots,-td^{M_D-1},-td^{M_D}}_{M_D}]$$

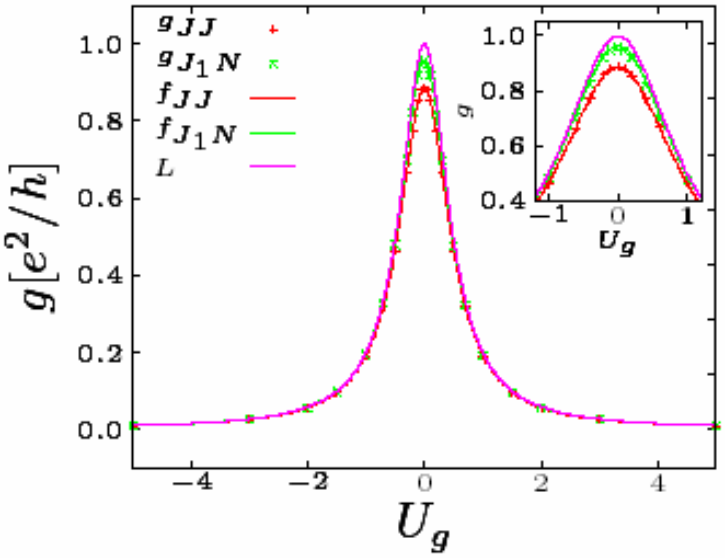
The parameter η, which in analytic calculations is a positive infinitesimal, must be chosen carefully in numerical calculations.

It must be larger than level splitting, arising from the finite system size, thus allowing transport.

It must be smaller than the widths of whatever resonances the system may exhibit.

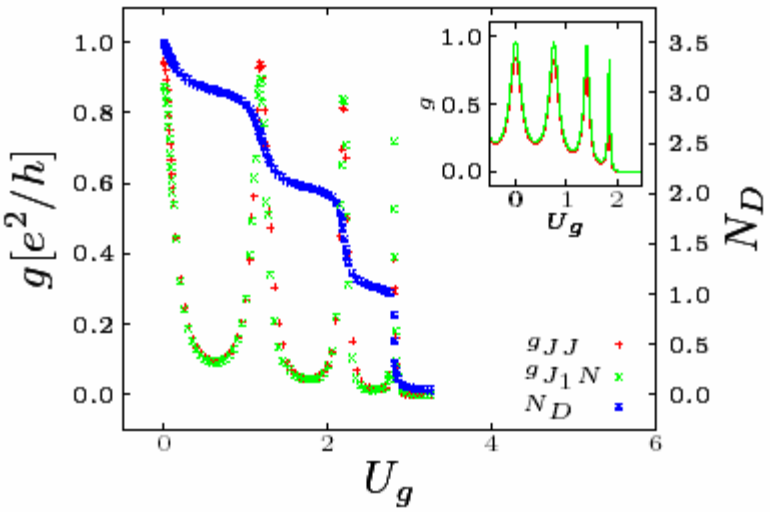
An appetizer for the results that can be obtained.

First bench-mark: compute the conductance for a non-interacting model, which can be solved by exact diagonalization.

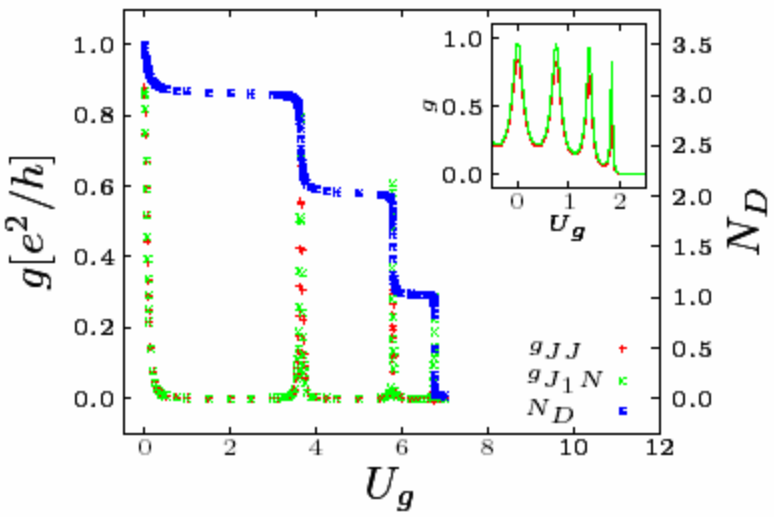


(a) Single resonant level. f's denote exact diagonalization results, g's denote DMRG results, and L denotes the exact Lorentzian result in the infinite lead limit. The inset shows an enlargement of the resonance peak.

Examples of results that **cannot** be obtained by "simple" techniques (the beauty of the approach is that interacting and noninteracting problems equally difficult/simple!!):

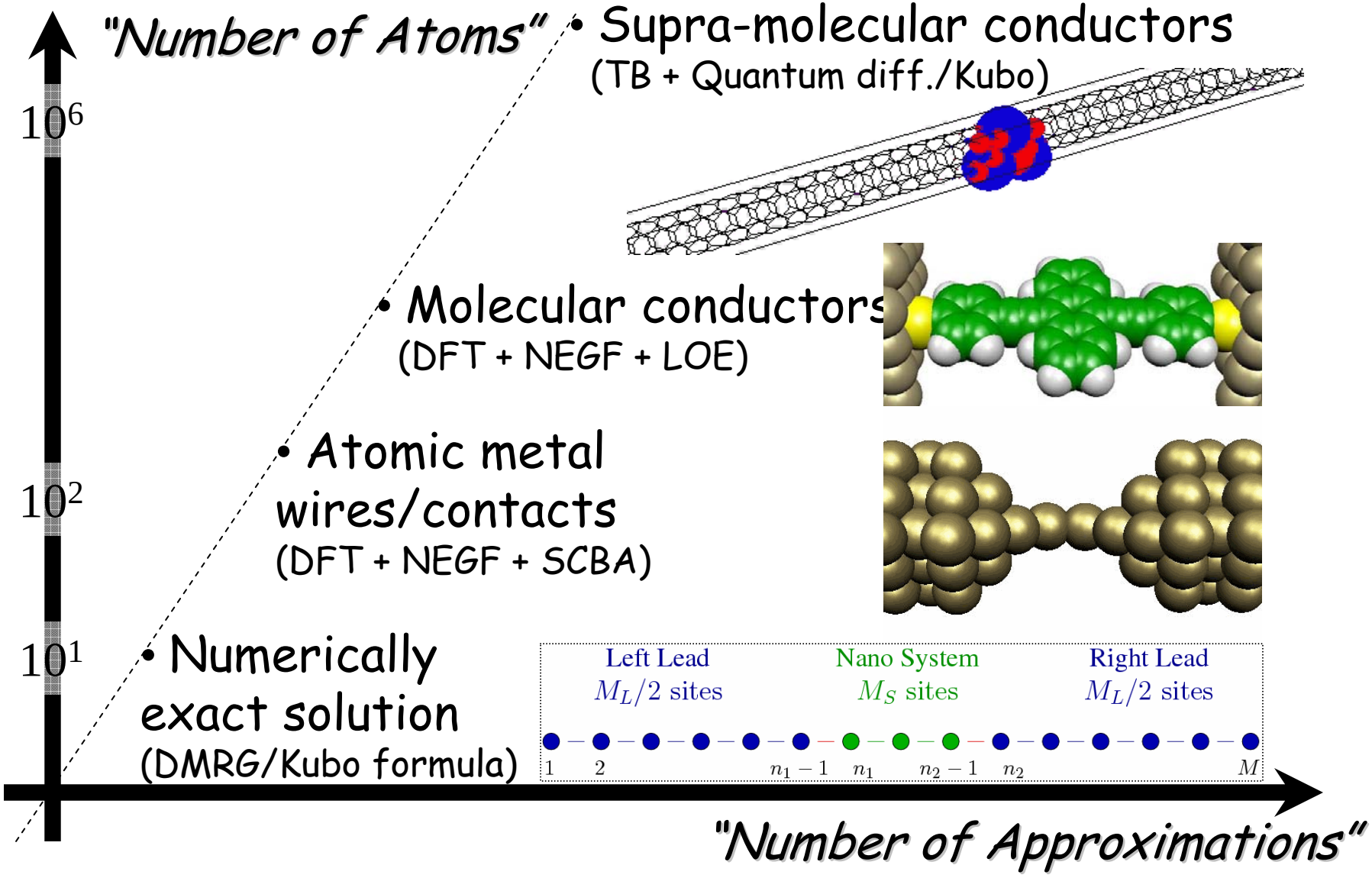


(b) Seven site nanostructure in the Luttinger Liquid regime, $V = 1.0$. The inset shows the conductance for the non interacting structure.



(c) Seven site nanostructure in the charge density wave regime, $V = 5.0$. The inset shows the conductance for the non interacting structure.

Molecular electronics/transport



Thank you for your attention!

All the hard numerical calculations were done by

- Dan Bohr (graduate student)
- Thomas Frederiksen (graduate student)
- Troels Markussen (Master's student)
- Magnus Paulsson (post-doc)
- Mads Brandbyge (associate professor)