Time Dependent Transport Through Molecular Devices

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Nano-device Transport Simulation

First principles Device Simulator

Input

Device Atomic Positions

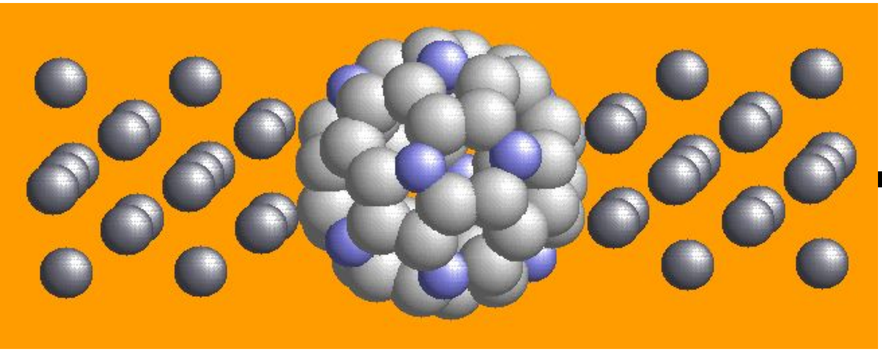
→

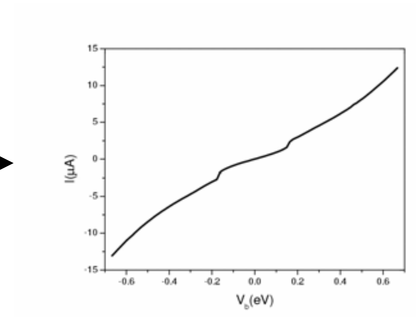
Device Simulator

→

Output

I-V curve
charge transfer
heat dissipation





Our method

→ NEGF + DFT

Taylor, Guo, and Wang, PRB **63**, 245407 (2001)

Non-equilibrium and non-linear quantum transport properties

Chemical information of molecular devices

Equivalent "KS" equation for an open system:

$$\begin{cases} -\nabla^2 U(x) = 4\pi\rho(x) = 4\pi i \int dE [G^<(E,U)]_{xx} \\ G^<(E,U) = iG^r(E,U)[\Gamma_L f_L + \Gamma_R f_R] G^a(E,U) \\ G^r(E,U) = \dfrac{1}{E - H - U - V_{ex}[\rho] - \Sigma^r} \end{cases}$$

G^r expanded using s, p, d real space LCAO basis set

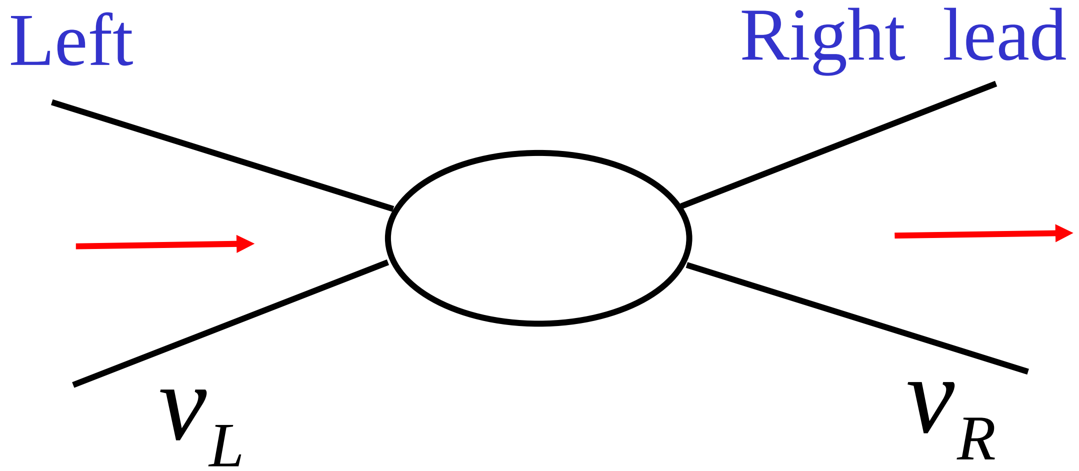
NEGF+DFT formalism

- When NEGF+DFT iteration converges, we obtain the potential landscape V on the Hartree level.
- This potential includes effect of charge transfer.
- Once the scattering potential V is known, it then becomes a scattering problem. We can use the NEGF formalism to calculate all kinds of quantum transport properties.

Issues in time dependent quantum transport

- Steady state ac transport
- Charge relaxation time
- Transient transport (response time)

ac transport theory

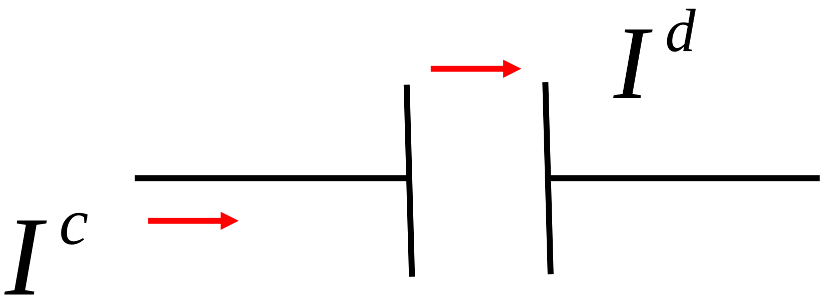


Conduction current : $$I^c = \frac{dq}{dt}$$

Continuity equation : $$I_L{}^c + I_R{}^c + \frac{dQ}{dt} = 0$$

⇨ Current is not conserved !

What is $\frac{dQ}{dt}$?



Parallel plate capacitor

Displacement current $J^d = \frac{1}{4\pi}\frac{\partial D}{\partial t}$ from Jackson's book

$$\frac{dQ}{dt} = \frac{\partial}{\partial t}\int \rho dV = \int \frac{1}{4\pi}\nabla \cdot \frac{\partial D}{\partial t} dV = \nabla \cdot \int J^d dV = I_L{}^d + I_R{}^d$$

Poisson Eq. $\nabla \cdot D = 4\pi\rho \longrightarrow \frac{\partial \rho}{\partial t} = \frac{1}{4\pi}\nabla \cdot \frac{\partial D}{\partial t}$

Total displacement current: $I^d = \frac{dQ}{dt}$

Q: charge accumulation in the device.

$$I_L^c + I_R^c + \frac{dQ}{dt} = 0$$

Conclusion

Current consists of two contributions at ac bias: the usual conduction current and the displacement current due to Coulomb interaction.

* Define total current $$I^t = I^c + I^d$$

So that $I_L^t + I_R^t = 0$ Current conservation

* One could partition the total displacement current into each lead

$$I_L^{\,d} + I_R^{\,d} = I^d$$

This has been done by Buttiker when external bias is small.

Buttiker et al, PRL **70**, 4114 (1993) and Wang et al, PRL **82**, 398 (1999).

* Dynamic conductance $\quad I^t(w) = G(w)(v_L - v_R)$

a). $w = 0 \quad G(0) \longrightarrow$ DC conductance, a real quantity.

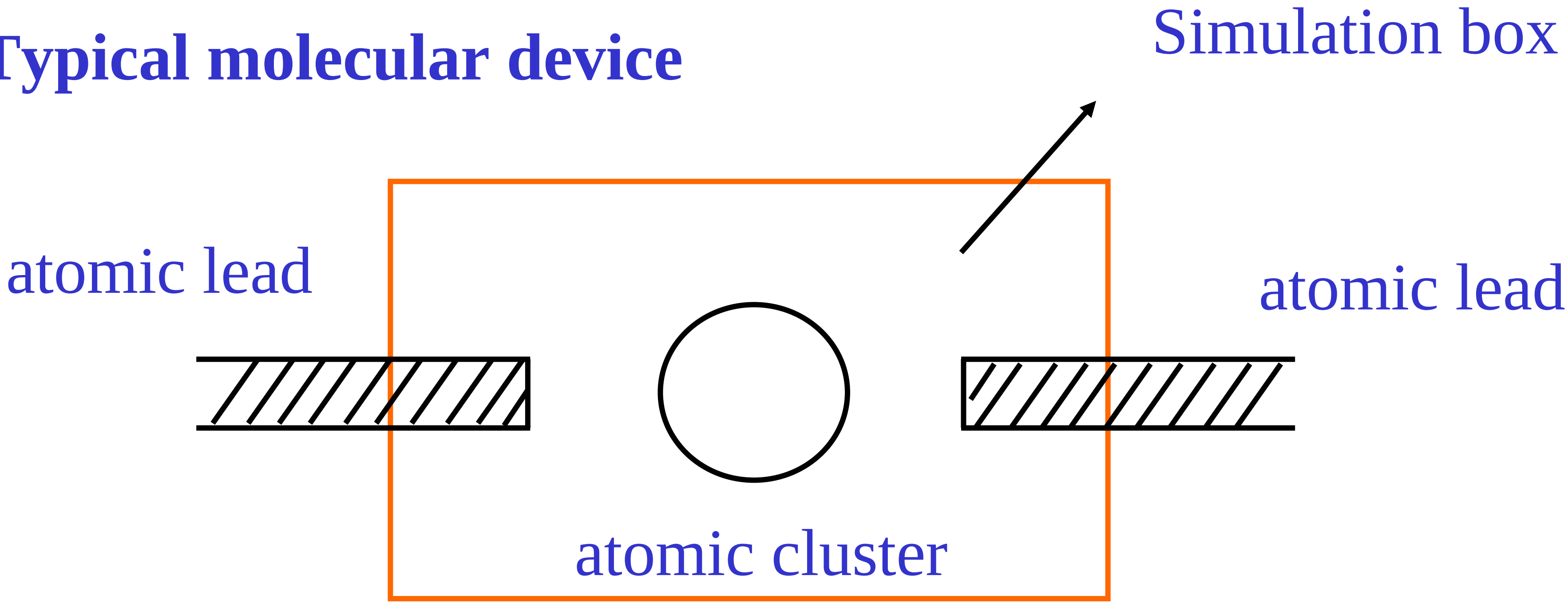
b). $w \neq 0 \qquad G(w) = G_R(w) + iG_I(w)$

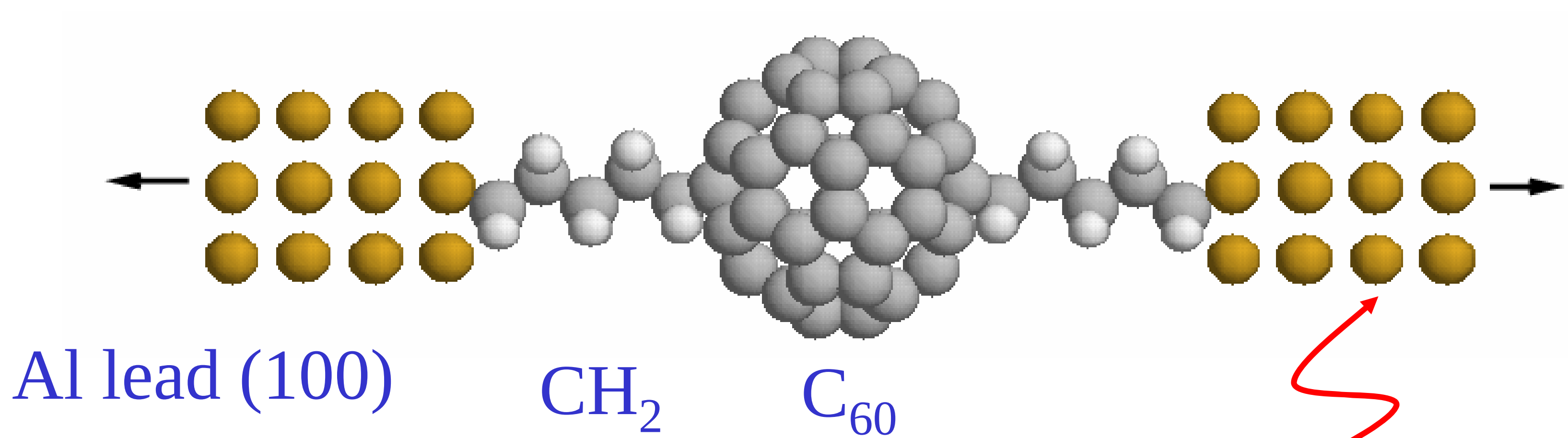
expressed in terms of non-equilibrium Green's function

dissipation

phase difference between I and v

Typical molecular device



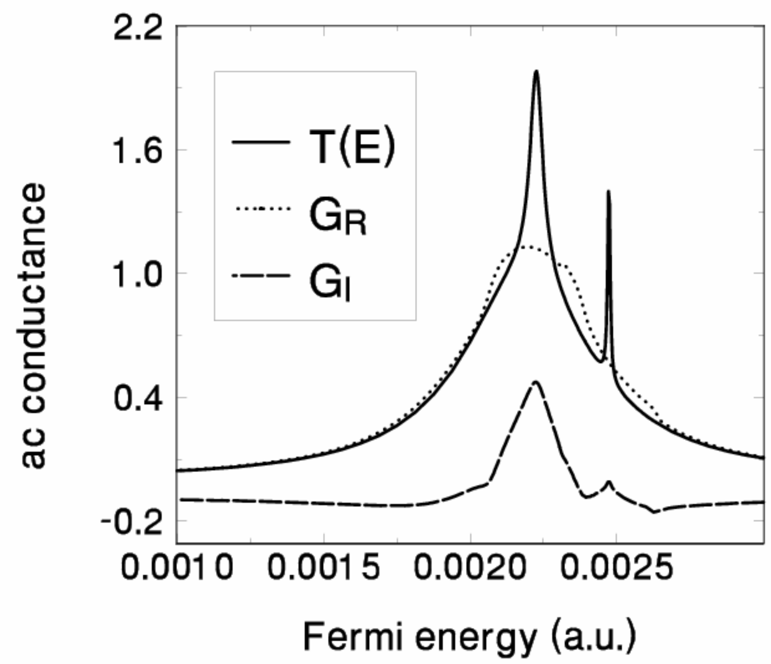


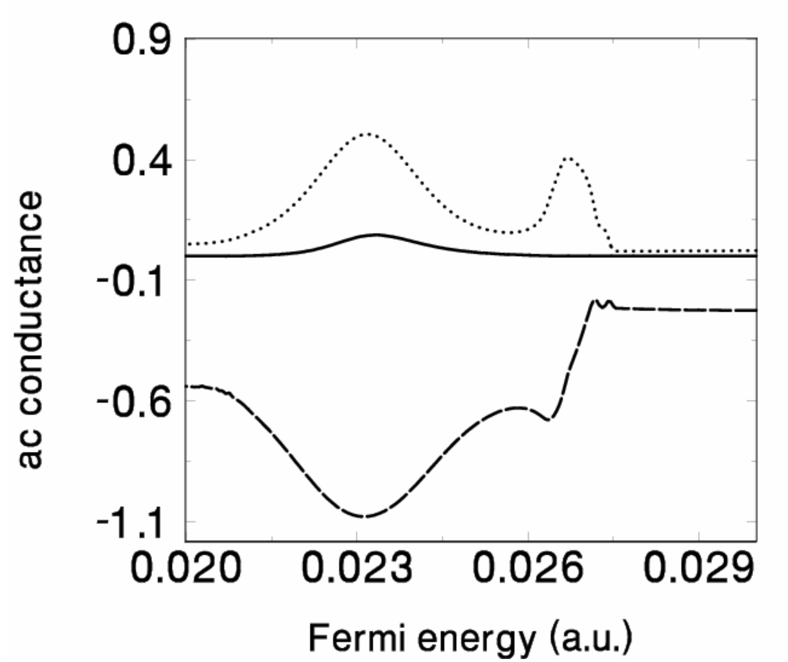
Due to highly resistive CH2 chain the system
is in the tunneling regime.

Results

J.L.Wu, B.G. Wang, J. Wang, and H. Guo, Phys. Rev. B 72, 195324 (2005)

Frequency: 100GHz





Fermi energy can be shifted by gate voltage
See Cronin et al, APL 84**, 2052 (2004).**

* two kinds of resonant behaviors

a). $T = 2,$ near the resonance, $G_R(w) < G_R(0)$

b). $T << 1,$ $G_R(w) > G_R(0)$

Classical circuit (qualitatively understanding)

*

R-L circuit

$$G(w) = \frac{1}{R - iwL} \qquad (Z = R - iwL)$$

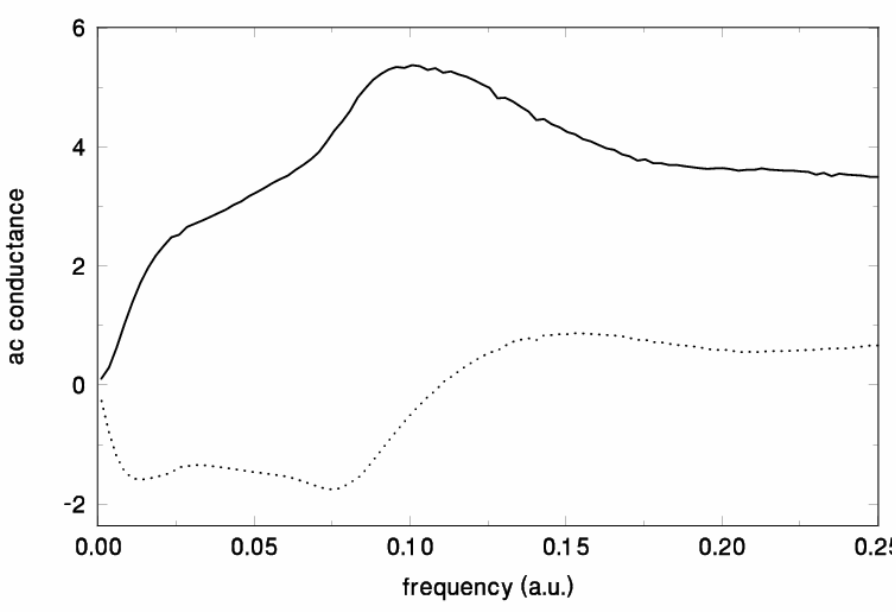
$$\approx \frac{1}{R} + \frac{iwL}{R^2} - \frac{w^2L^2}{R^3} + \ldots$$

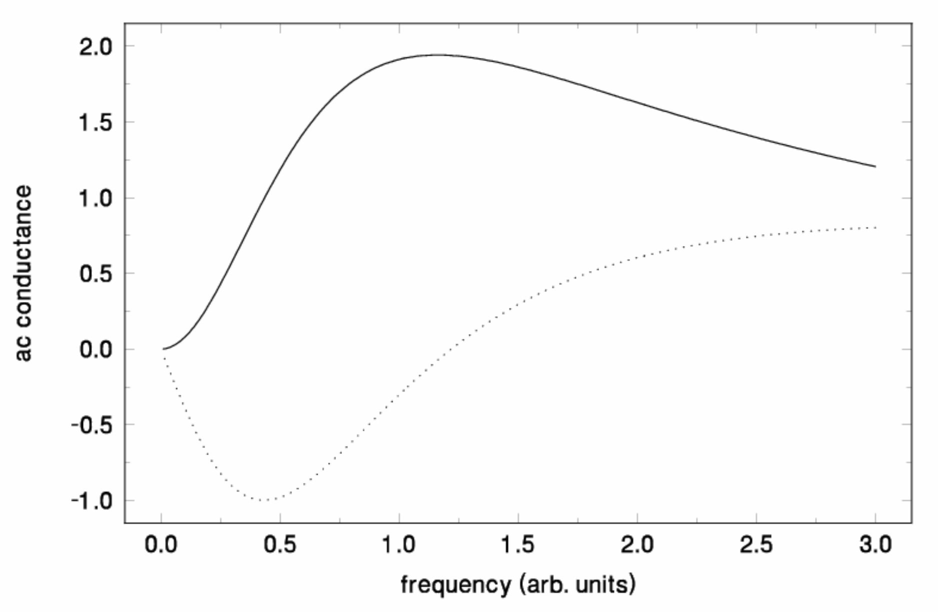
*

R-C circuit

$$G(w) = \frac{1}{R - \frac{1}{iwc}} = \frac{-iwc}{1 - iwcR} = -iwc + w^2c^2R$$

Comparison (at fixed Fermi energy)

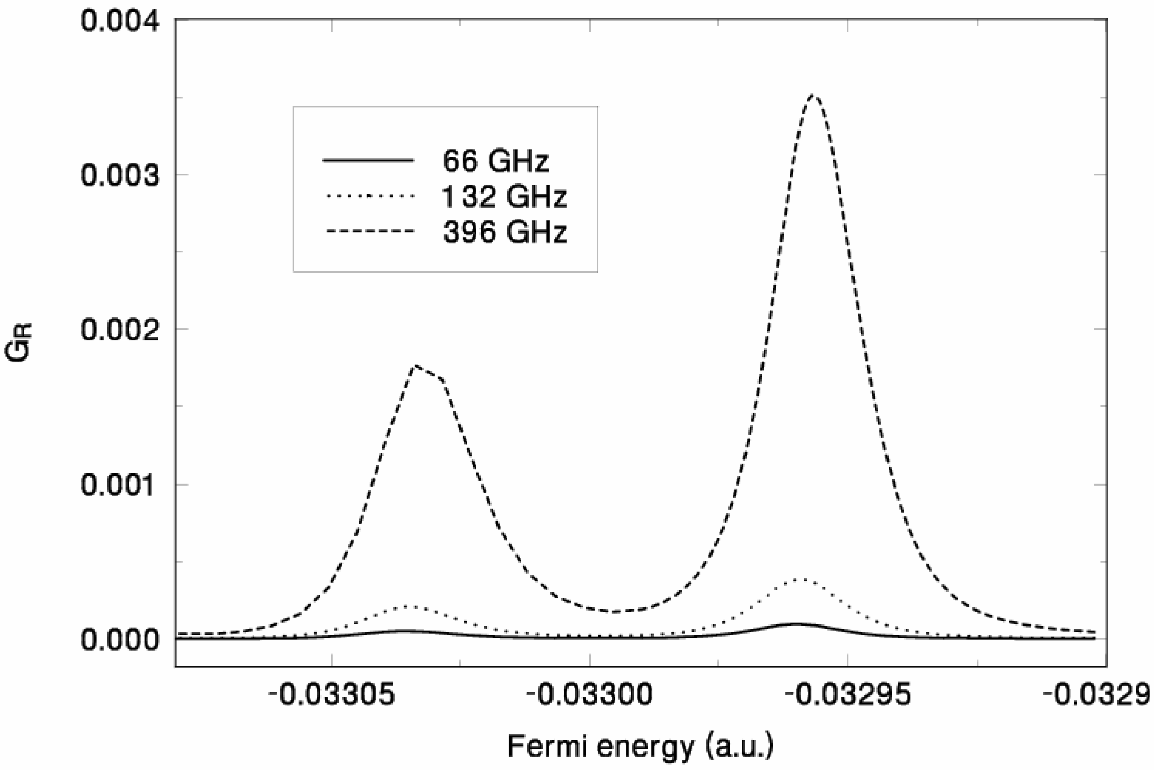


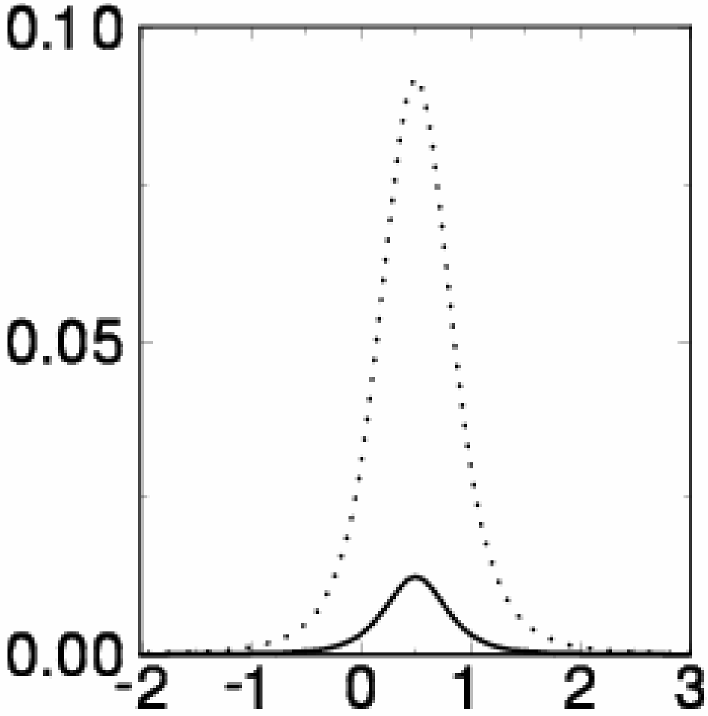


Ab initio calculation

Model calculation

Large magnification





Ab initio calculation

Model calculation

On experimental side

- Oscillation up to 0.7 THz has been achieved in InAs/AlSb resonant tunneling diodes. (Brown et al APL **58**, 2291 (1991).)
- SWNT transistor operated at 50GHz has been achieved. (Rosenblatt et al APL **87**, 153111 (2005).)
- It would be a challenge to measure the large magnification of ac conductance at 100GHz.

Issues in time dependent quantum transport

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For a nano-capacitor

Frequency dependent conductance

$$G(\omega) = -i\omega C_{\mu} + \omega^2 C_{\mu}^2 R_q$$

C_{μ} Electrochemical capacitance

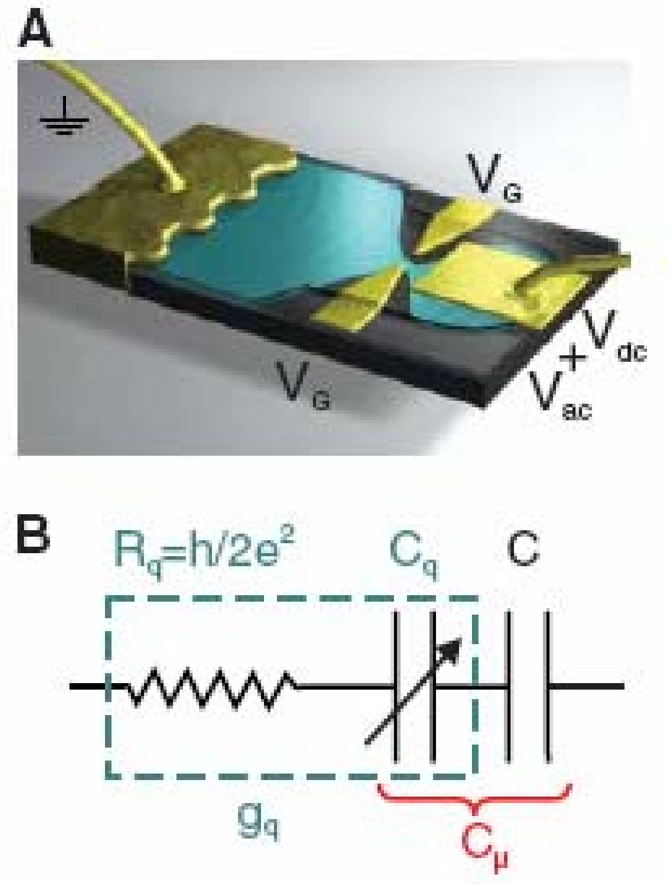
For nano-capacitor $$\frac{1}{C_{\mu}} = \frac{1}{C} + \frac{1}{C_{DOS}}$$ T.P. Smith et al, Phys. Rev. B32, 2695 (1985)

R_q Charge relaxation resistance, h/2e*e for a single channel

$\tau = R_q C_{\mu}$ Charge relaxation time

Recent experiment on mesocopic systems,

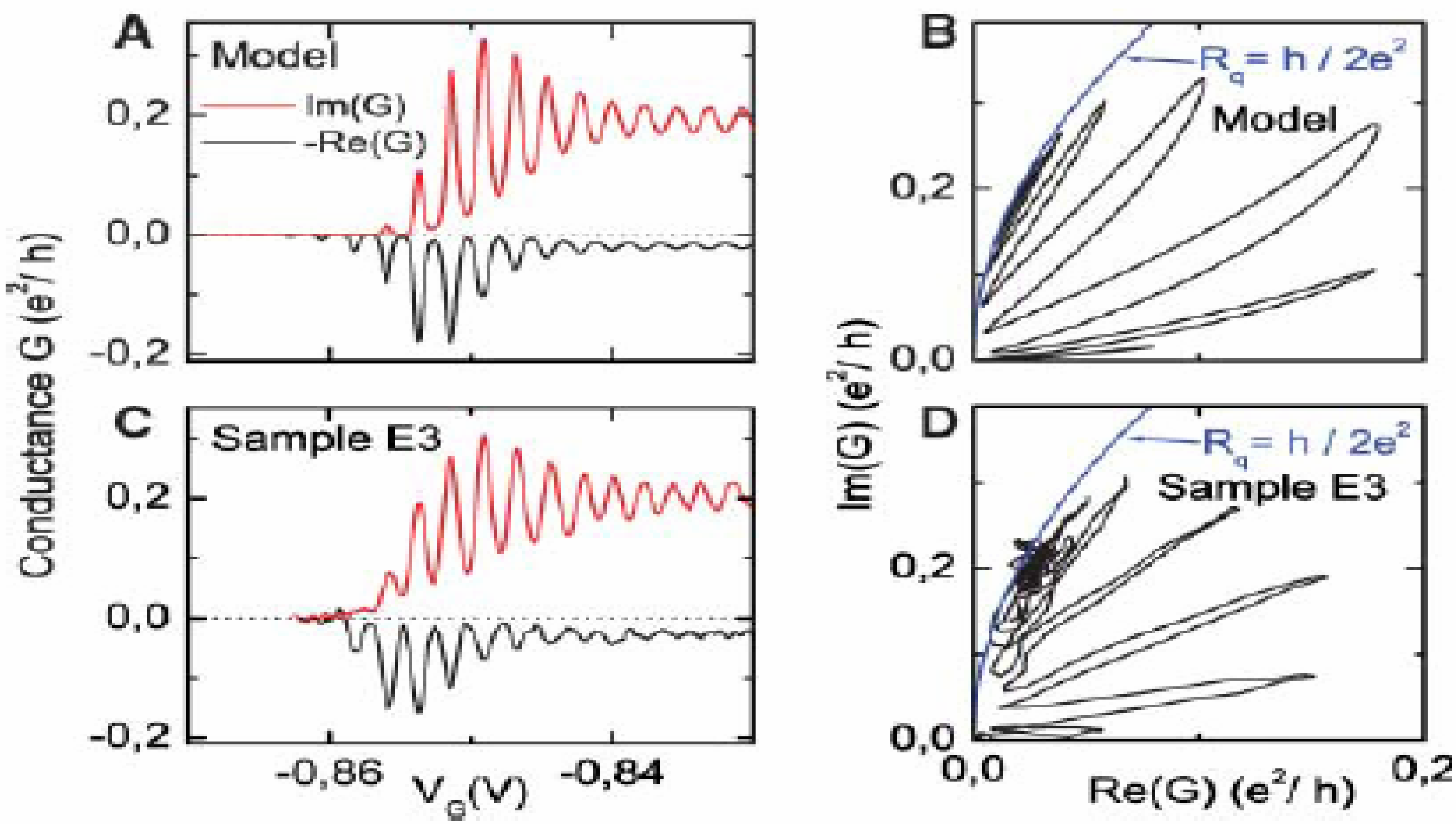
Gabelli et al, Science 313, 499 (2006)



$$\frac{1}{g_q(\omega)} = \frac{h}{2e^2} + \frac{1}{-i\omega C_q}$$

Frequency=1.2GHz

$$G = \frac{-i\omega C g_q(\omega)}{-i\omega C + g_q(\omega)} = \frac{-i\omega C_\mu (2e^2/h)}{-i\omega C_\mu + (2e^2/h)}$$



What is the nature of charge relaxation for atomic devices?

In addition,

- Non-equilibrium relaxation resistance?
- What happens for a leaky nano-capacitor?
- Frequency dependent capacitance and charge relaxation resistance?

Issues in time dependent quantum transport

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How fast a nano-electronic device can turn on/off a current ?

- For nano-devices, a quantum mechanical first principle calculation is needed.
- Problem of atomic leads: must go beyond the wideband limit.
- Our theoretical formalisms:

  (1). Time demain, Zhu et al, Phys. Rev. B 71, 075317 (2005)

  (2). Exact solution in k-space for a pulse, Maciejko, Wang, and Guo, Phys. Rev. B 74, 085324 (2006).
- Our theoretical results on model calculation show that there is a big difference between wideband limit and exact solution.

Difficulty with ab initio calculation

- Numerical problem with a triple integral.
- An ansatz can be used so that the triple integral reduces to a double integral.
- Very calculational demanding.
- Alternatively solution: find out all the poles of retarded Green's function.

Thank You!