Conductance of Single Molecular Junctions

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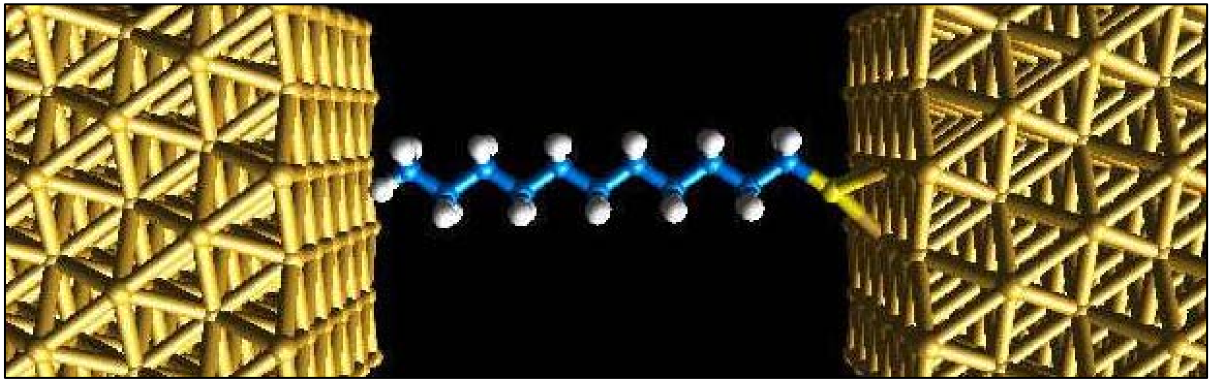


October 1, 2008

Outline:

1. Introduction

Why molecular electronics?

2. Comparison with experiments

Alkanethiol molecules

3. What is the single-molecule conductance?

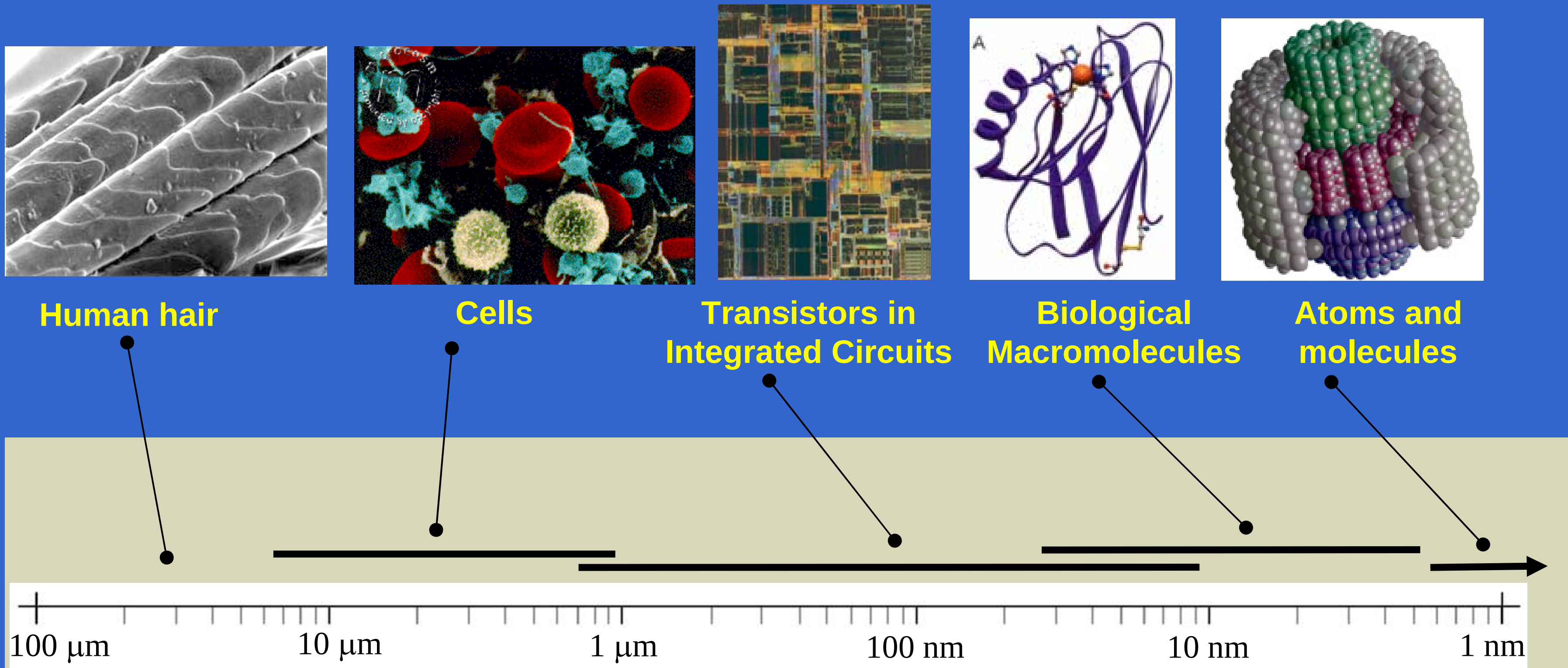
An alkanedithiol molecule

4. Spontaneous oscillation of current

A C_{60} molecule

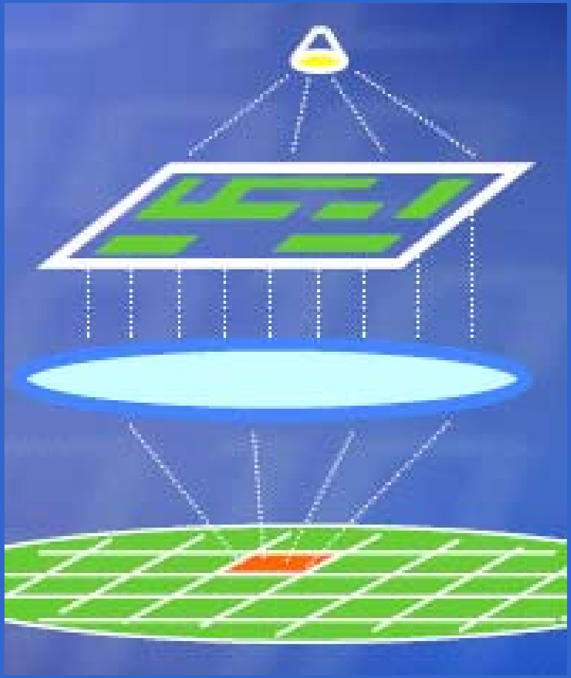
5. Summary

1. Introduction:



Nanotechnology: works at the atomic, molecular and supra-molecular levels, at the **0.1 – 100 nm scale**, with **fundamentally new properties.**

What’s the problem?



45 nm now

Physical limit:
Diffraction of light.

Economical limitation:
Too expensive.

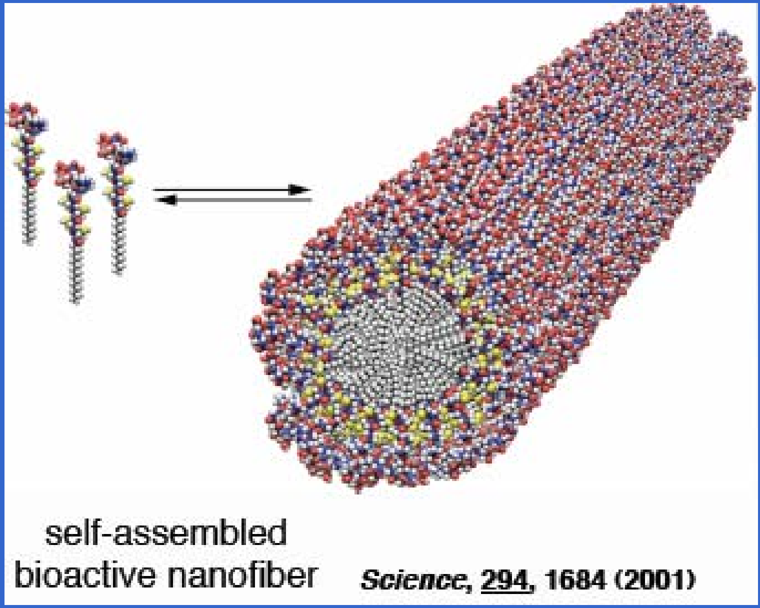
Molecular electronics: A solution

The main idea: use molecules to create analogues of today's IC chips.

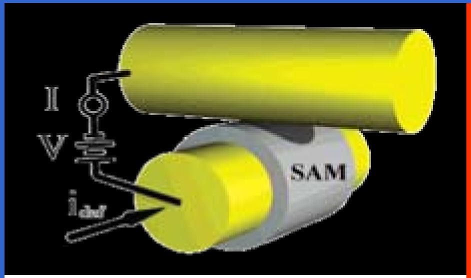
Because molecules are small and can form structures by self-assembly.

Aviram & Ratner, (1974).

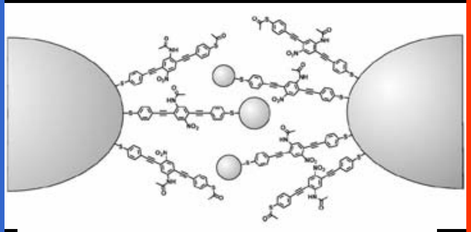
For example ..



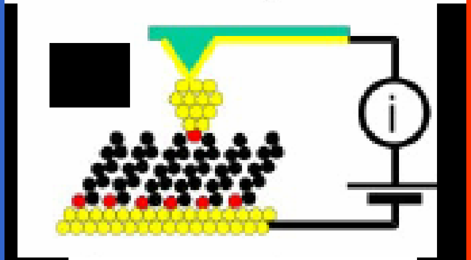
Some experiments



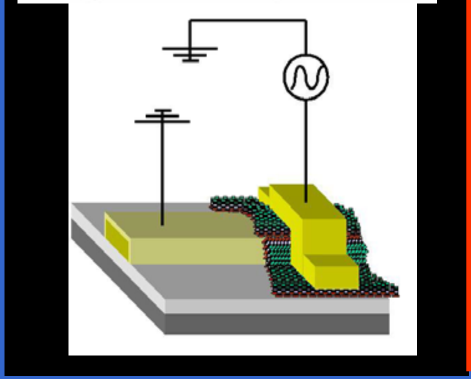
J.G. Kushmerick
NanoLetters '03



H.B. Weber
APL '03



S.M. Lindsay
Science '03



D. Stewart
NanoLetters '04

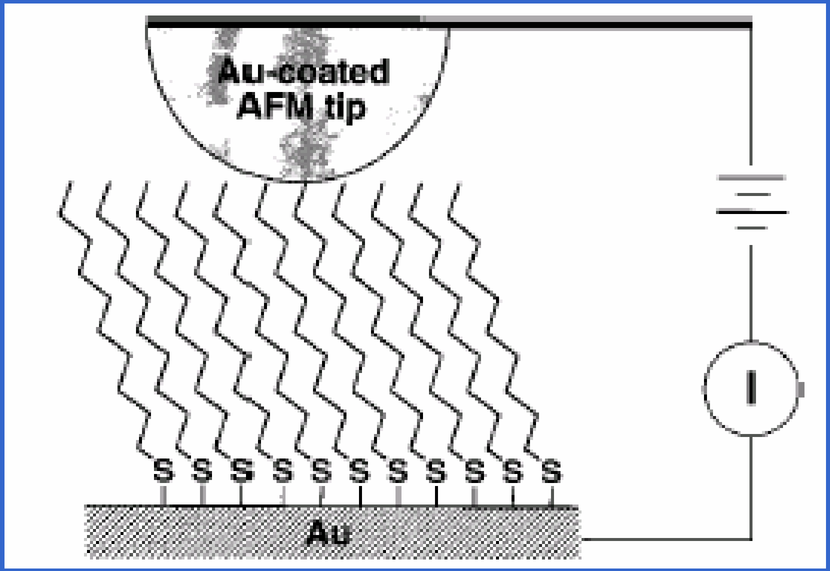
But, there is a big problem:

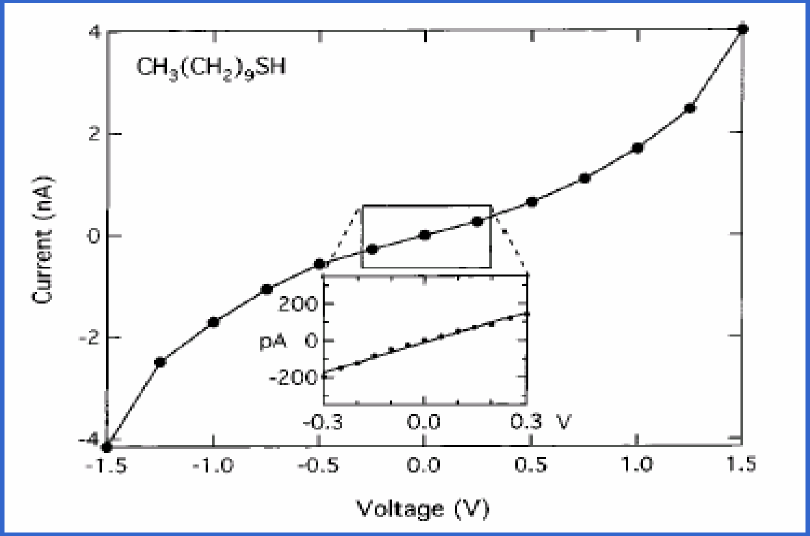
Most experimental data can not be reproduced by other groups!

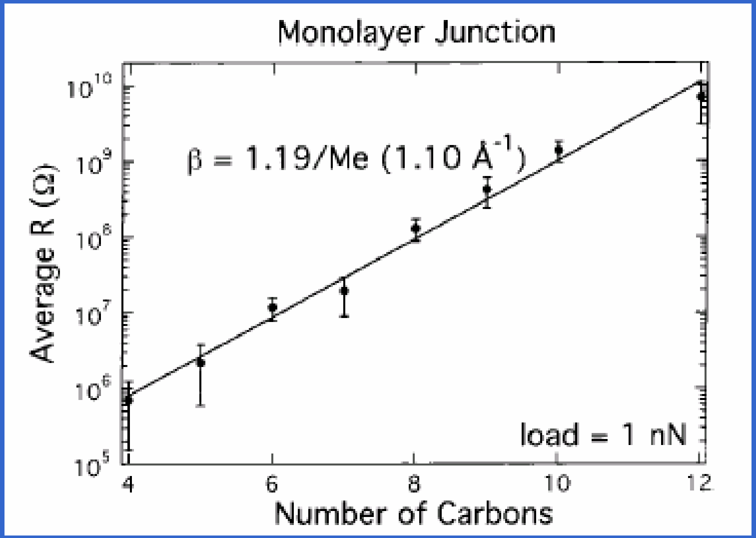
Except….

A SAM measurement: Alkanethiol molecular wires.

Wold and Frisbie, JACS 123, 5549 (2001)







Rather similar results from other groups: M. Reed et al (2003); Lindsay et al, Nanotechnology, 13, 5 (2002).

Can we simulate these experimental data from first principles?

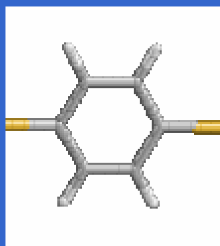
How to calculate current?

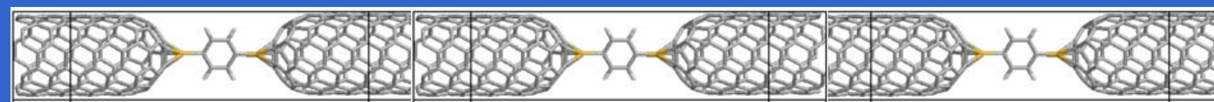
Landauer formula:

$$I(V_b) = \frac{2e^2}{h} \int_{-\infty}^{+\infty} T(E, V_b)\left(f_l - f_r\right) dE$$

Real space DFT plus Keldysh Green's functions: **Taylor, Guo, Wang, PRB 63, 245407(2001)-----McGill-Device-CALculator (McDCAL); Brandbyge, et al, PRB 65, 165401(2002)---Transiesta.**

Conventional Density Functional Theory (DFT) solves two kinds of problems:





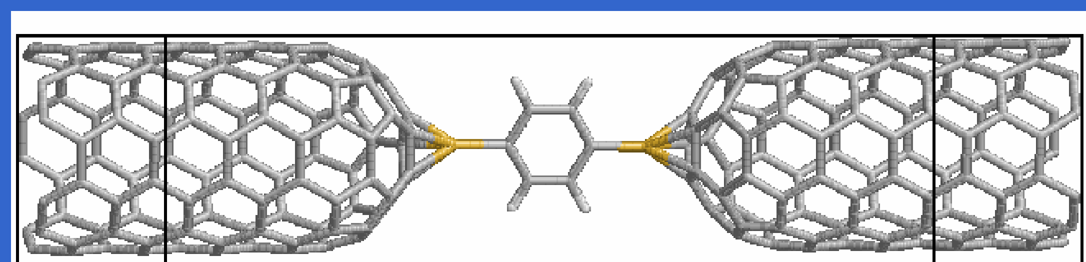
Periodic systems

VASP

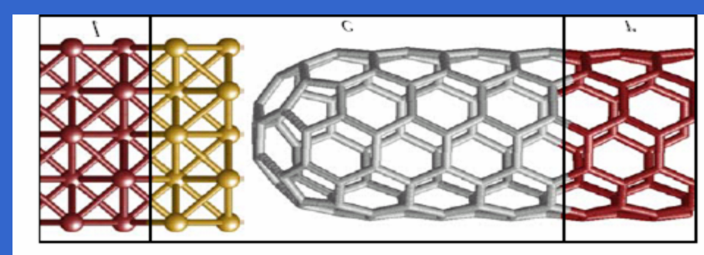
Finite isolated system

Gaussian-03

Quantum transport:

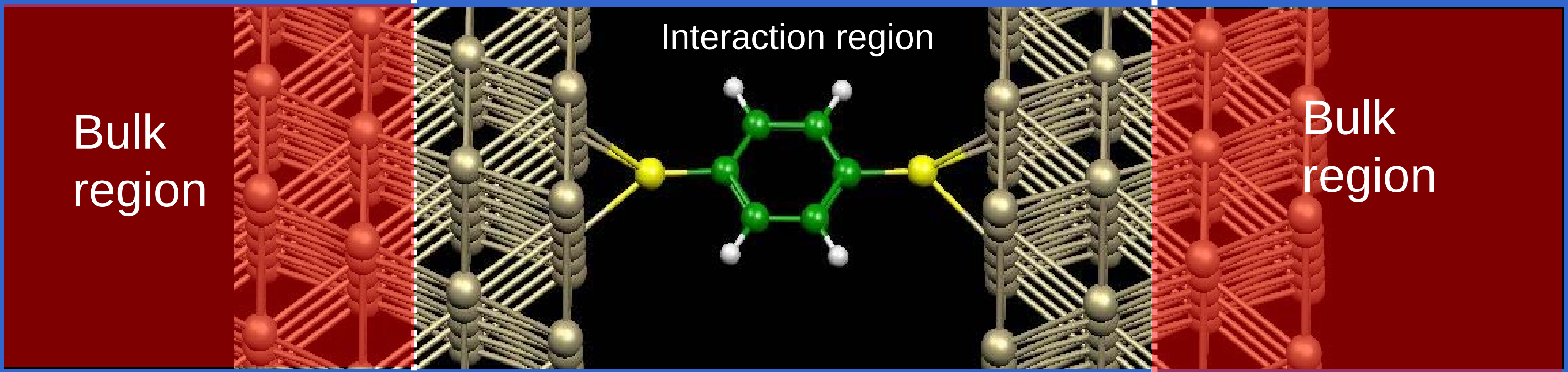


A device is neither finite nor periodic, and is in non-equilibrium



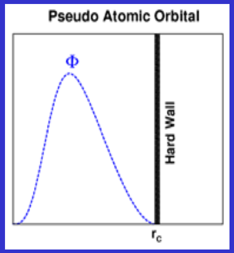
McDcal

Computational modeling



Electronic structure

- Density Functional Theory
- LCAO
- Pseudopotentials



ρ

H

Nonequilibrium physics

- Full description of electrodes using ab initio self-energies
- Non-equilibrium electron distribution using NEGF

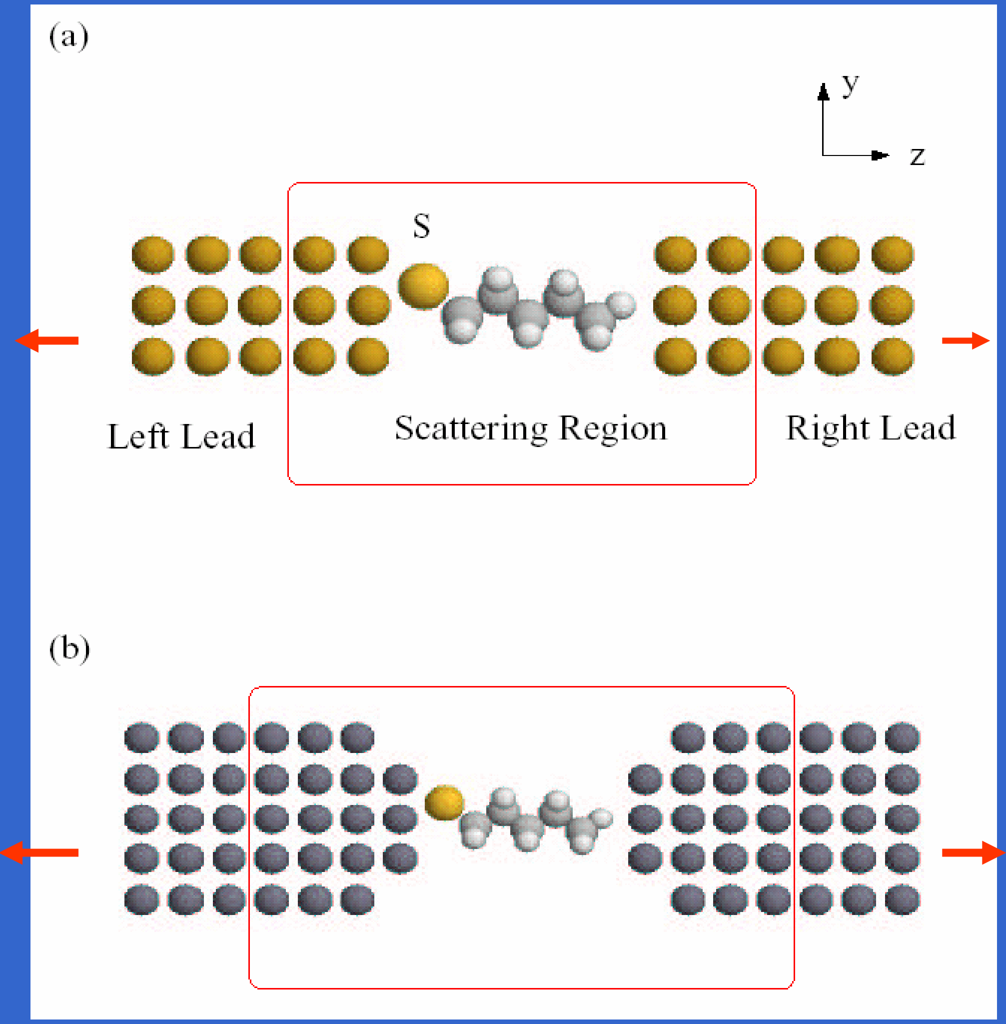
- Calculation of electron current

2. Comparison with experiments: Alkanethiol molecules

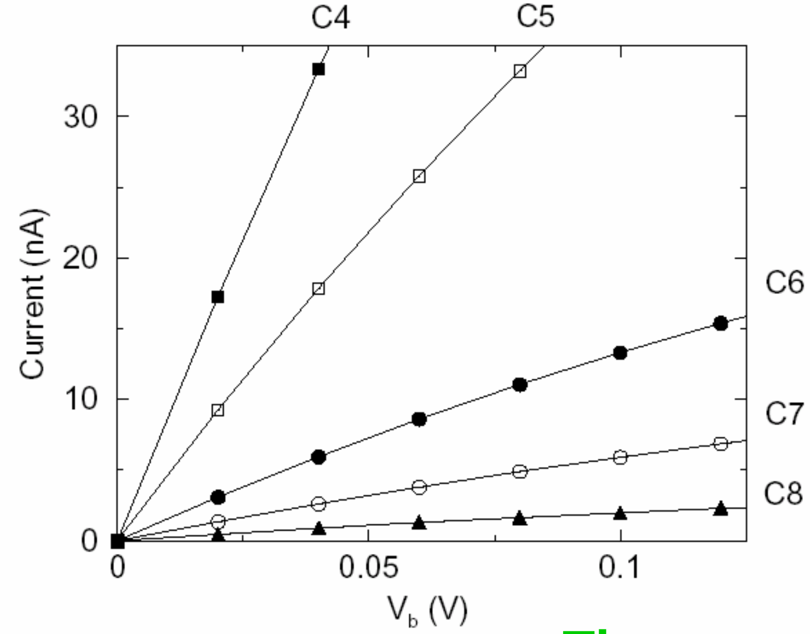
Our model:

Au electrodes

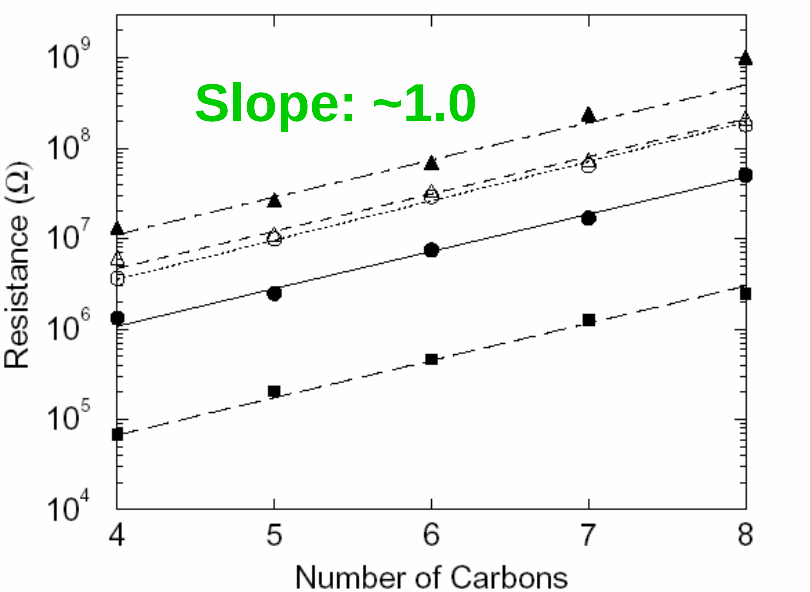
Al electrodes



Kaun & Guo, Nano Lett. 3, 1521 (2003)

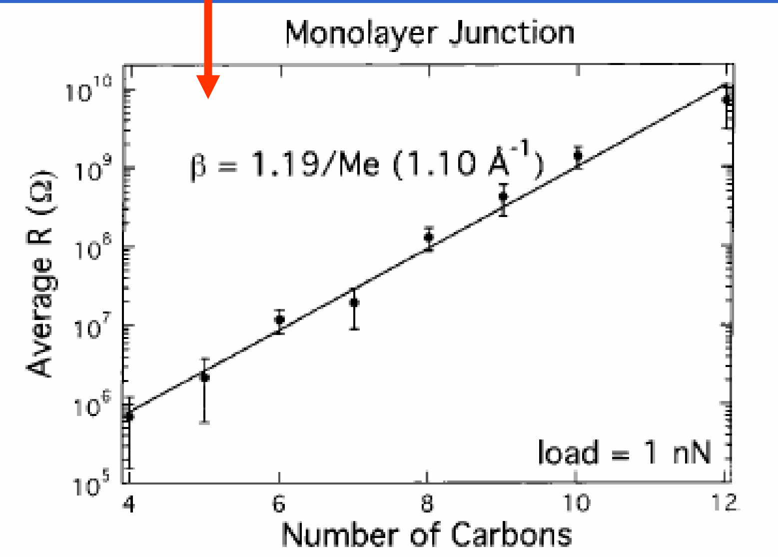


Theory



Quantitative agreement with measurements

Experimental: average slope (beta) is close to 1



From alkanethiol to alkanedithiol

- Our calculation: still shows $R_n = R_o \exp(\beta n)$;
- Our calculated beta is still about 1.0;
- Our R_o is smaller than that of alkanethiol by about a factor of 18.

Experiments so far:

1. Cui et al, J. Chem. Phys. 106, 8069 (2002): $\beta = 0.57$

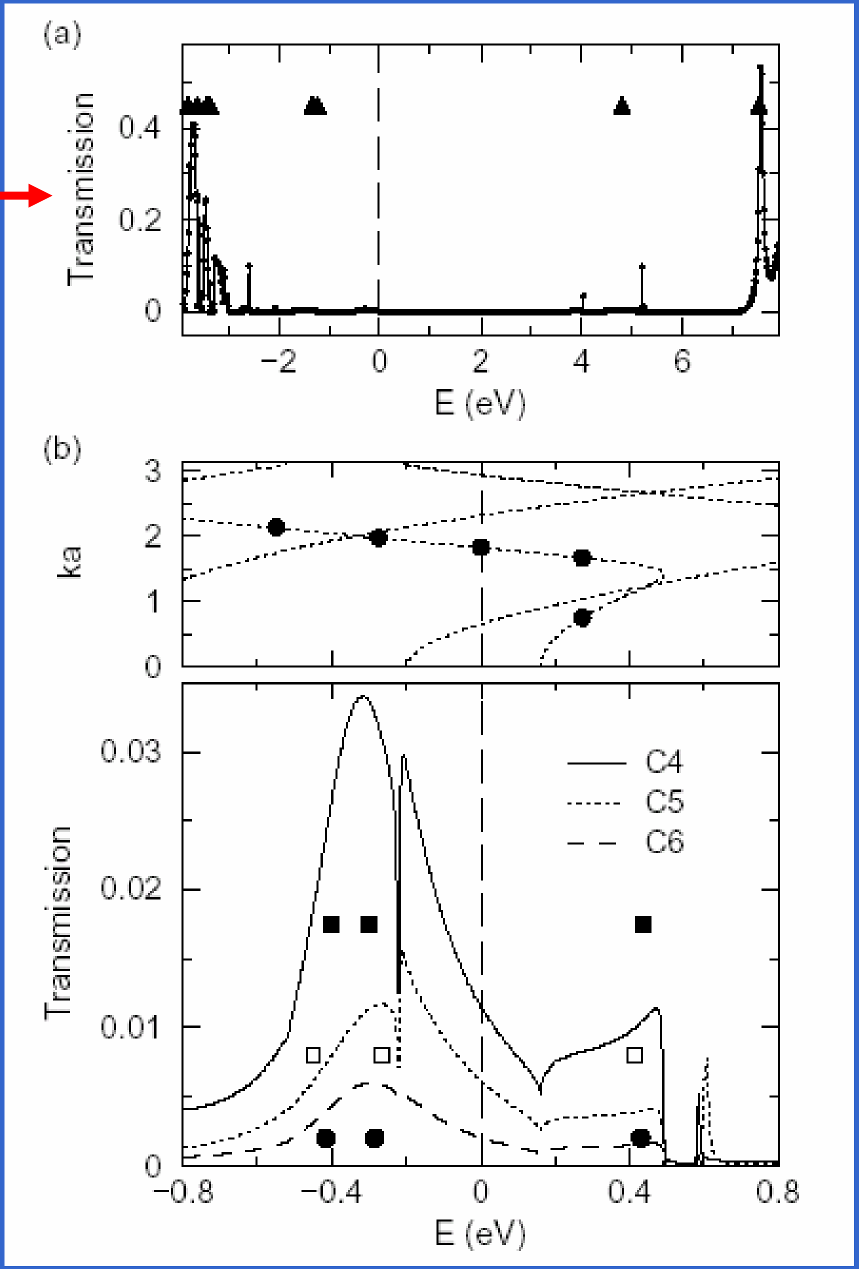
2. Engellkes et al (Frisbie lab) (2003):

   Xu and Tao, Science (2003):

   Lee and Reed, J. Phys. Chem (2004):

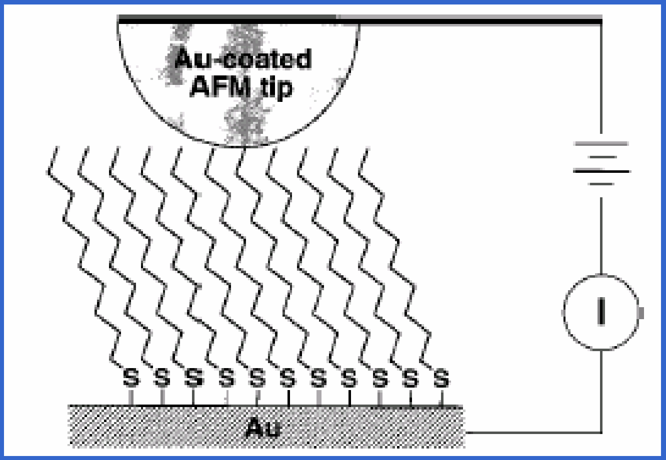
$\beta = 1.05$

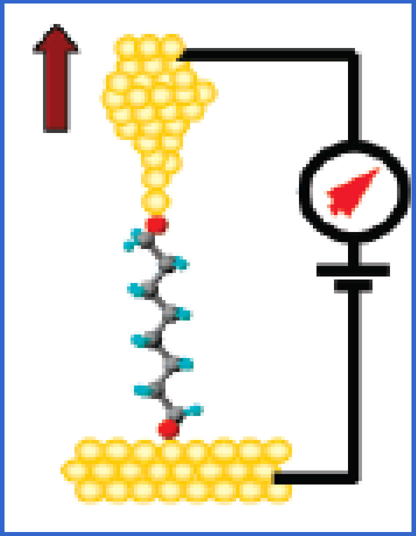
Alkane has a large HOMO-LUMO gap, ~10eV. The Fermi level is inside the gap, but closer to HOMO.



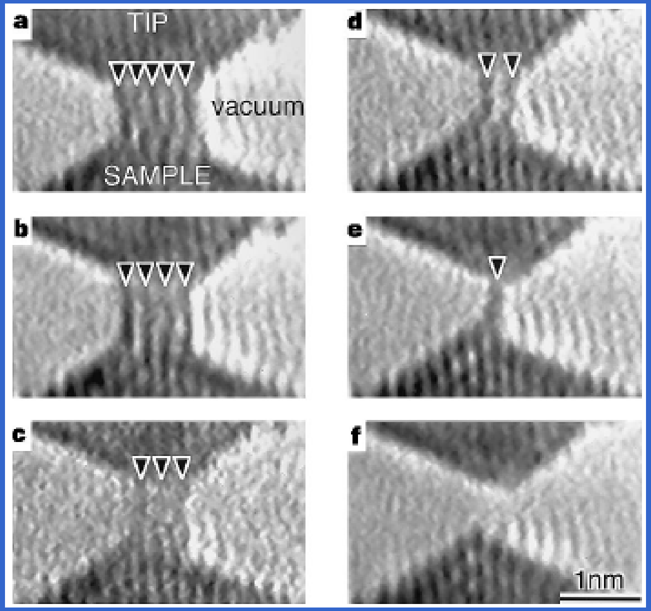
There is a tiny feature near Fermi level which determines the resistance.

3. What is the single-molecule conductance?

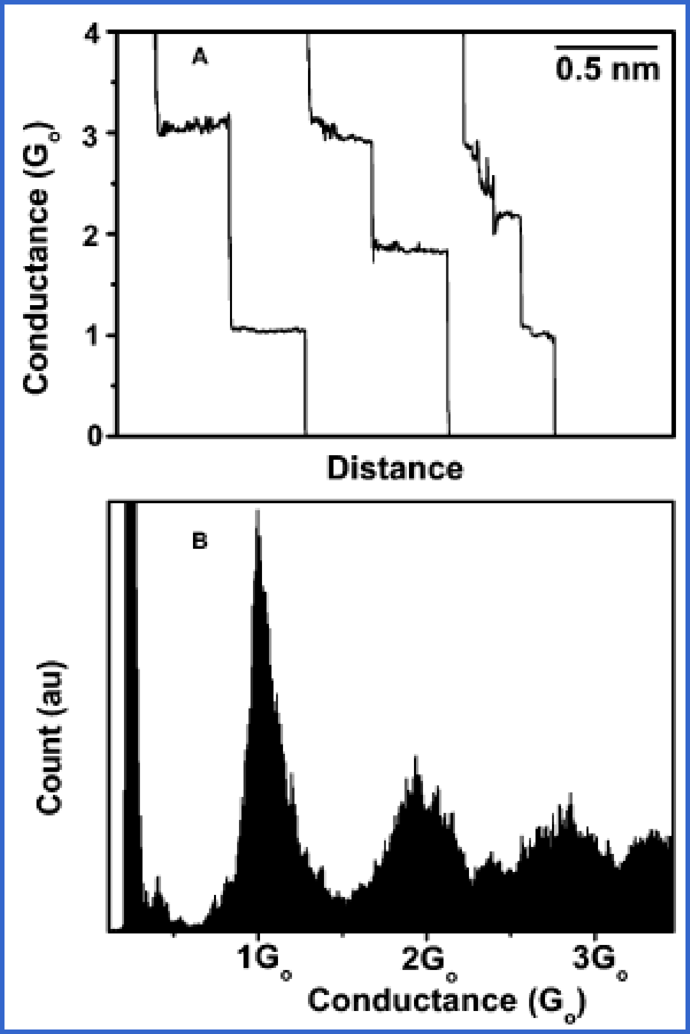




Conductance of a Au nanowire:

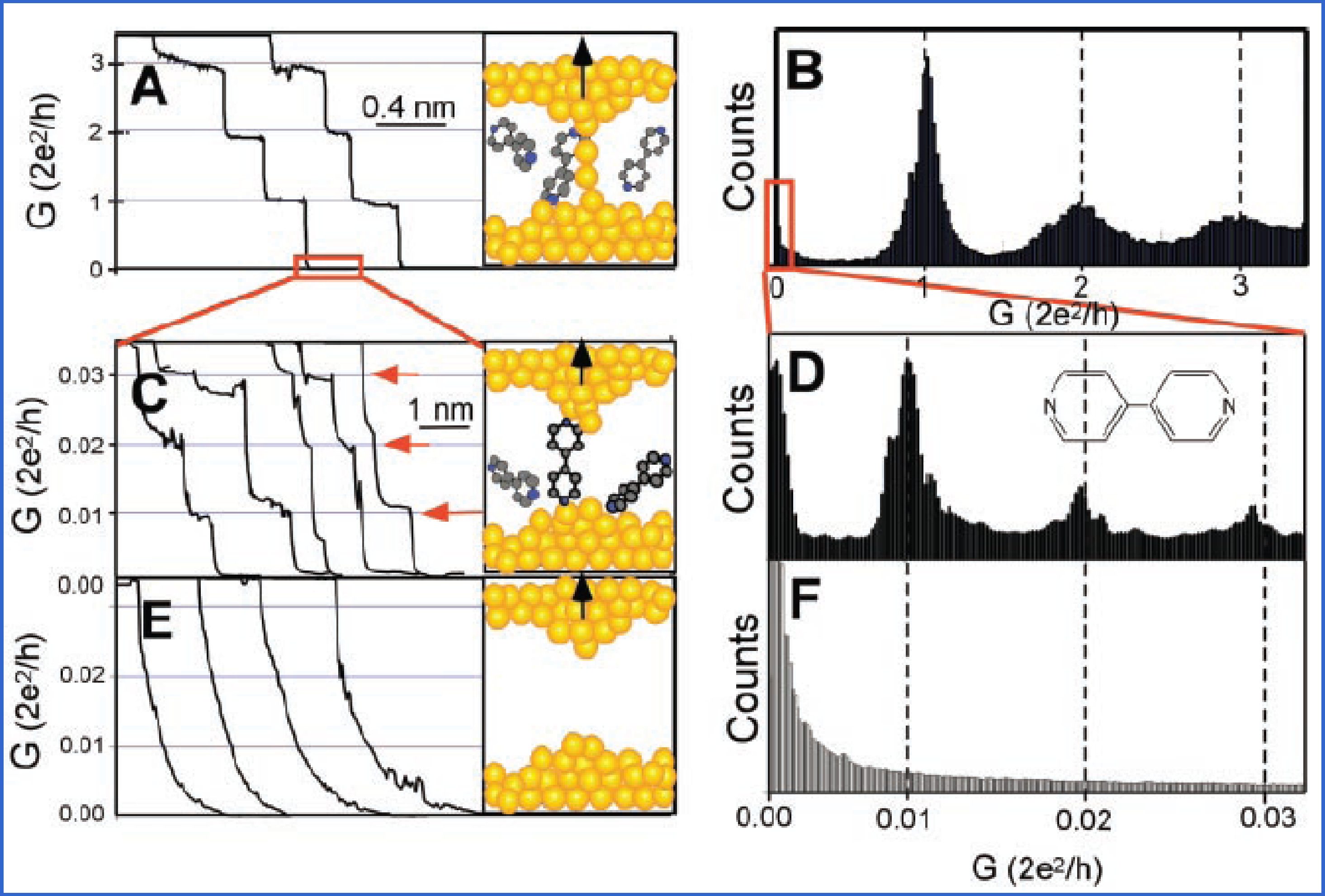


Nature 395, 780 (1998)



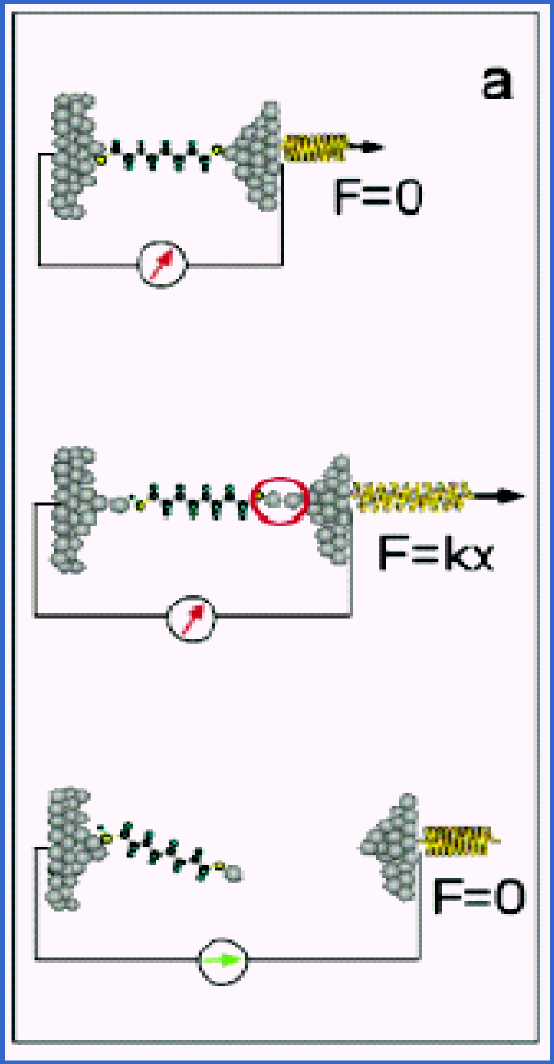
Nano Lett. 6, 2362 (2006)

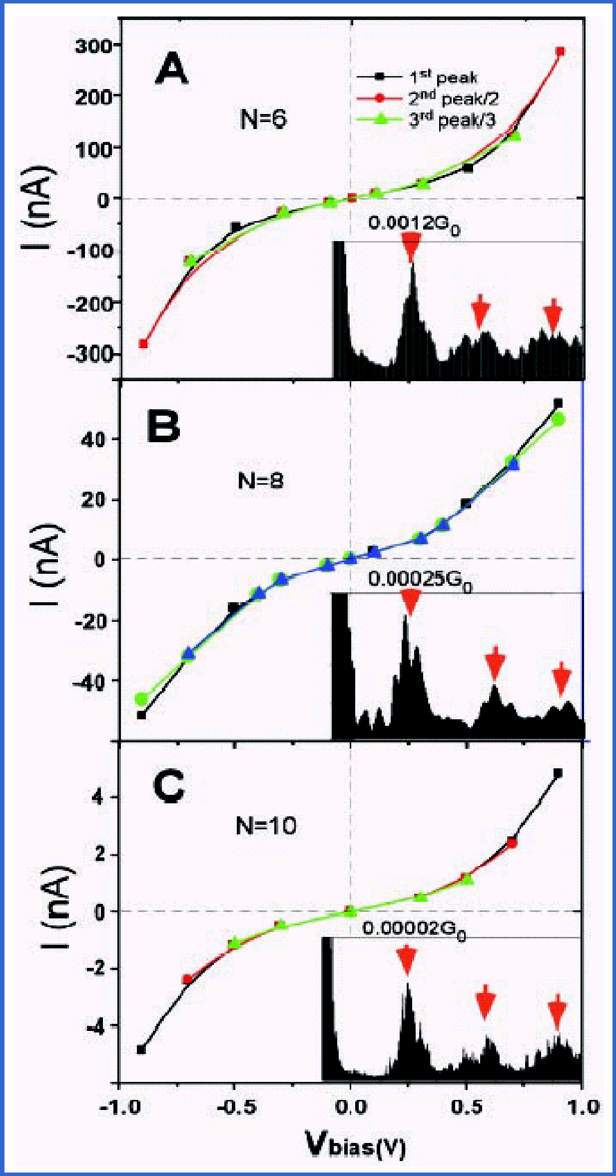
Conductance of a single molecule



J. Tao et al, Science (2003)

New measurement on single alkanedithiol molecule





J. Tao et al, JACS (2003); Science (2003)

Previous modeling:

PRL 95, 156803 (2005) PHYSICAL REVIEW LETTERS week ending 7 OCTOBER 2005

Conductance of an Ensemble of Molecular Wires: A Statistical Analysis

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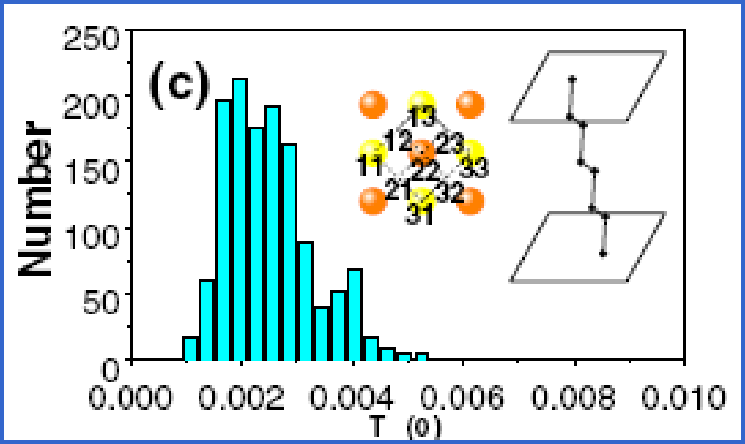
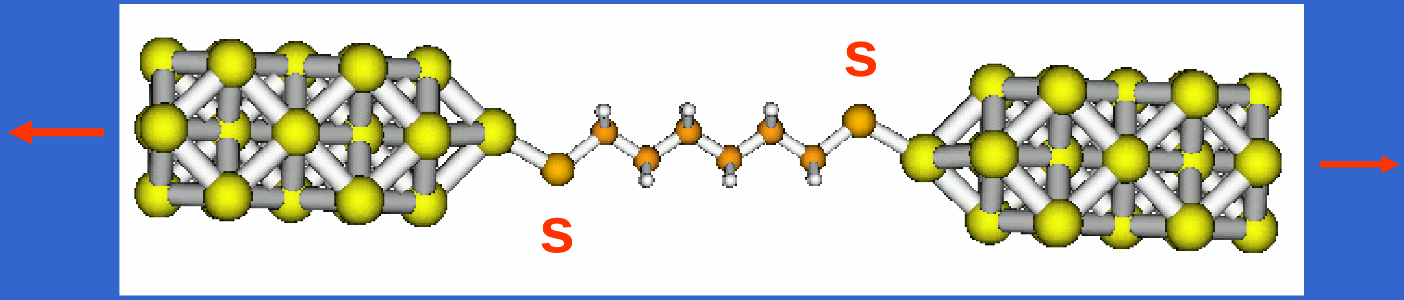


		Calculation	Experiment
N = 6	G =	0.0025	0.0012

Unit: G_0

Our model:



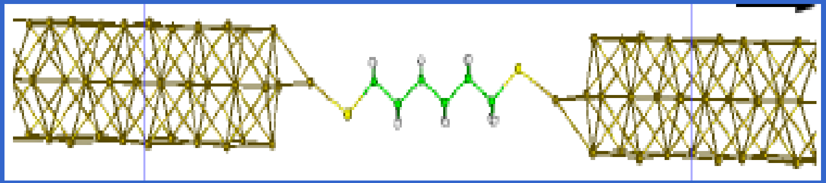
		Calculation	Experiment
N = 6	G =	0.0010	0.0012
N = 8	G =	0.000 13	0.000 25
N = 10	G =	0.000 02	0.000 02

Unit: G_0

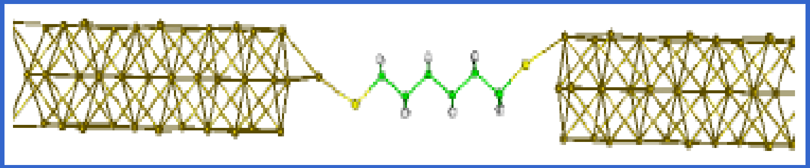
Kaun & Seideman, Phys. Rev. B 77, 033414 (2008)

Contact effect (N=6):

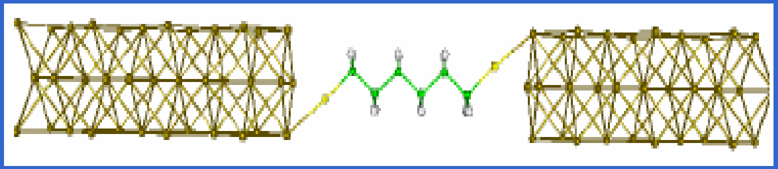
a

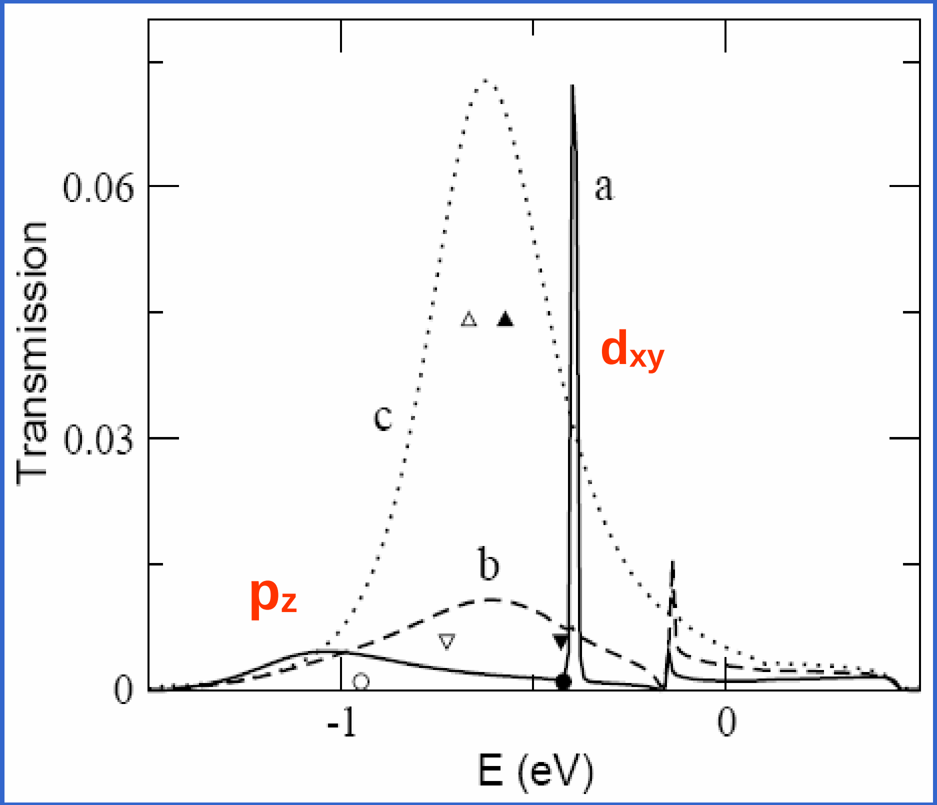


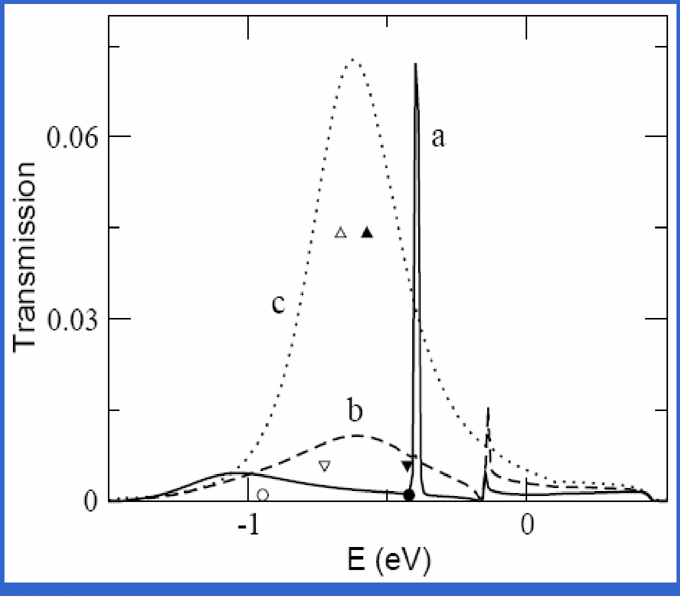
b



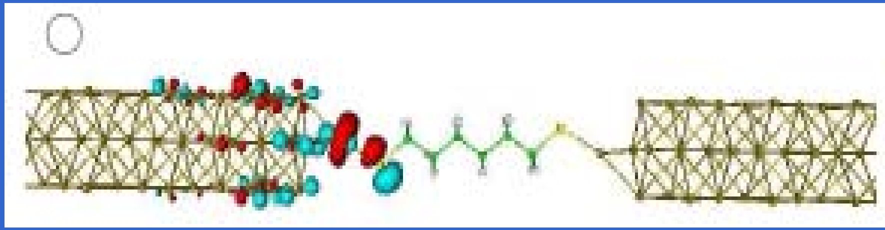
c

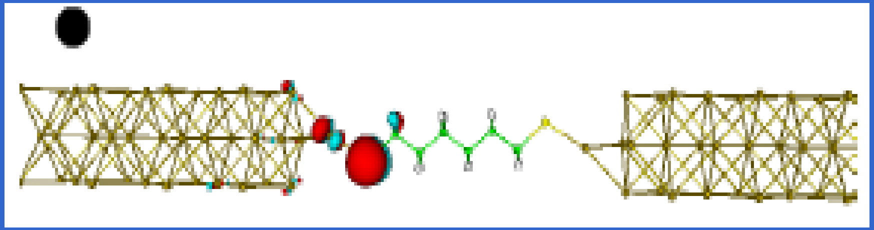


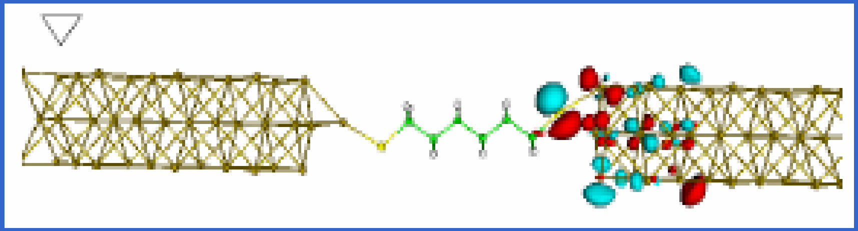


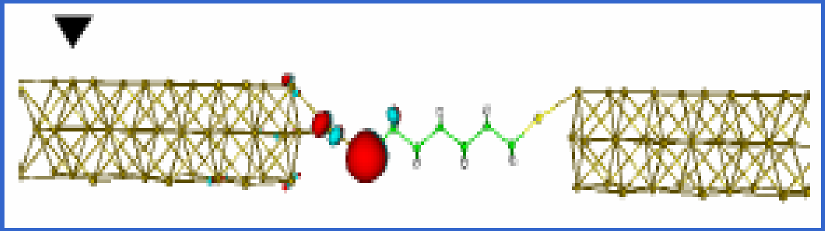


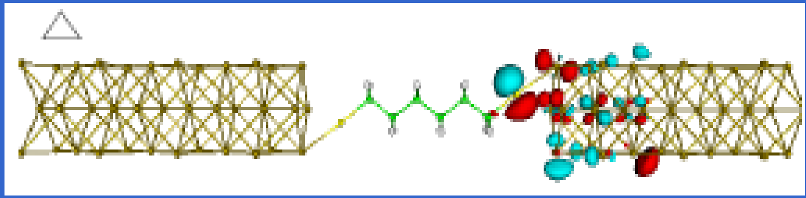
Au surface states and Au-S hybridization (from lead s, p_z band)

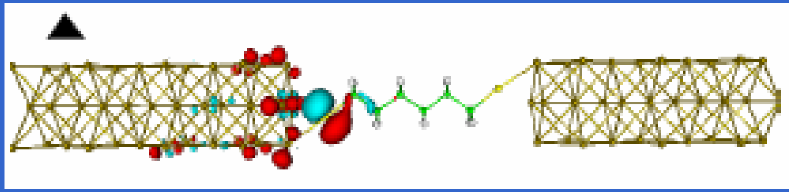




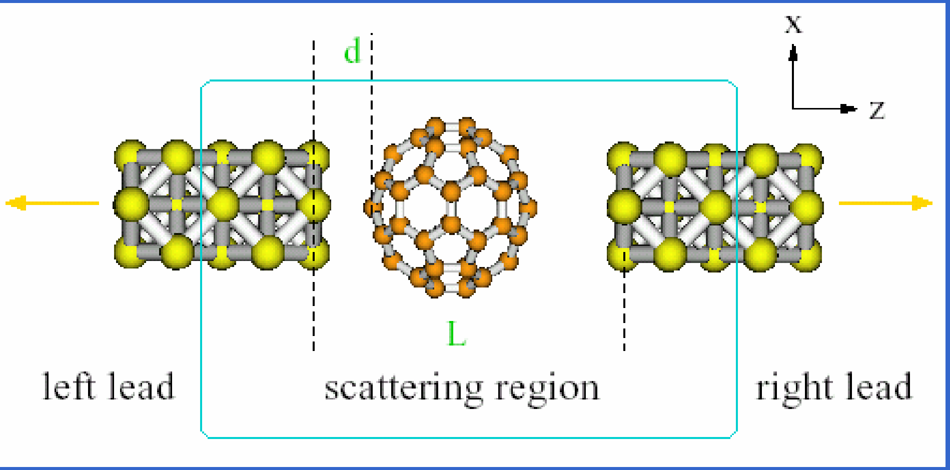




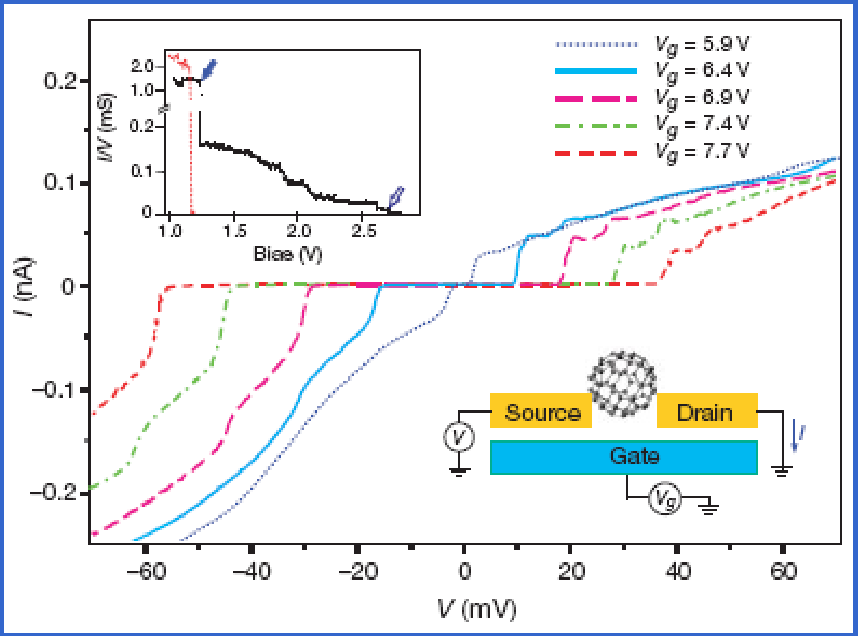


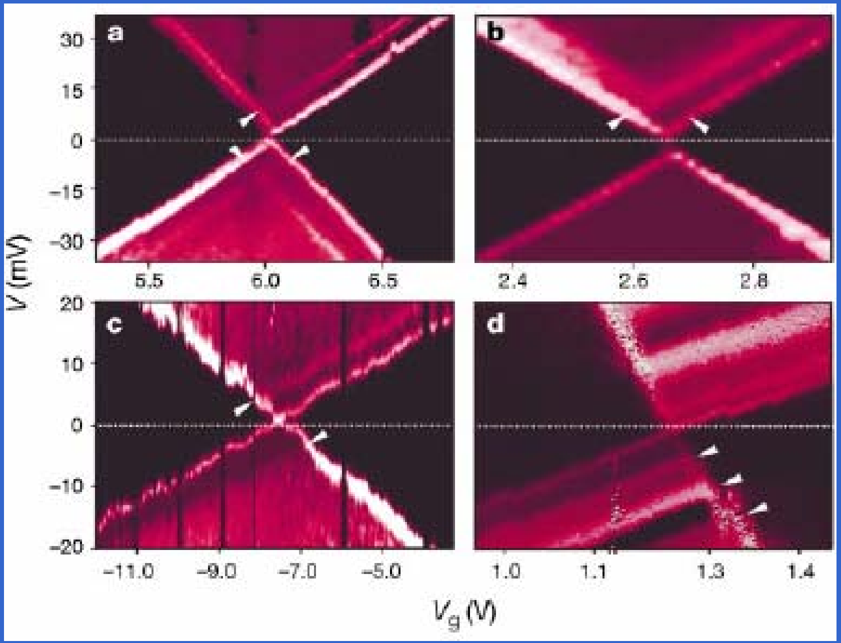


4. Spontaneous oscillation of current



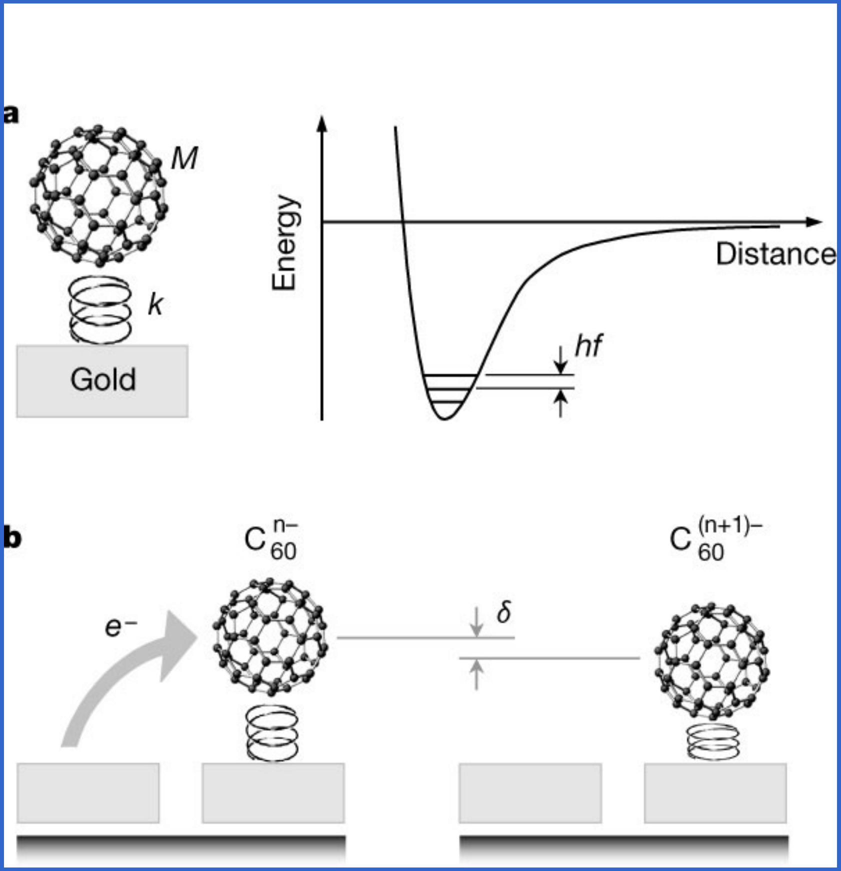
Nanomechanical oscillations in a single-C_{60} transistor





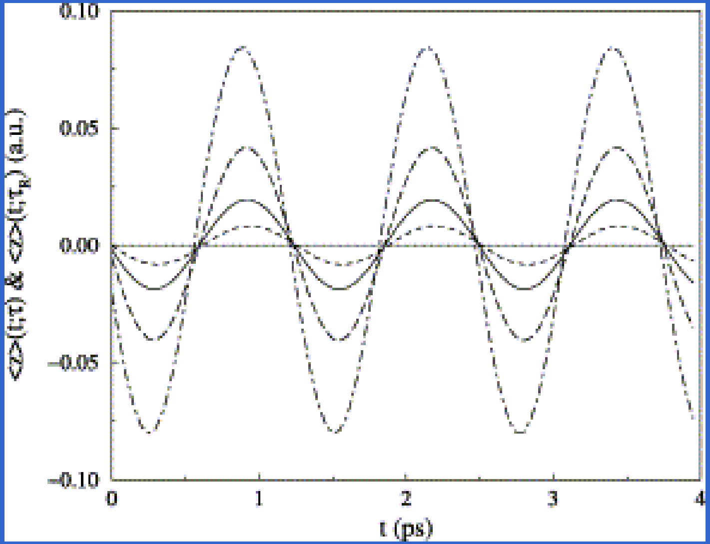
H. Park, et al, Nature (2000)

Current-driven oscillations:



The bouncing Bucky ball

H. Park, et al, Nature (2000)



Predictions from calculations

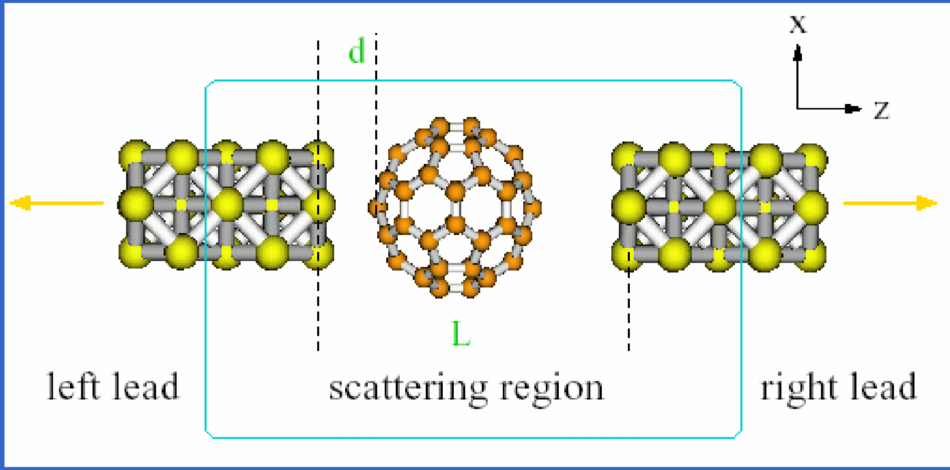
T. Seideman, et al, Chem. Phys. (2002)

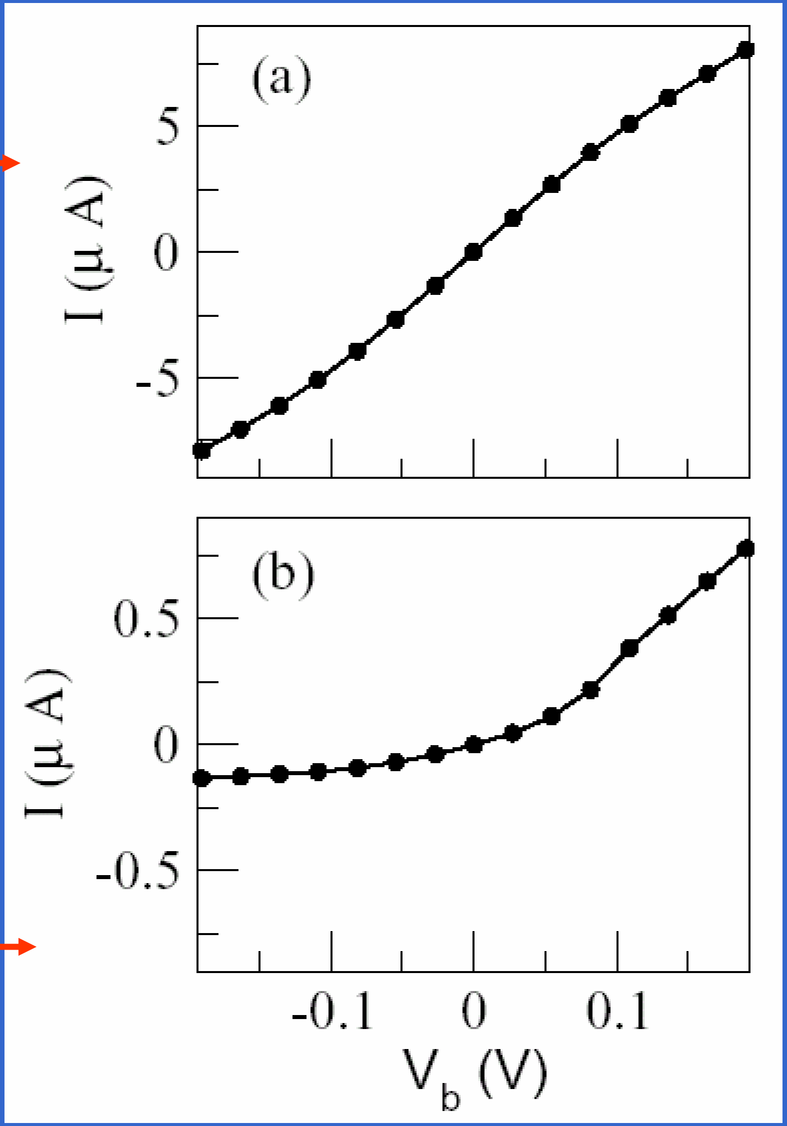
<Z> ⟵ the lifetime of resonance

f ⟵ the C_{60} mass

Our model:

Symmetric coupling

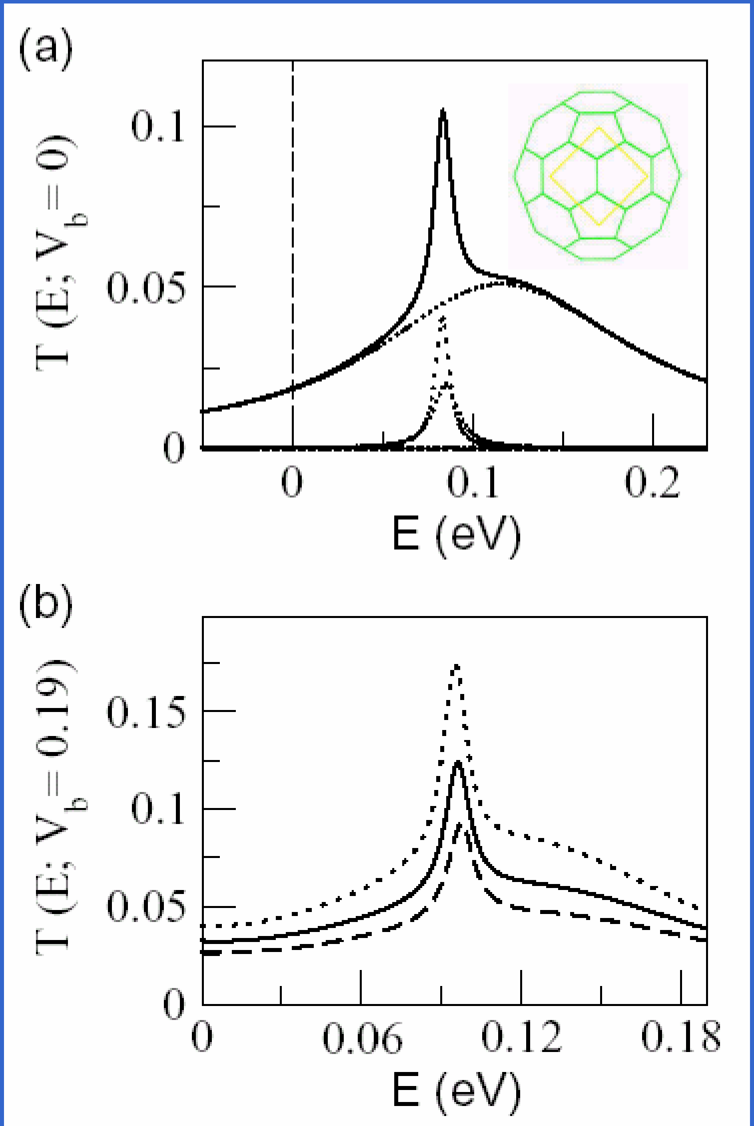




Asymmetric coupling

(L = 26.42 a.u.)

Transmission spectra:

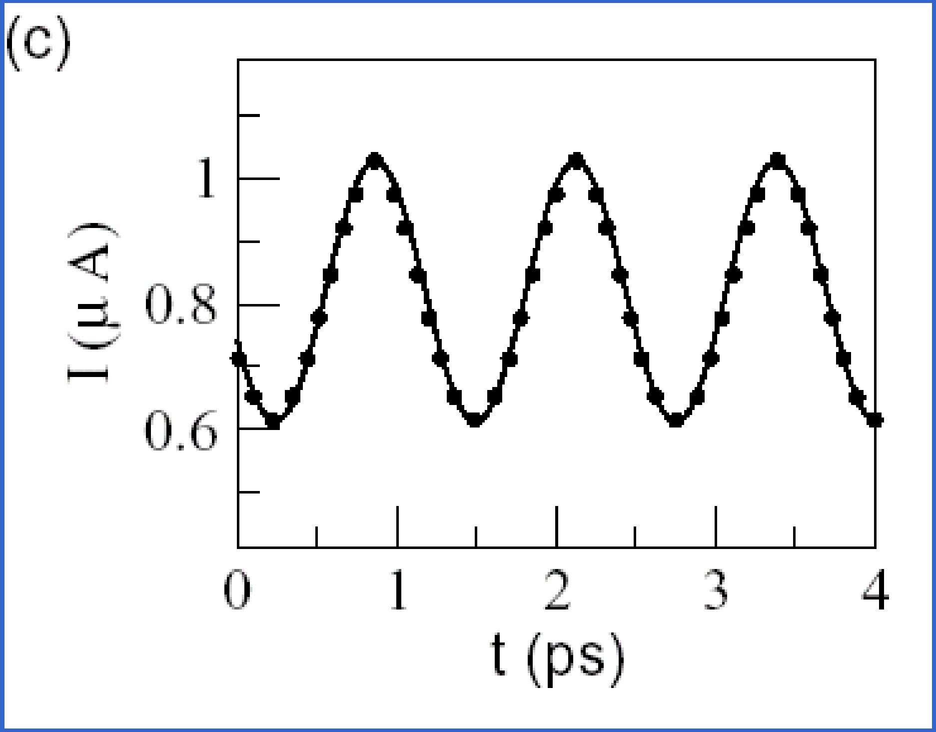


Three channels

One induces the motion; the other probes it.

Different locations

Current oscillates as the molecule vibrates



The ac/dc ratio, the power output efficiency, is 0.26 (L = 26.42 a.u.)

When L = 25.42 a.u., the ratio is 0.07

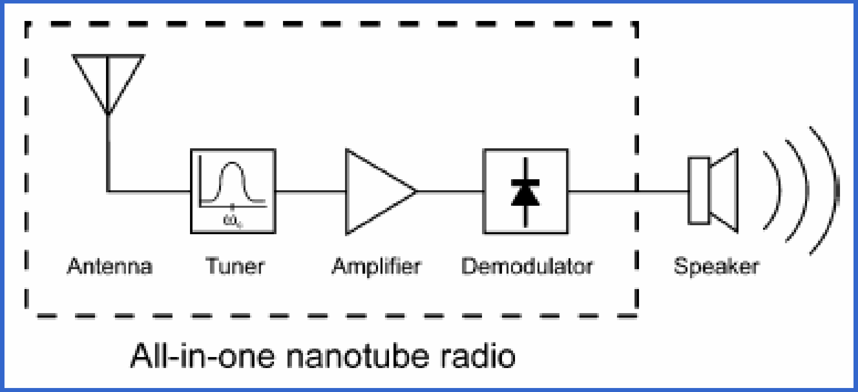
Only a range of L permits both a large ratio and high average conductance

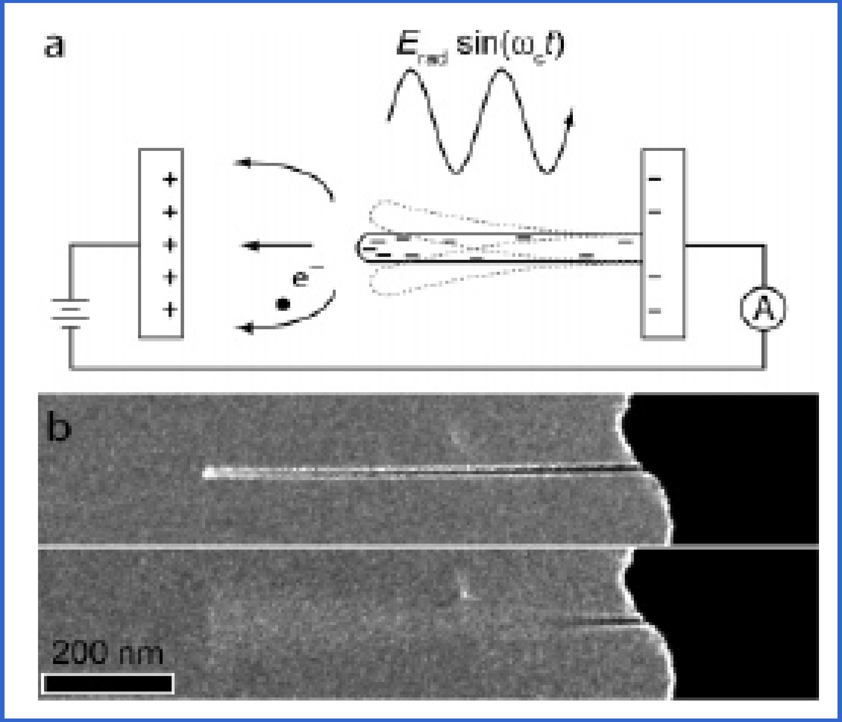
Kaun and Seideman, PRL 94, 226801 (2005)

Applications:

- A nanoscale generator of a radiation field, thus a THz optoelectronic device.
- A miniature mass spectrometry.
- The direct, time-domain probing of the current-driven dynamics in nanojunctions.

Experimentally ....   Nanotube radio





A. Zettl et al, Nano Lett. 7, 3508 (2007)

5. Sumary:

- Conductance are quantitative consistent to experimental data
- Contacts play important roles
- Au-S hybridization states dominates the conduction
- Current-driven dynamics can be used to produce oscillating current in molecular junctions

Acknowledgements:

- Prof. Tamar Seideman Northwestern Univ.
- Prof. Hong Guo McGill Univ.