Guest lecture: Atomistic Modeling of Nanoelectronics

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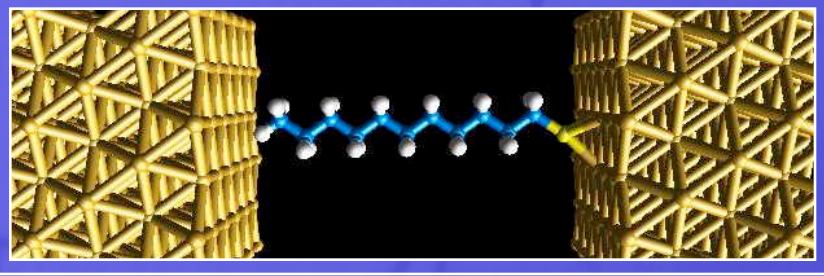
- General introduction to nanoelectronic device physics
- NEGF-DFT approach to atomic modeling of quantum transport
- Spin injection in magnetic tunnelling junctions
- Transient current and AC: progress report
- Summary



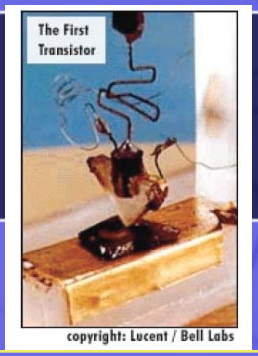
Acknowledgements:

Dr. Jeremy Taylor	AtomistiX, Denmark	mcdcal ver.1
Prof. Jian Wang	Univ. of Hong Kong	NEGF theory
Dr. Brian Larade	QuantuModeling	mcdcal ver.2
Dr. Derek Waldron	McGill	matdcal
Dr. Pawel Pomorski.	Western Ontario	capacitance
Dr. Hatem Mehrez	General Electric	nanotube devices
Dr. Devrim Guclu	Cornell University	Quantum dots
Prof. C.C. Kaun	Academic Sinica, Taipei	Molecular wires
Joseph Maciejko	Stanford	transient theory
Dr. Nikolai Sergueev	UT-Austin	electron-phonon at nonequilibrium
Dr. Dan Roubtsov	Chalk River National Lab	inelastic current
Prof. Q.R. Zheng	Grad. School, CAS, Beijing	Molecular electronics
Prof. Wengang Lu	Institute of Physics, CAS, Beijing	nanotubes and molecular wires
Dr. C. C. Wan, Dr. N.J. Zhu	Synopsis	Transport theory

Guo group: Tao Ji, Ning, Ke, Massen, Smeu, Feng, MacGuire, Eric Zhu, Timochevski, Liu, YB Hu, Wei Ji, Zahid, Arijit Sen

Paul Haney, Prof. Allan MacDonald (UT-Austin), Prof. Xia Ke (IoP) spintronics

Nano-fabrication (top-to-bottom)

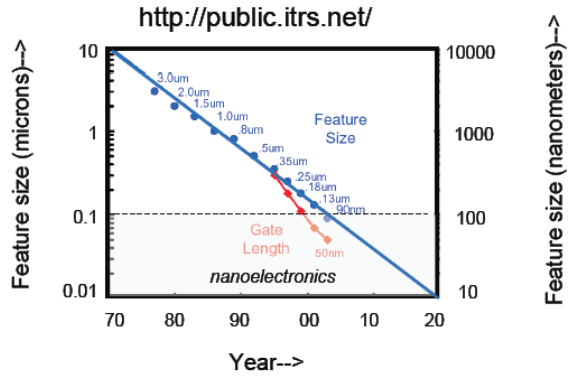


1st transistor was born in , size is $1cm^2$

During the development of microelectronics, Electronics Design Automation (EDA), namely modeling, played a important role.

After several decades, gate length of a transistor was reduced to 45nm in 2007: there can be a hundred million or more transistors in a chip. This size is expected to reach atomic scale in about 15 years.

Moore's Law



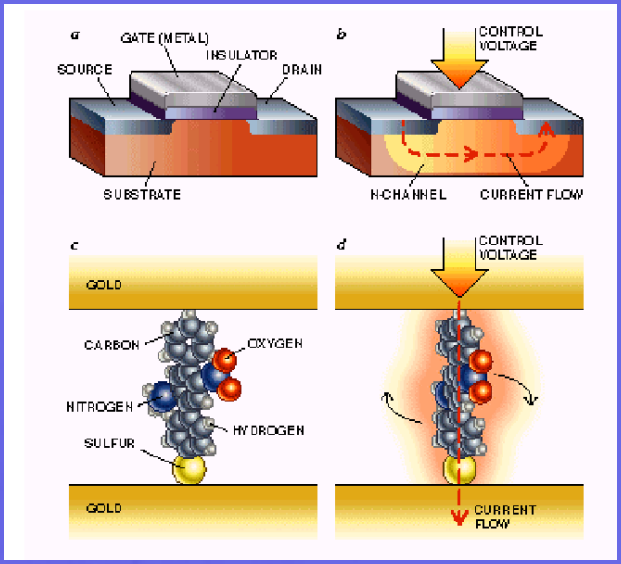
L = 6 nm (IBM, 2002)
L = 5 nm (NEC, 2003)

Traditional EDA was established on classical or semi-classical physics. At nano-scale, these theories are increasingly in adequate to deal with quantum and materials effects.

Therefore, we wish to develop a theory that can quantitatively describe nanoelectronics.

Self-assembly: bottom-to-top

In another direction, people learned how to make nanosystems by self-assembly.



Molecular electronics

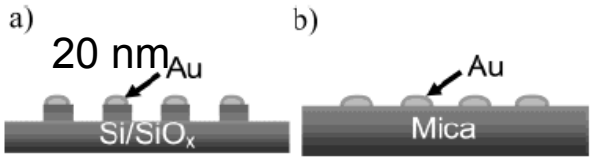


Figure 6. Schematic cross-sectional view of (a) Au islands formed on a Si/SiO_x substrate and (b) Au islands formed on a mica substrate.

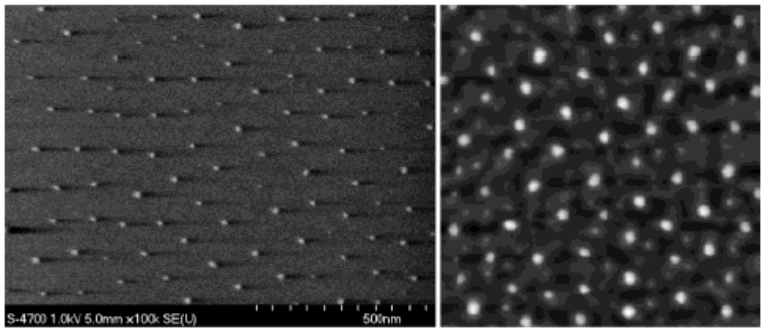
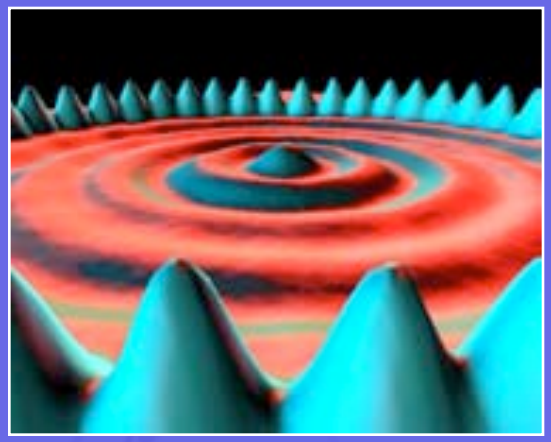


Figure 7. FEGSEM (left) and AFM (right) images of Au islands on mica. The small (<4 nm) particles randomly located between the patterned Au domains (only visible in the AFM image) are most likely remnant Au particles left behind after the ion-milling process.

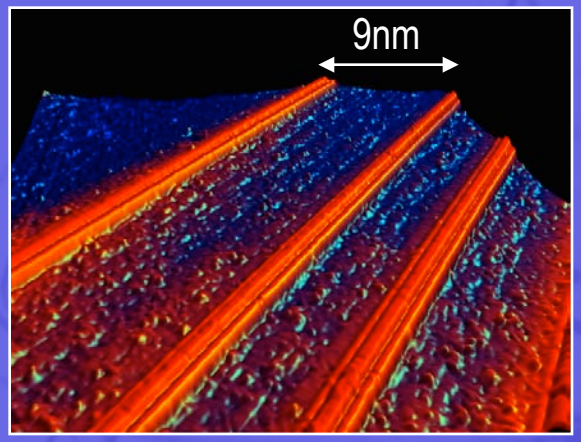
Meli and Lennox, Langmuir 19, 9097 (2003)

How do we make predictions in these molecular systems? Need a quantitative theory and modeling method.

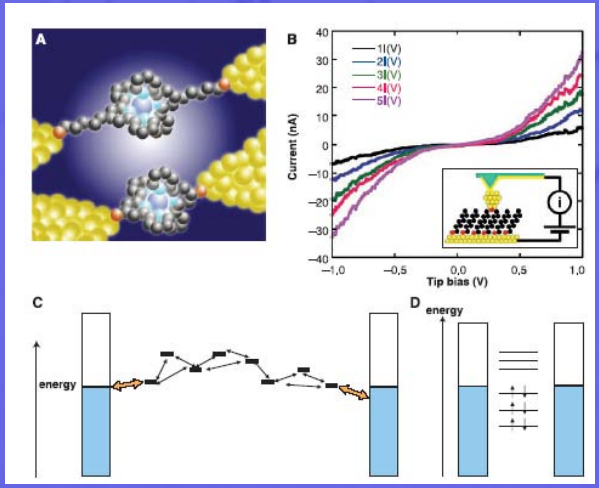
Three ingredients for nanoelectronics thoery



1. Quantum physics Eigler (IBM)



2. Materials physics Williams (HP)

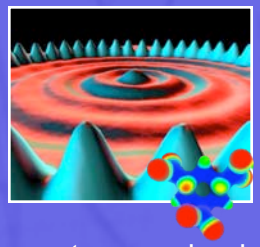


3. Nonequilibrium physics (picture from Ratner)

To make quantitative predictions without phenomenological parameter, a formalism is needed that includes these ingredients. It should also be calculable as required by the device community.

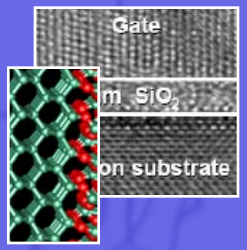
The goal of nanoelectronic device theory

- Starting from quantum mechanical first principles, calculate device Hamiltonian and potential including all atomic/chemical/materials details of the device under external bias potentials, make quantitative predictions on transport features of realistic devices.

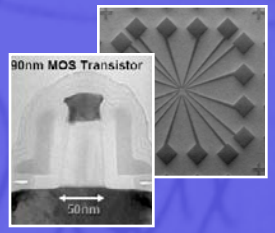


quantum mechanics
Physics

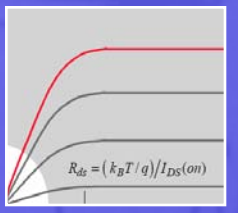
atomic simulations
materials, chemistry,
physics



device modeling
< 50nm (1000 atoms)



large scale
device modeling



device parameters

Nano-EDA

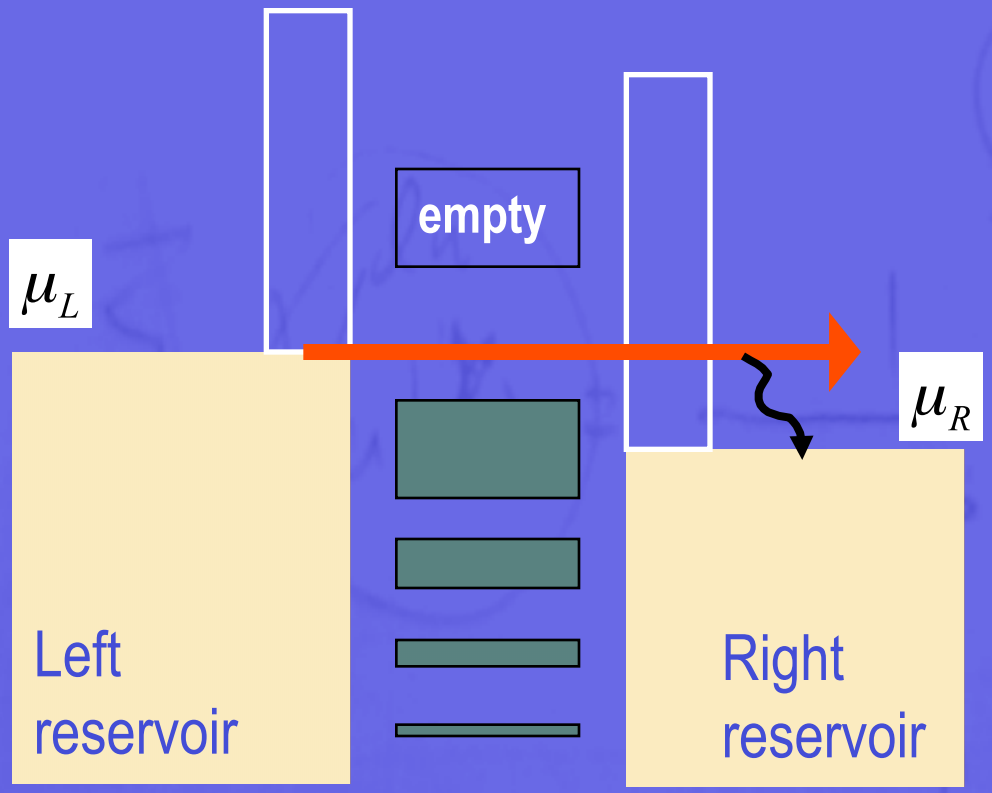
Goal of nanoelectronic device theory is along this line

Steady-state transport within NEGF-DFT

$$H = H_{leads} + H_{device} + H_{coupling}$$

- Calculating electric current flow driven by a finite bias voltage is a non-equilibrium problem, the first physics consideration is the non-equilibrium statistics of the device scattering region.
- A second consideration is the calculation of device Hamiltonian H. H determines the energy levels of the device. How to fill these levels is given by the non-equilibrium statistics.
- Keldysh non-equilibrium Green's function (NEGF) is a natural approach to determine the non-equilibrium statistics. One may also evolve a non-equilibrium density matrix from some initial equilibrium initial condition, somehow.
- What kinds of H are used concern numerical accuracy: effective mass, kp, TB, HF, DFT, GW, QMC, CI… In the end, one has to compare with experimental data.

Transport model: Landauer theory

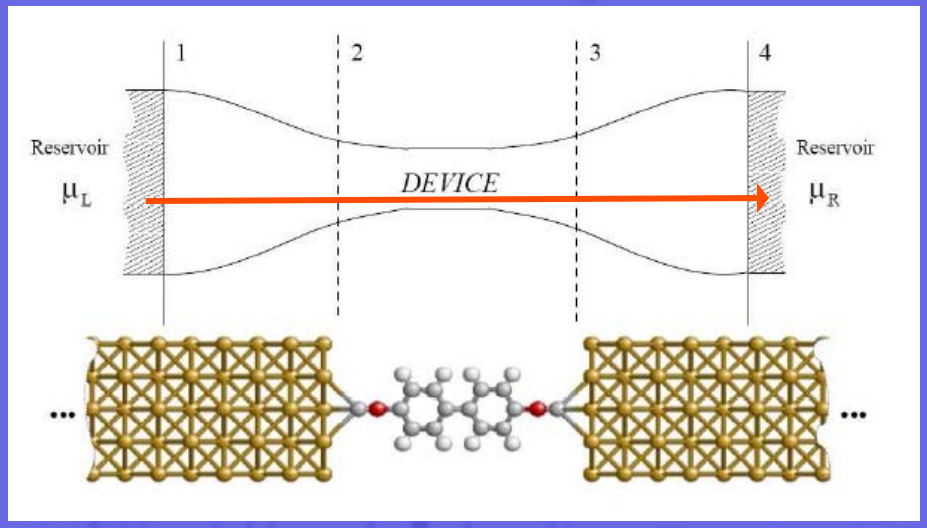


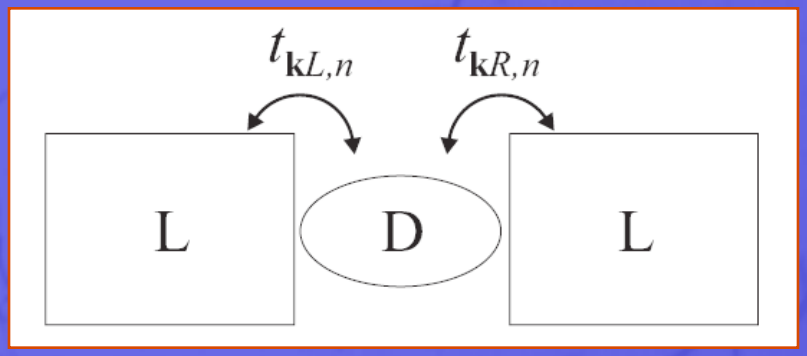
Under a voltage bias, electrons ballistically traverse the device from left to the right. They are "hot" electrons on the right, and some dissipation occurs and electrons end up inside the right reservoir.

We compute the transmission process from left to the right.

Transport : H plus reservoirs.

Again: Landauer-Buttiker transport model

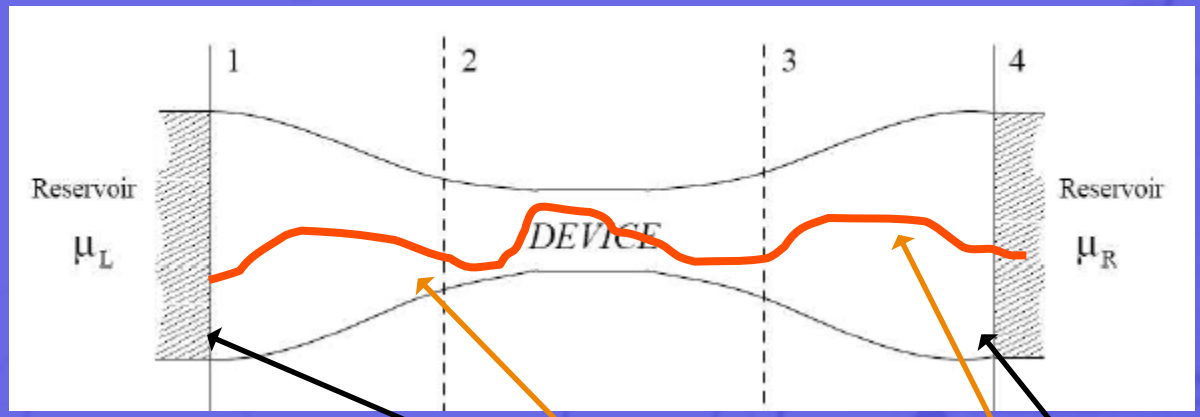




It's a scattering problem:

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

Transmission coefficients:



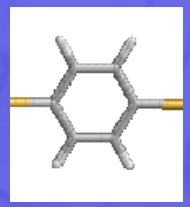
$$T(E,\Delta V) = Tr[G^r \Gamma_L G^a \Gamma_R]$$

(This is one of several ways of getting T)

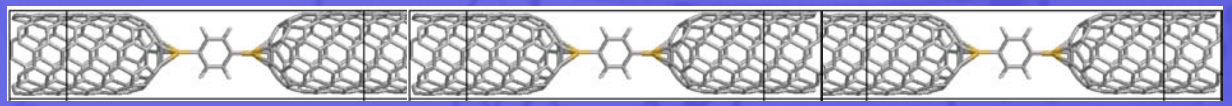
How to calculate the Hamiltonian of a device?

In order to go beyond just a few atoms, a practical way is to use DFT to calculate the device Hamiltonian.

But conventional DFT solves two kinds of problems: isolated or periodic

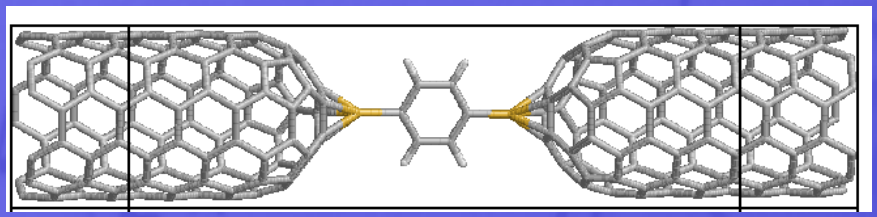


Isolated (Gaussian)

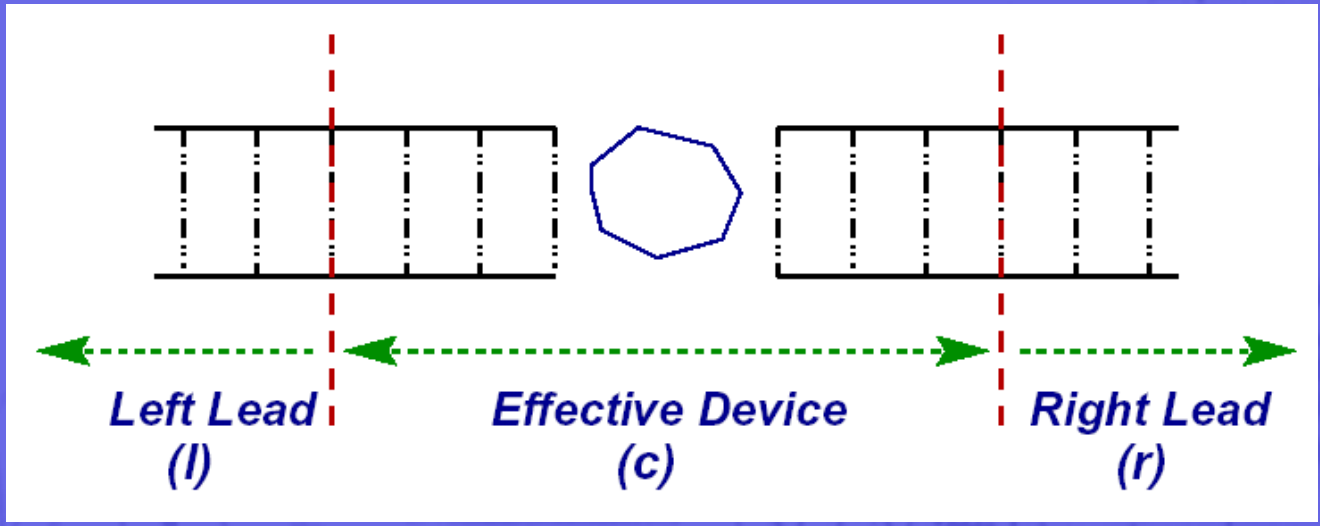


Periodic (VASP)

A device is neither isolated nor periodic, and is often at nonequilibrium.



A device is an OPEN system:

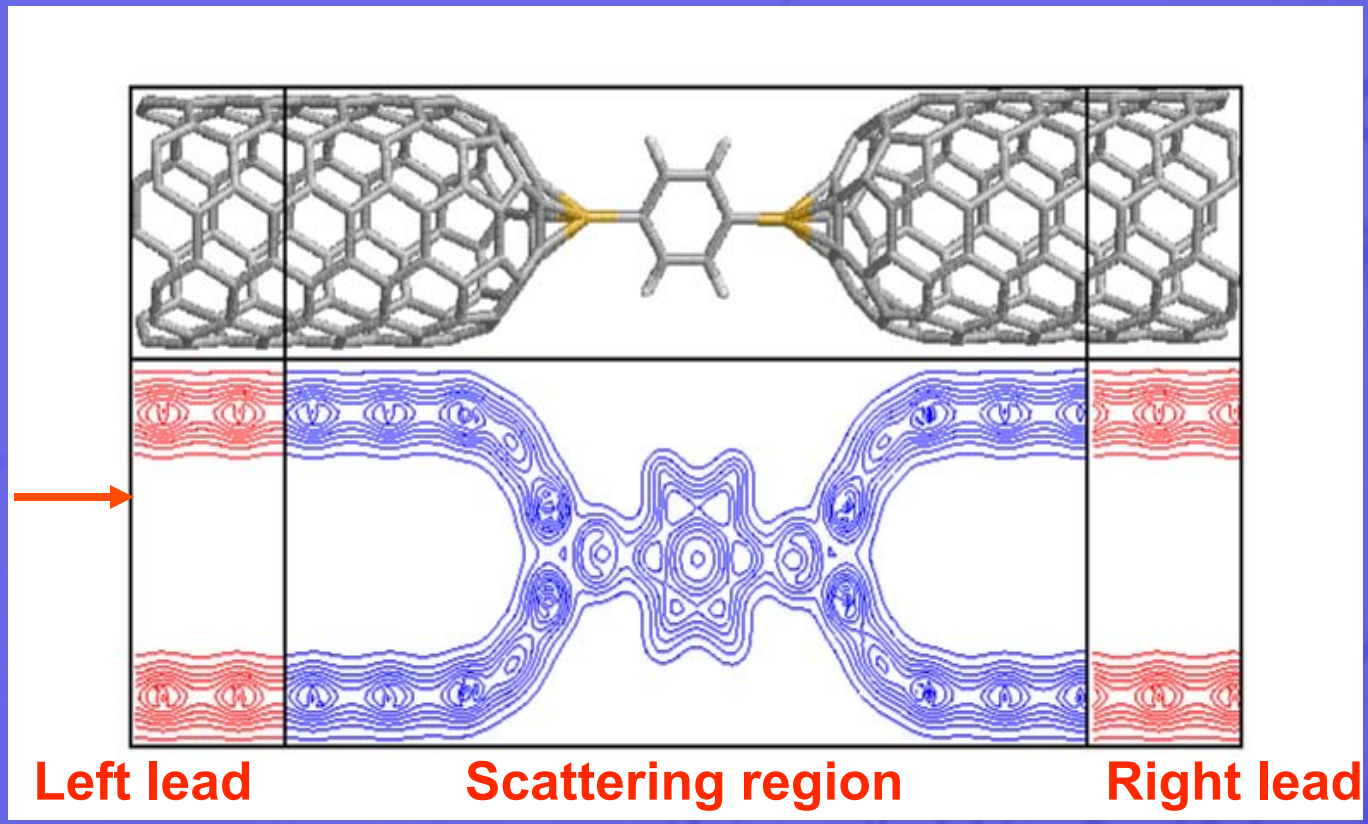


Two new problems must be solved:

1. How to reduce the infinitely large system to something calculable on a computer?
2. How to compute charge density when there is an external bias potential (non-equilibrium)?

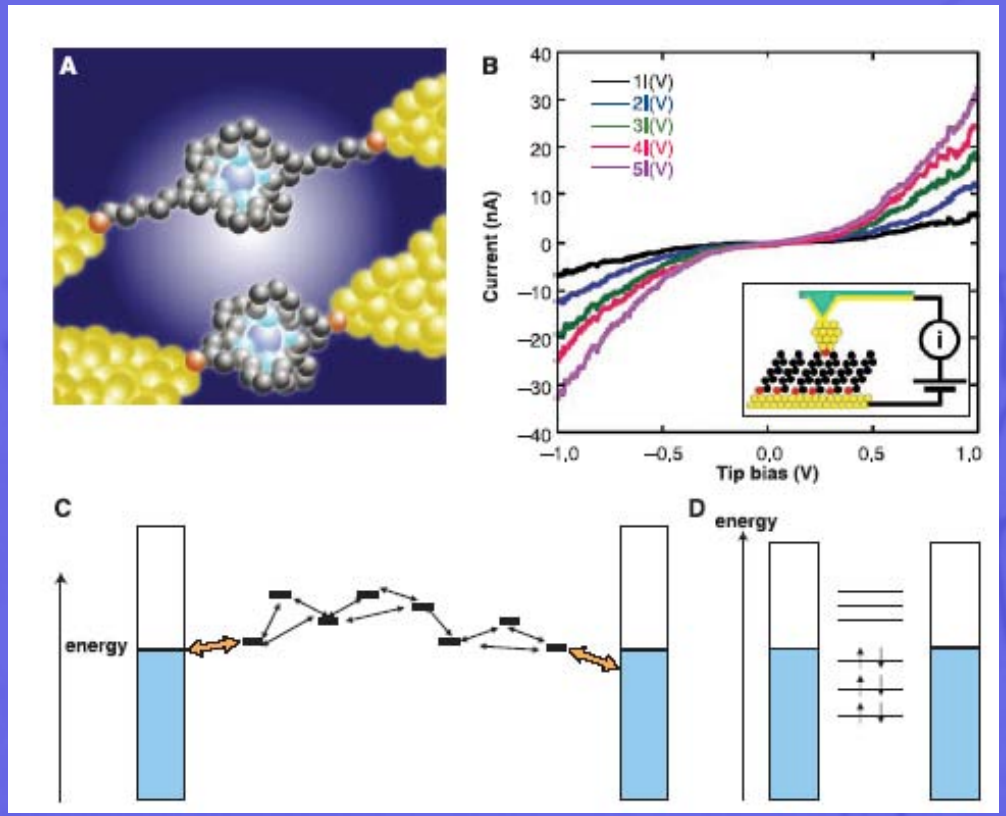
Reducing the infinitely large problem:

A device is “infinitely” large due to the large leads. We divide the system into 3 parts: left/right leads and central scattering region. By potential matching at boundaries, we reduce the problem.



Within screen approx., we only have to worry about a finite scattering region .

Nonequilibrium physics:

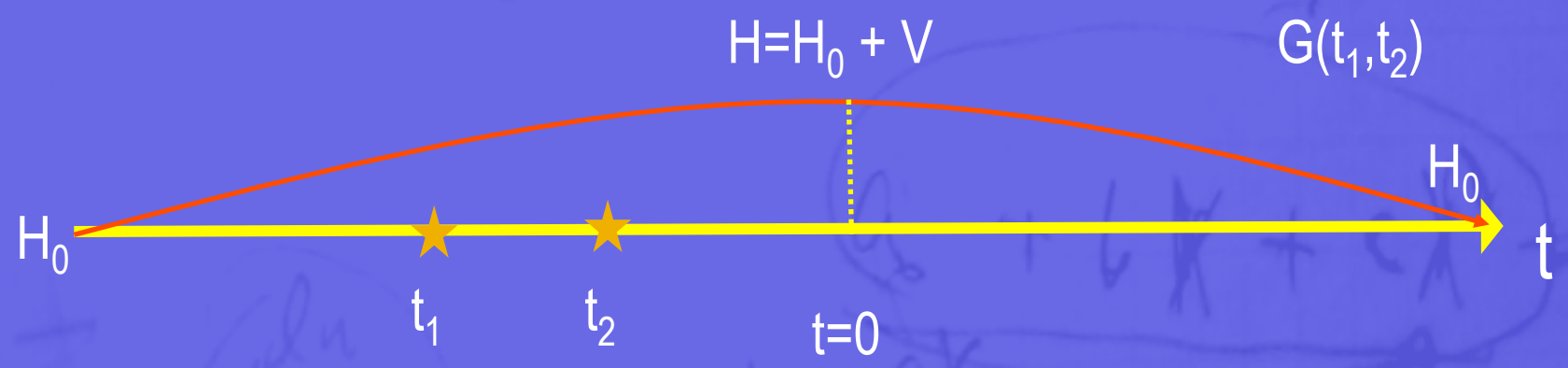


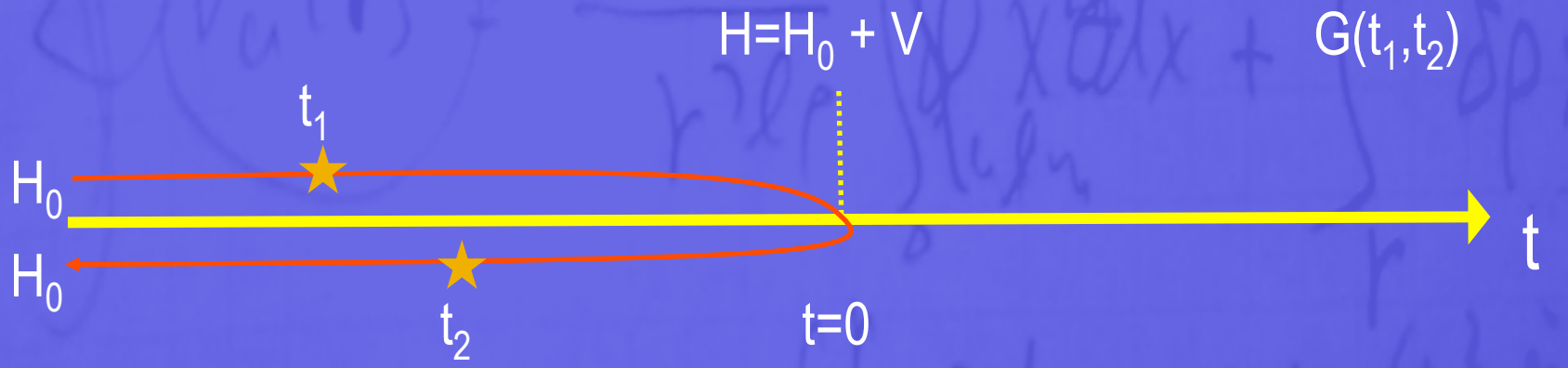
(Picture from Ratner)

After we have a Hamiltonian, we get energy levels of the device. How do we populate these levels so that we can construct a density matrix?

We use Nonequilibrium Green's functions (NEGF).

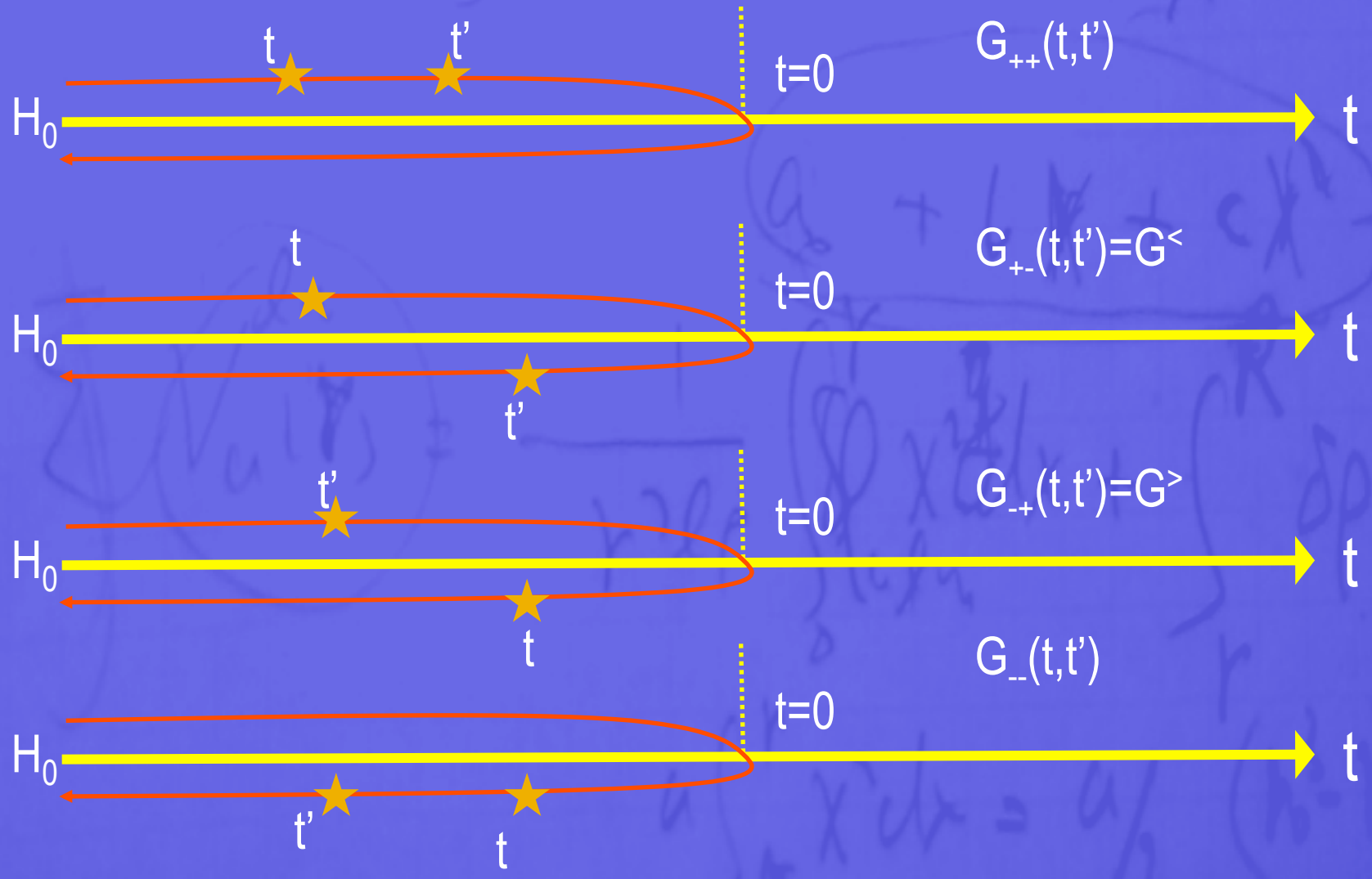
Equilibrium vs Non-equilibrium: adiabatic turning on





Book of Kadanoff and Baym, *Quantum Statistical Mechanics*

Non-equilibrium: 4 basic Green's functions

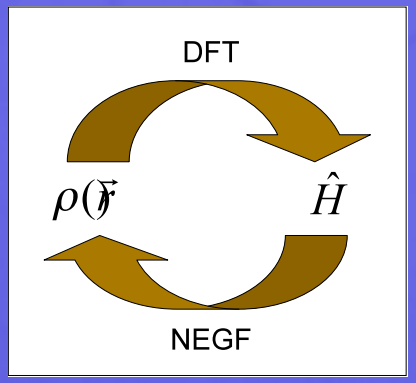


NEGF-DFT:

- Using density functional theory (DFT) to compute H of the device;
- Using Keldysh non-equilibrium Green's function (NEGF) to populate the electronic structure given by H (quantum statistics);
- Using real space numerical techniques to deal with the device boundary conditions.

Taylor, Guo, Wang, PRB 63, 245407(2001); Waldron, Haney, Larade, MacDonald, Guo, PRL 96, 166804 (2006)

NEGF: $$\hat{\rho} = \frac{1}{2\pi i}\int dE\, G^{<}$$



$$G^{<} = G^{R}\Sigma^{<}G^{A}$$

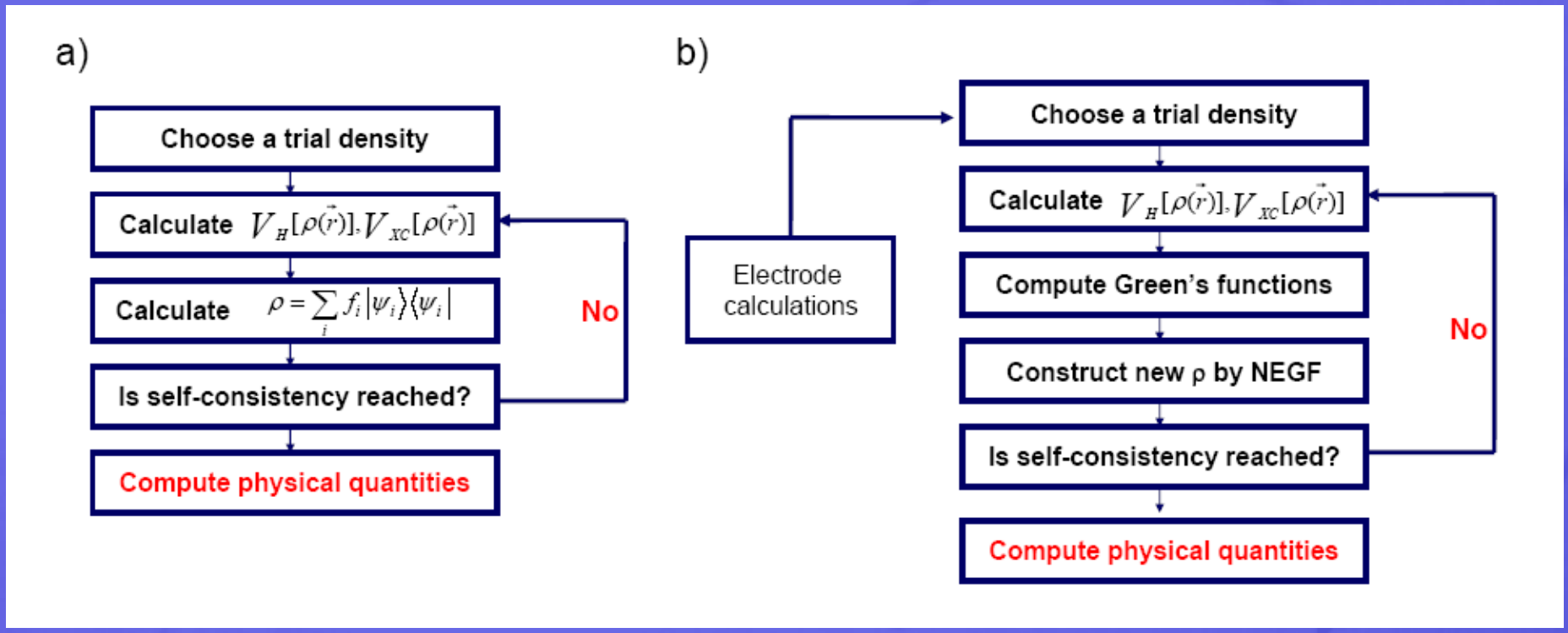
$$G^{R}(E,U) = \frac{1}{E - H_0 - eU - V_{ps} - V_{xc} - \Sigma_R}$$

$$\Sigma^{<} = i f_l(E;\mu_l)\Sigma^{R}_{l,l} + i f_r(E;\mu_r)\Sigma^{R}_{r,r}$$

$$\nabla^2 U = -4\pi\rho$$

Book of Jauho; book of Datta; Wang et al. PRL 82, 398(1999)

Algorithm implementation: DFT vs NEGF-DFT:



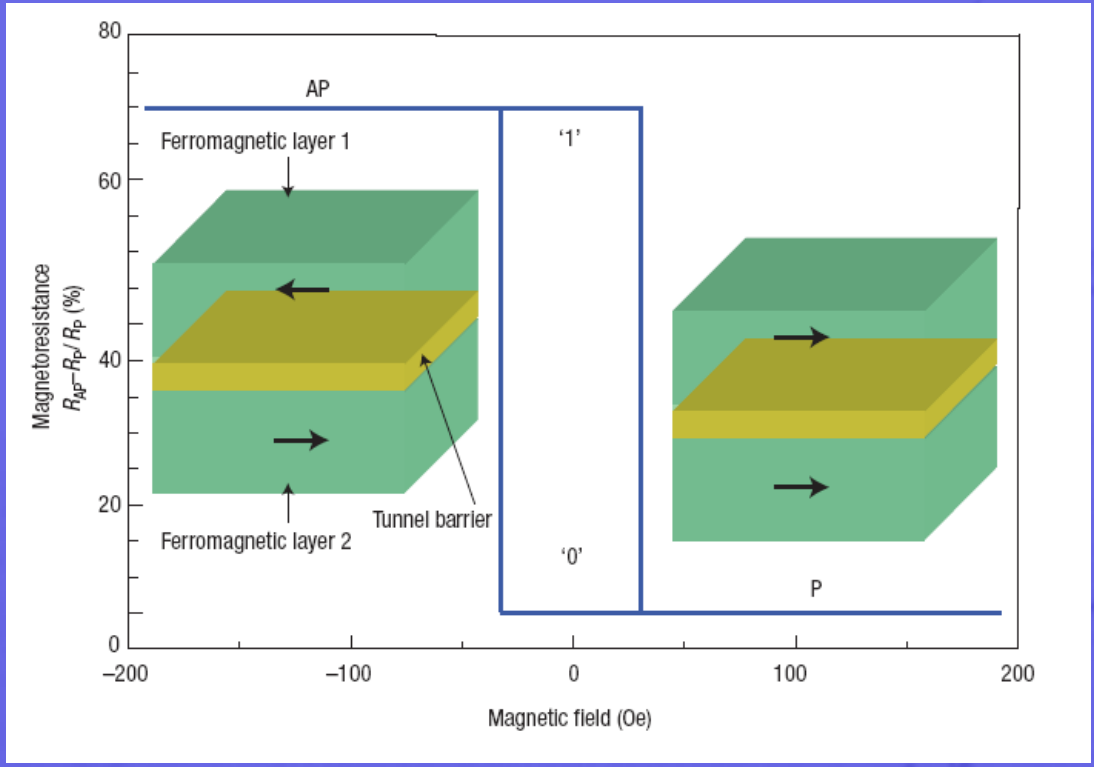
DFT NEGF-DFT

Example 1.

Spintronics: Fe/MgO/Fe magnetic tunnel junction

D. Waldron etal, PRL, 97, 226802 (2006).
P. Haney etal., PRB 76, 024404 (2007).

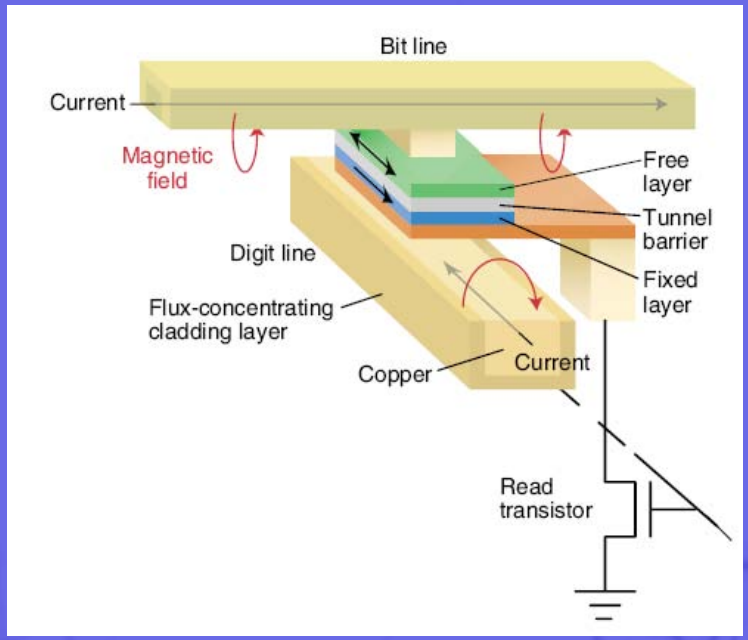
Magnetic tunneling junction (MTJ)

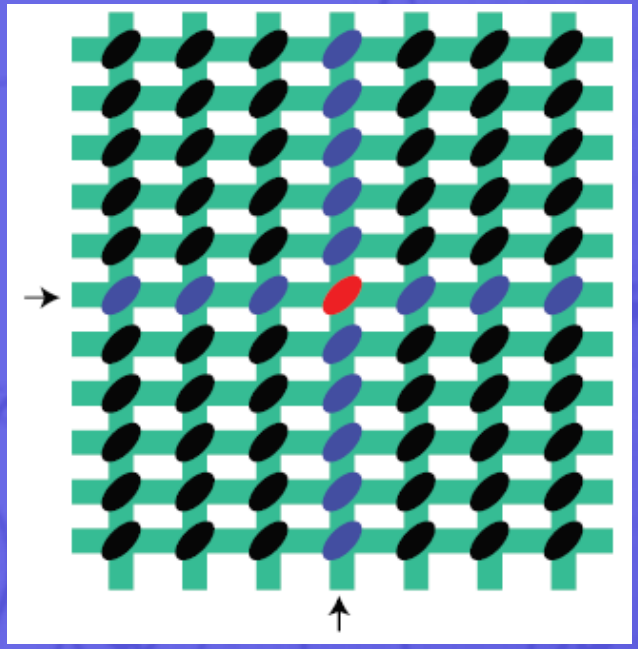


$$\text{TMR} = \frac{I_{tot}^{\uparrow\uparrow} - I_{tot}^{\uparrow\downarrow}}{I_{tot}^{\uparrow\downarrow}}$$

Picture from W. Butler, Nature Mat., 3, 845 (2004)

Why MTJ is useful ? It's a switch and can build MRAM





	Speed	Density	Power
DRAM		✓	
SRAM	✓		
FLASH			✓
MRAM	✓	✓	✓

In principle:

"Universal memory"

J. Akerman, Science, 308, p508 (2006).

4 MB MRAM commercially available (since July 2006)

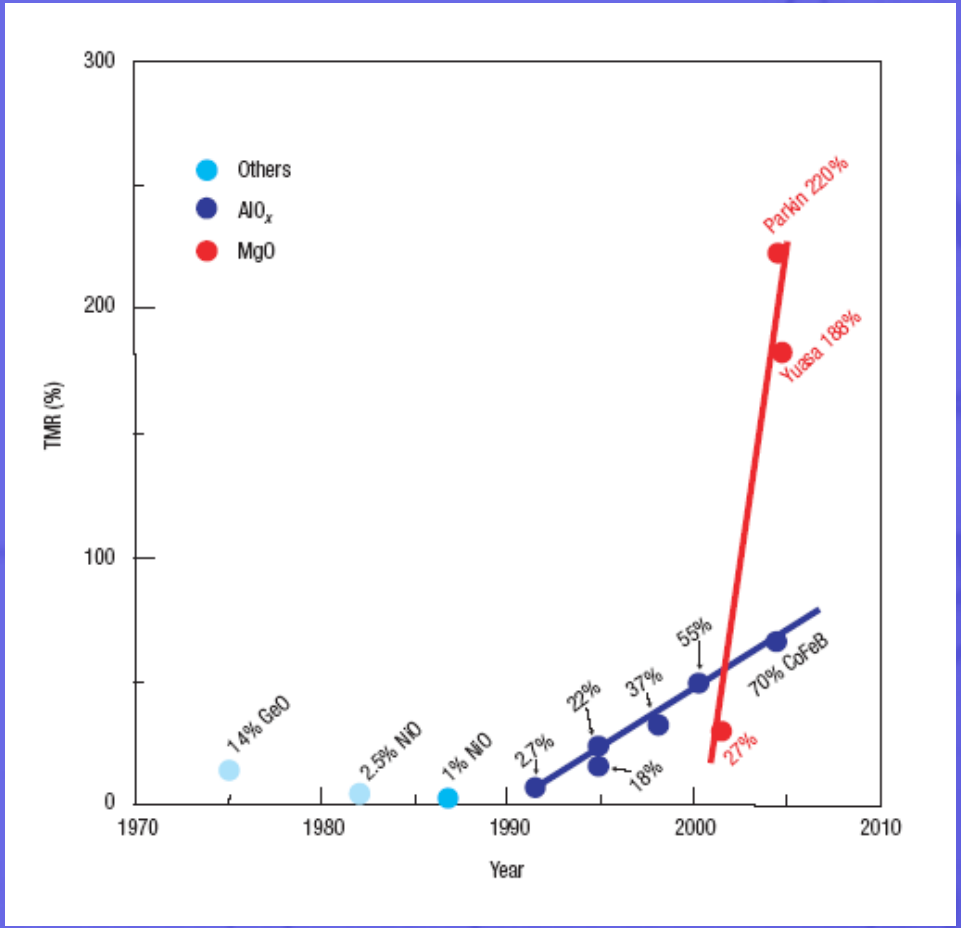
Some device merits:

- The greater the TMR, the more sensitive the device. The value of TMR appears to sensitively depend on the tunnelling structure. There is a need to understand these materials issues.
- The smaller the junction, the more devices can be packed per unit area. There is a need to understand tiny junctions made of molecules, clusters, etc.
- All of these requires calculation schemes which take into account material and chemical details of the device structures, namely NEGF-DFT.

Fe/MgO/Fe MTJ

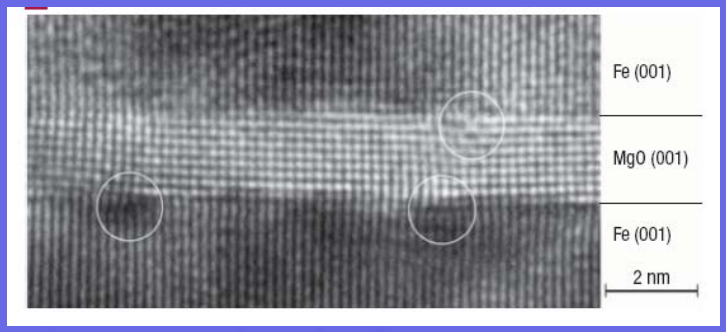
- Rapid progress on TMR ratio in recent years due to progress in materials science

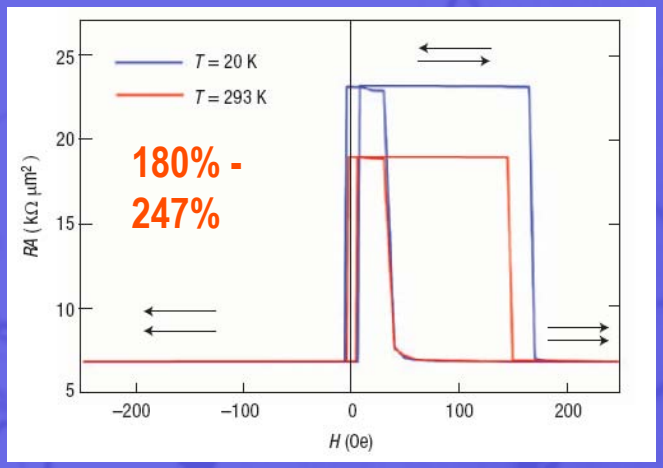
Picture from M. Coey, Nature Mat. 4, 9(2005).

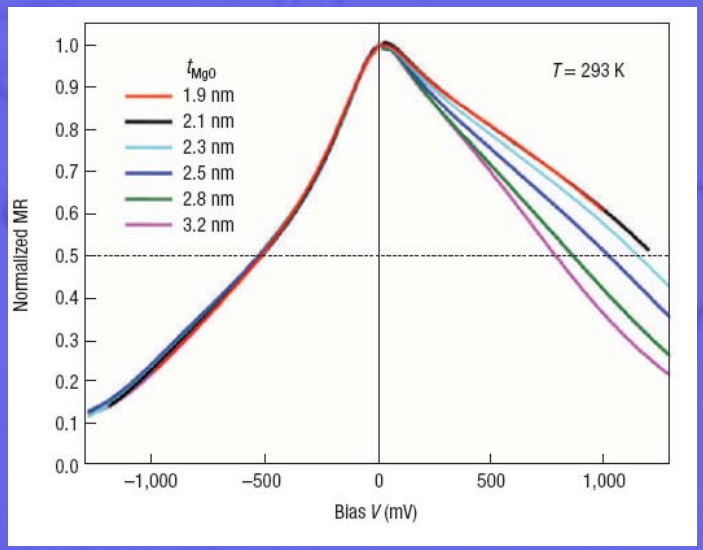


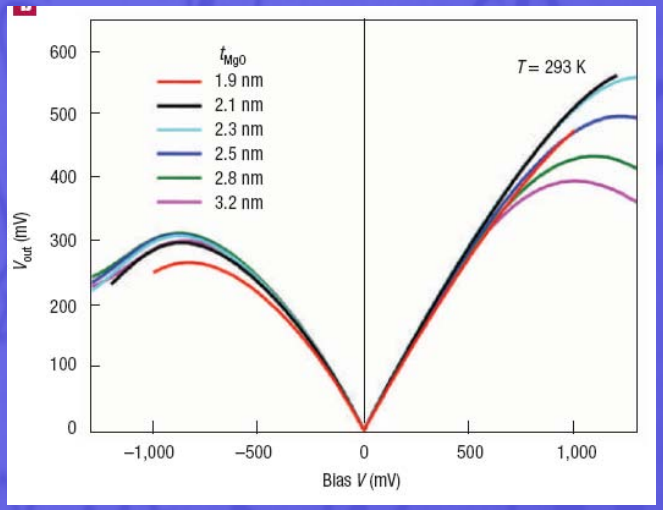
1st theory work on MgO: Butler etal PRB **63**, 054416 (2001).

Solid state device: Fe/MgO/Fe magnetic tunnel junction









Yuasa et al., Nature Mat. Vol.3, 868 (2004).

Two puzzles for Fe/MgO/Fe MTJ:

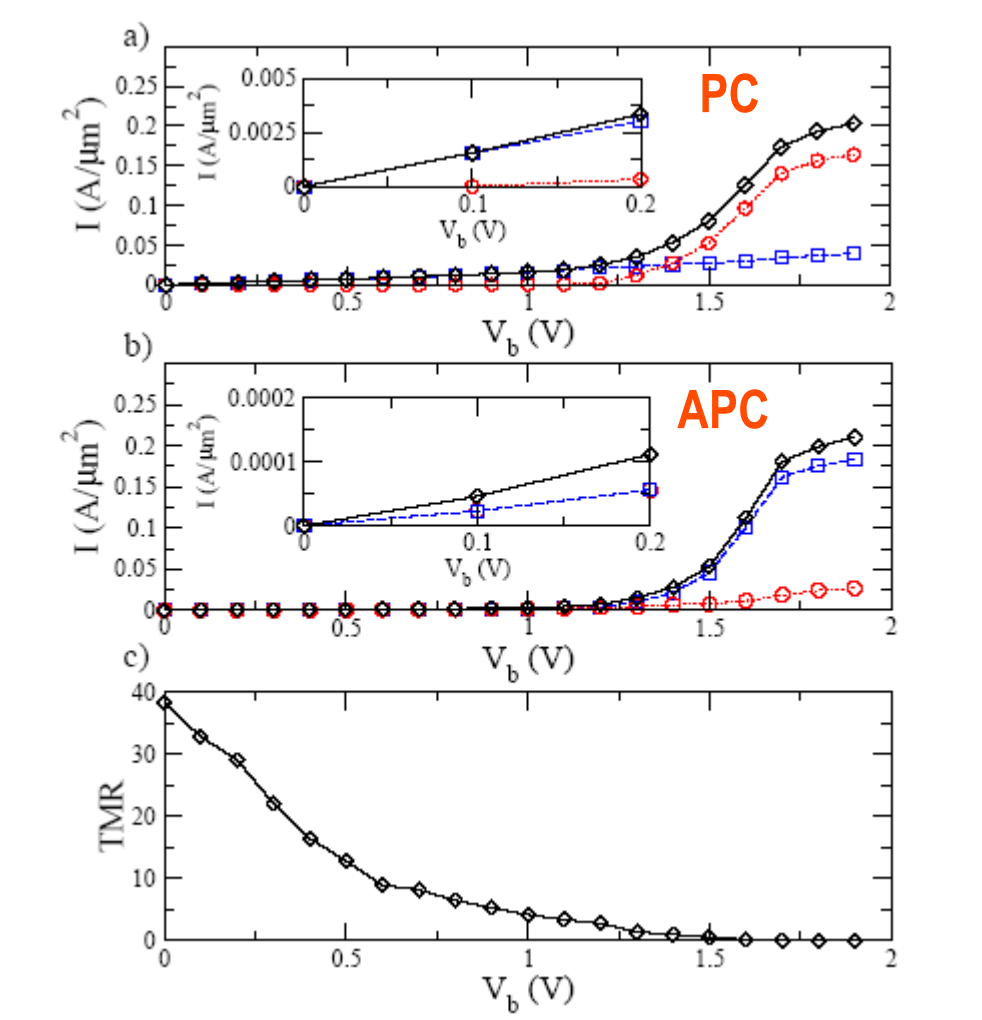
- **Zero bias TMR ratio:**
- **Theory:** many thousands percent.
- **Experiment:** several hundred percent.

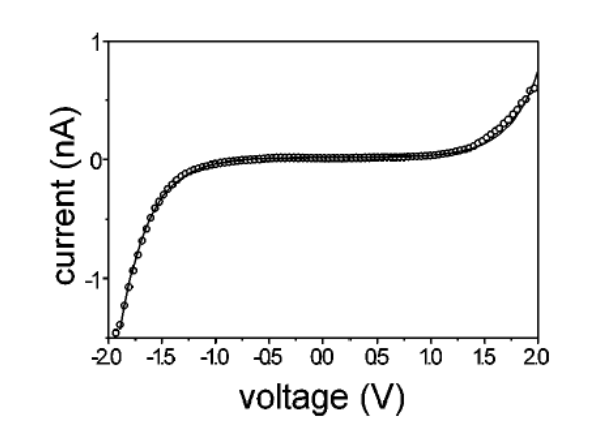
- **TMR versus bias voltage:**
- **Theory:** either increase with bias, or no dependence.
- **Experiment:** reduce with bias.

Can we understand these things? Warm up problem.

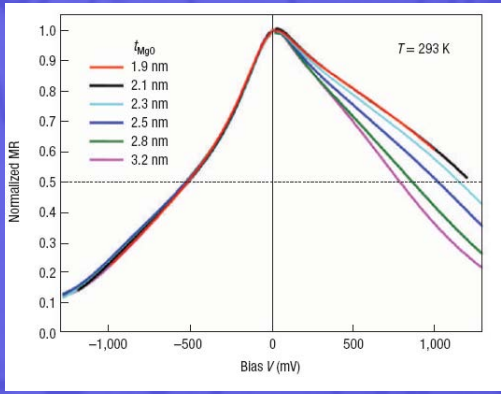
5 layer MgO:

$$TMR = \frac{I_{tot}^{\uparrow\uparrow} - I_{tot}^{\uparrow\downarrow}}{I_{tot}^{\uparrow\downarrow}}$$



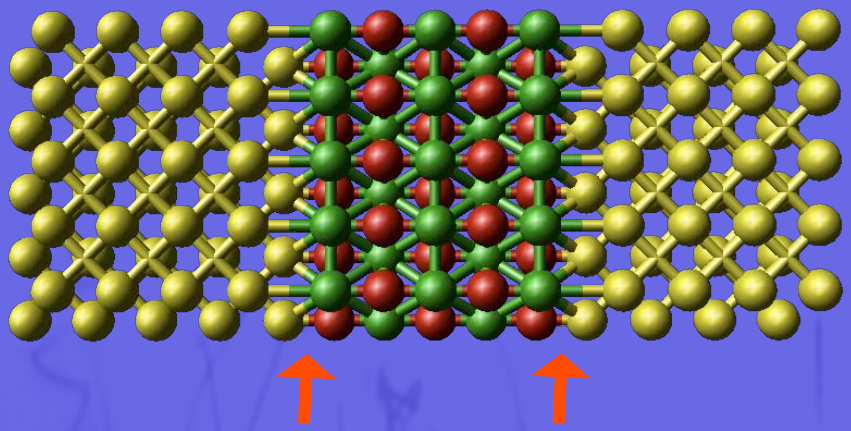


5-layer MgO, measured by Wulfhekel et al APL 78, p509 (2001). STM-MgO-Fe

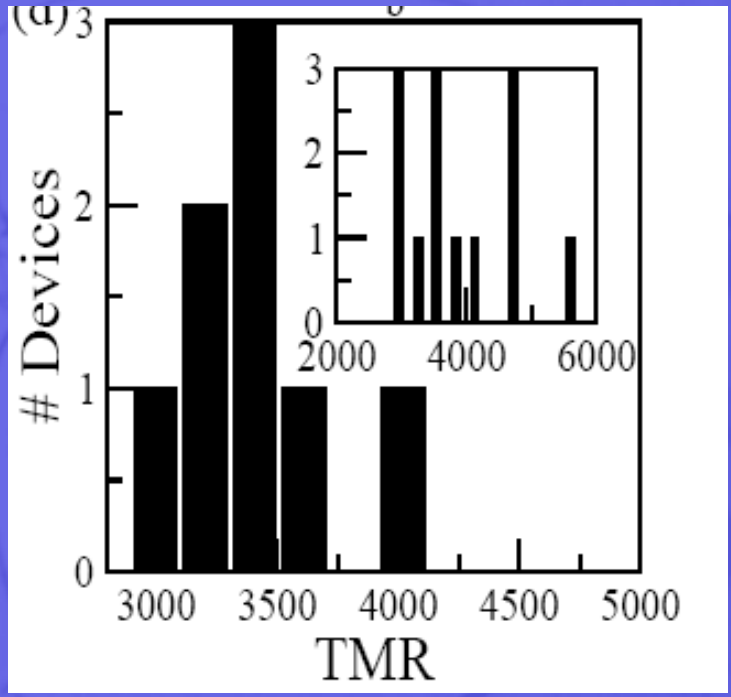


Yuasa et al., Nature Mat. Vol.3, 868 (2004).

Zero bias TMR: effects of small structure changes



Randomly change
interface bonds by ~1%.



Also, randomly change all bonds by ~1%.

These small structure changes do vary the value of TMR, but not enough to reduce it to the current experimental level (250%).

The remaining puzzle: zero bias TMR too large

Experimental data : ~200%;

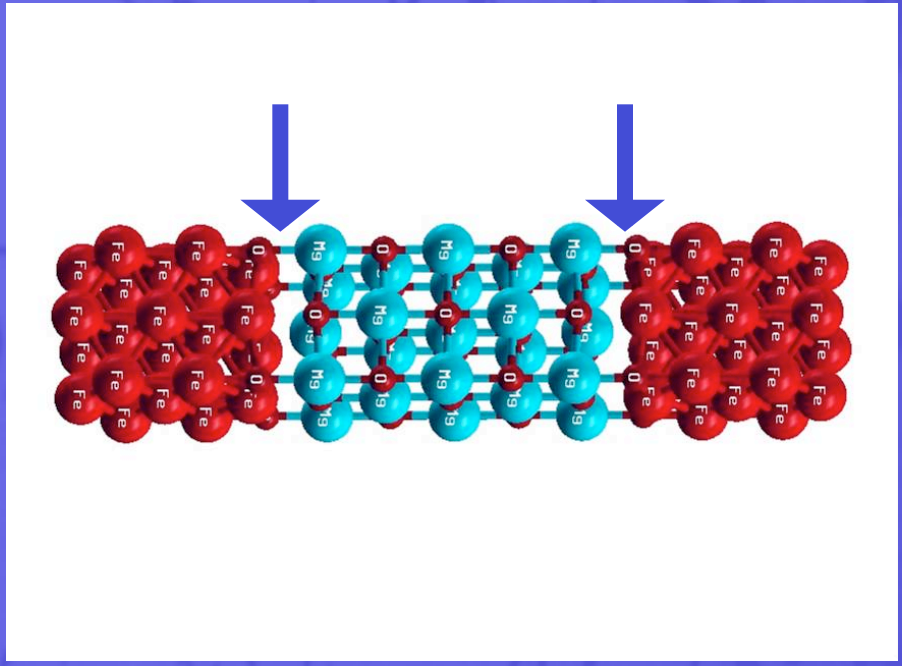
Theoretical result: ~3700%.

Possible reason: oxidizationlayer exists at the Fe/MgO interfaces.

Our results:

100% oxidization: TMR dropped to 169%. 50% oxidization: TMR dropped to 1040%.

Experimental structure: 60% oxidization (Meyerheim etal, PRL 87, 076102 (2001).

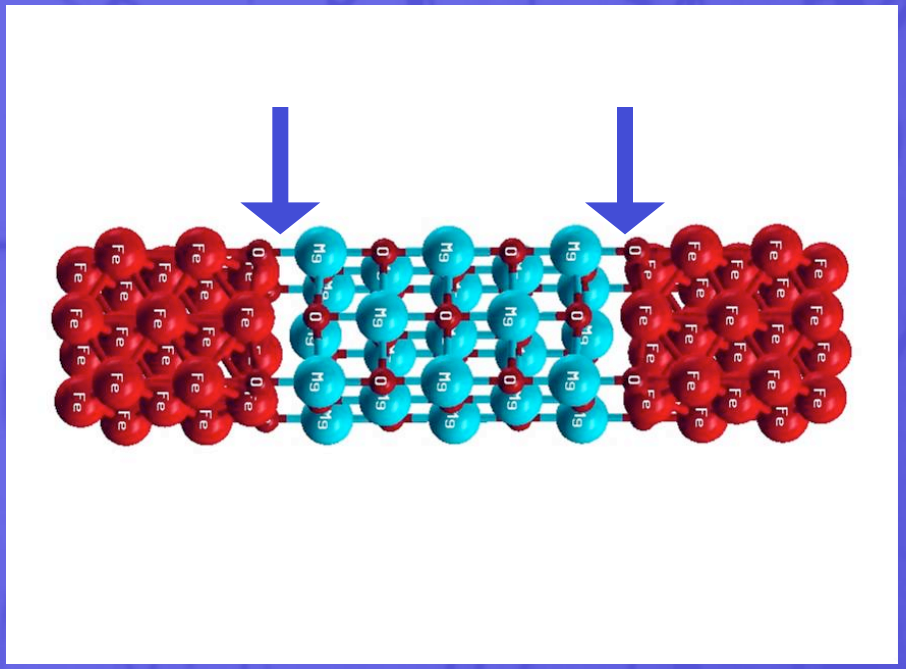


D. Waldron etal, PRL, 97, 226802 (2006).

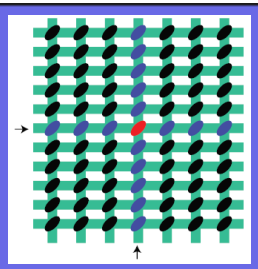
Strong interaction ?

For bulk FeO, LSDA predicts to be metal; but in fact it is an insulator. This well known problem can be addressed by LSDA+U.

Now, for Fe/MgO/Fe device, there are interface FeO bonds. Will these bonds help to localize conduction electrons so as to affect the tunnel junction?



E. Marcotte, V. Timochevski and H.G. (2008).



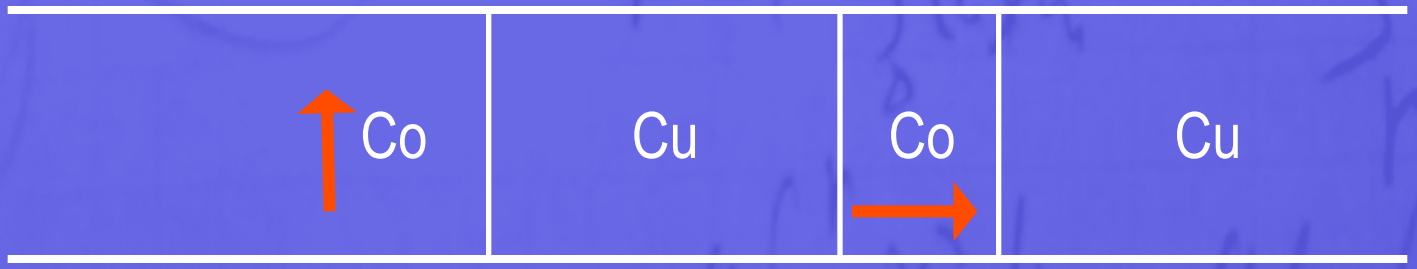
How to change PC to APC ? Spin transfer torque (STT)

STT is the torque exerted on magnetization by spin polarized current. It has important implications to MRAM technology.

$$\frac{d\mathbf{s}_i}{dt} = \frac{1}{i\hbar}\sum_{i<j} \mathrm{Tr}\left[H^T_{i,j}\boldsymbol{\tau}G^<_{i,j} - \mathrm{h.c.}\right] + 2(\boldsymbol{\Delta}_i \times \mathbf{s}_i)$$

Spin current

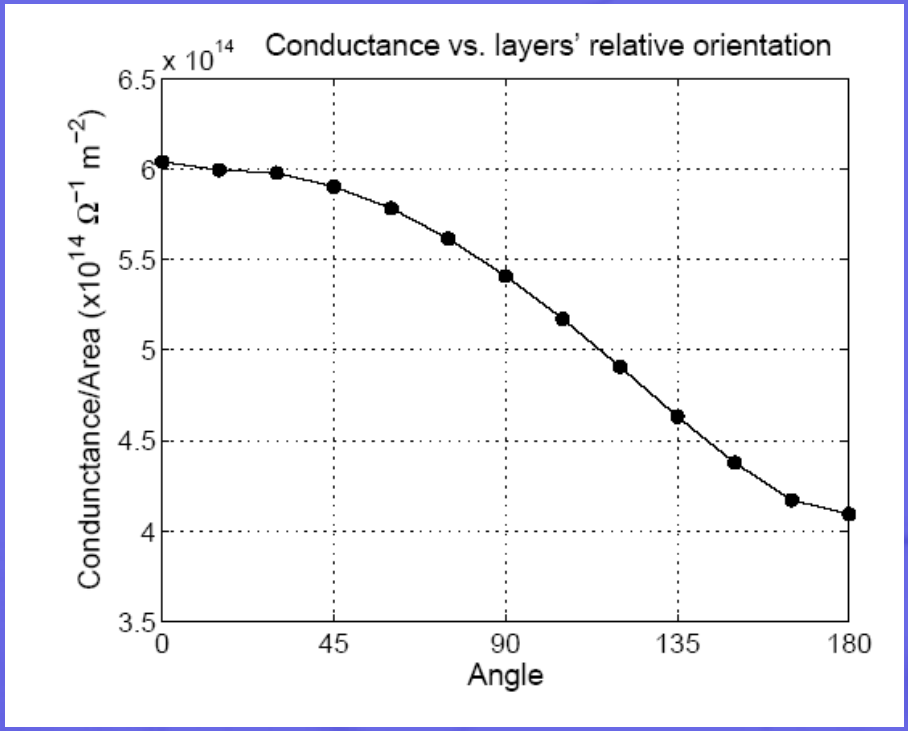
precession

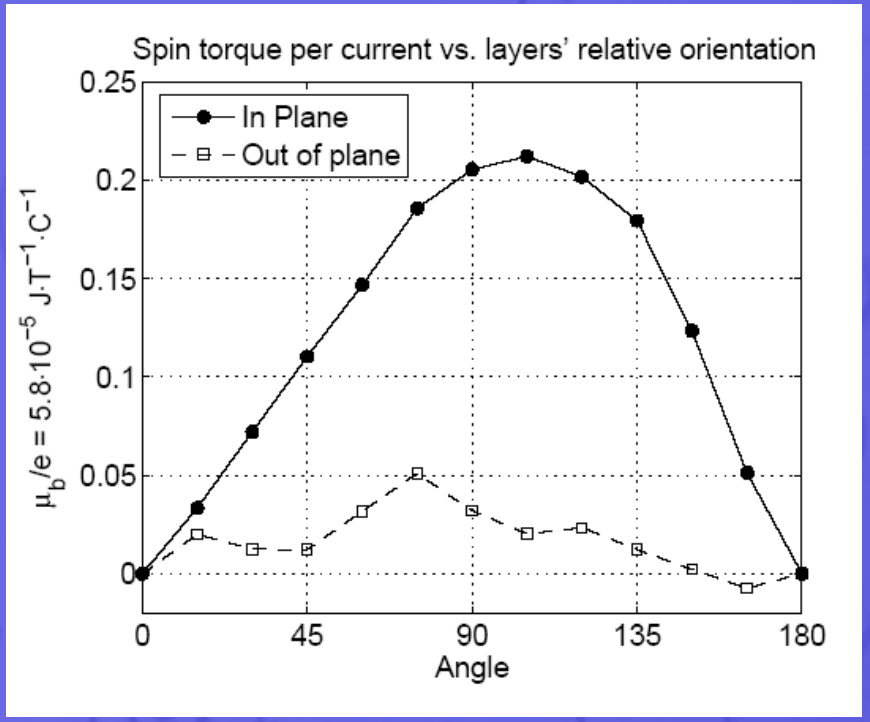


P. Haney etal., PRB 76, 024404 (2007).

Spin Transfer Torque

Haney etal PRB 76, 024404 (2007)





The angular pre-factor of STT has been measured experimentally in PRL 84, 3149(2000). The calculated value is within a factor of 3 if assuming bulk Co damping factor, and agrees almost perfectly if assuming a reasonable value of thin film damping factor.

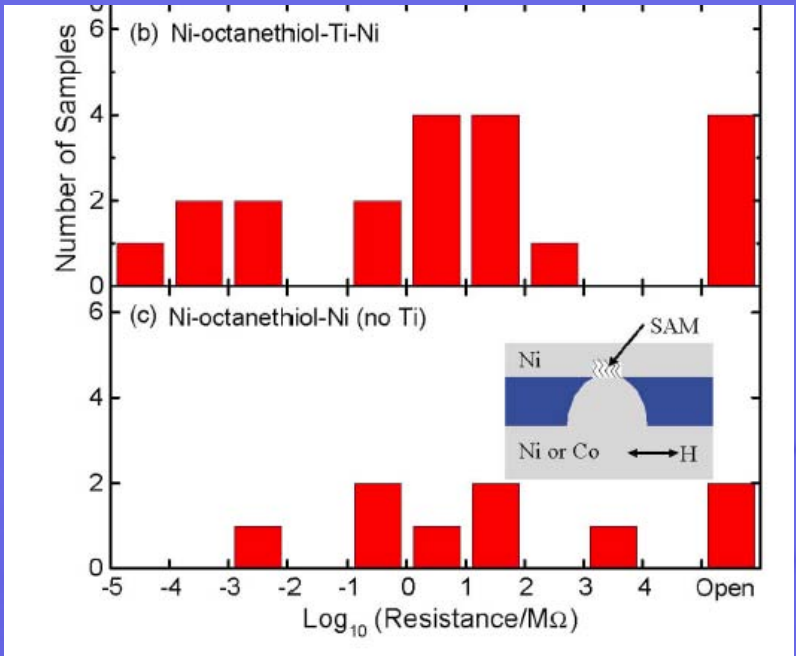
Example 2.

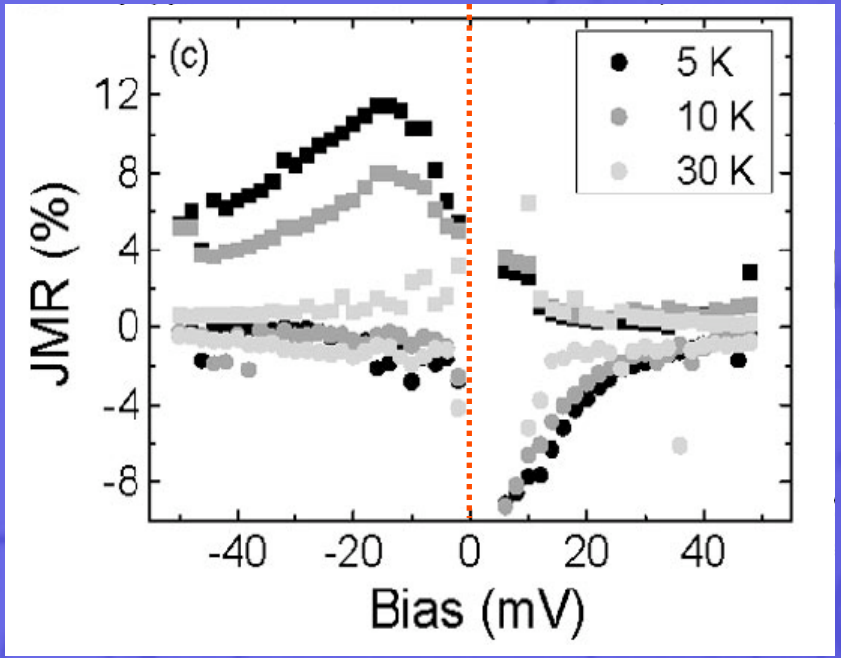
Spintronics: Molecular tunnel junction

Zhanyu Ning, Yu Zhu, Jian Wang and Hong Guo, PRL **100**, 056803 (2008).

Waldron etal PRL 96, 166804 (2006).

Ni-Octanethiol-Ni TMR: Experiments by Ralph etal, PRL93,136601(2004).

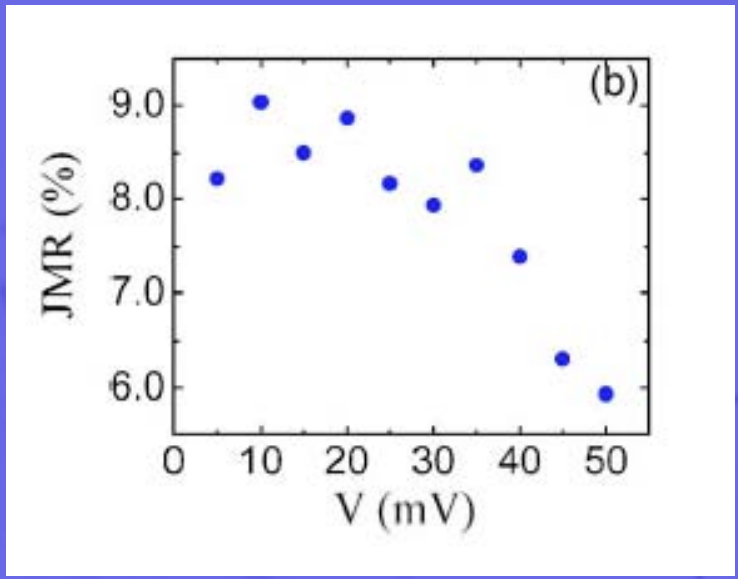




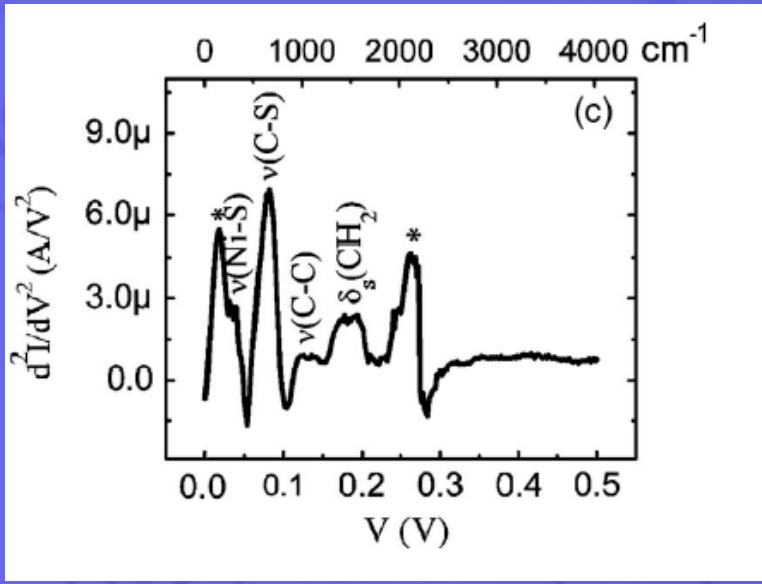
In all devices, highest JMR: 16% at 4.2K.

This device: TMR is asymmetric vs bias;
Peak TMR 12%;
Peak at -15mV negative bias;
Peak TMR decays to zero with a voltage scale of ~60mV.

Another experiment:



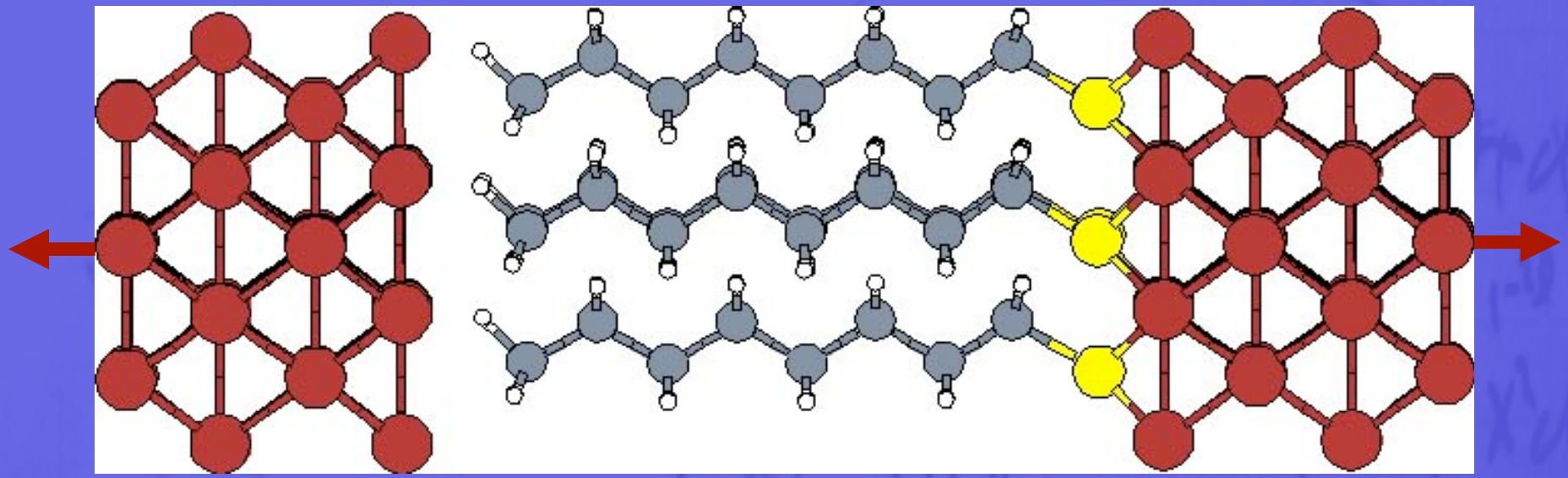
TMR vs bias



IETS spectra

W. Wang and C.A. Ricter. Appl. Phys. Lett. 89, 153105(2006).

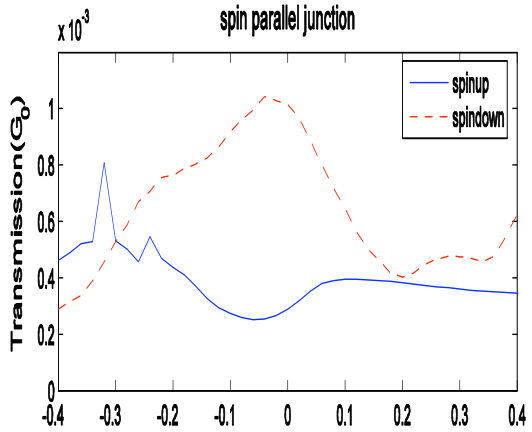
Our model of Ni(100)-Octanethiol-Ni(100) MTJ:

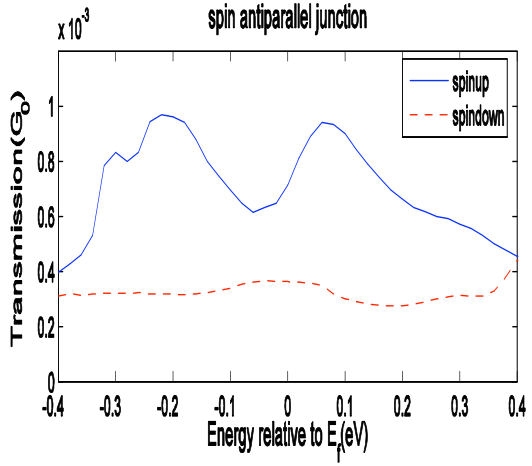


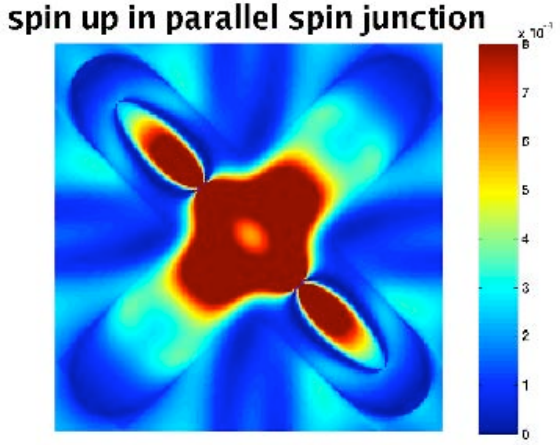
In this model, the molecules periodically align along the nickel surface to simulate an octanethiol monolayer. The geometric structure is fully relaxed before transport analysis is done.

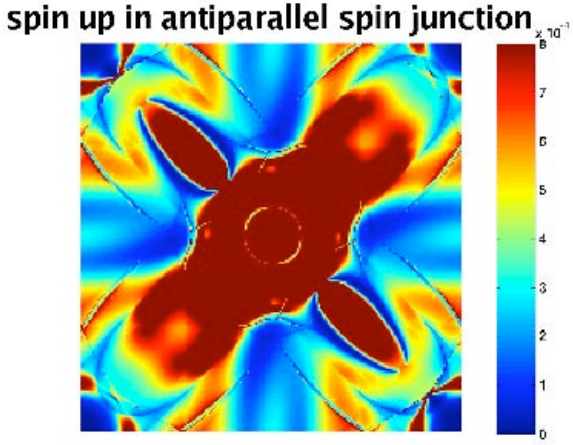
Zhanyu Ning, Yu Zhu, Jian Wang and H.G, PRL 100, 056803 (2008).

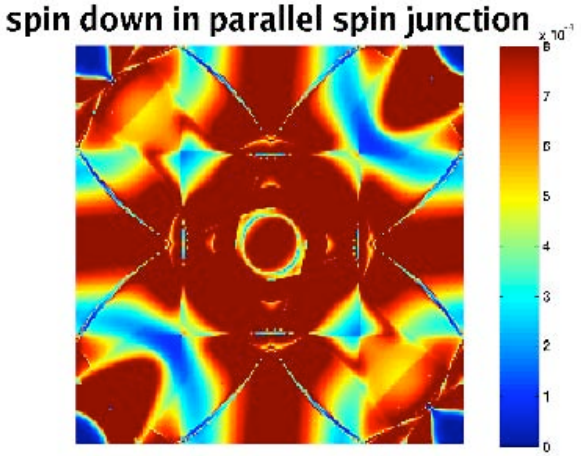
Transmission hot spots in 2D Brillouin Zone (zero bias)

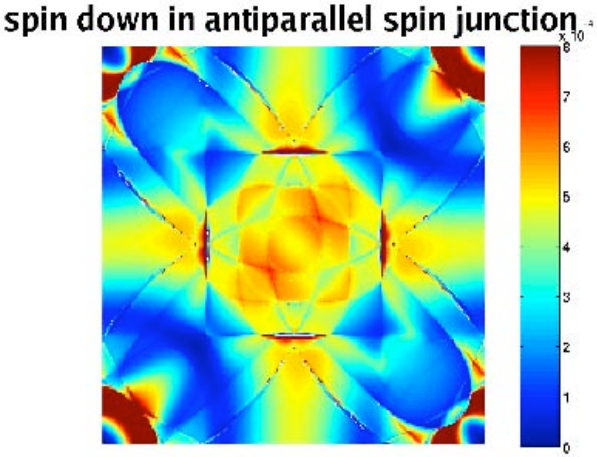






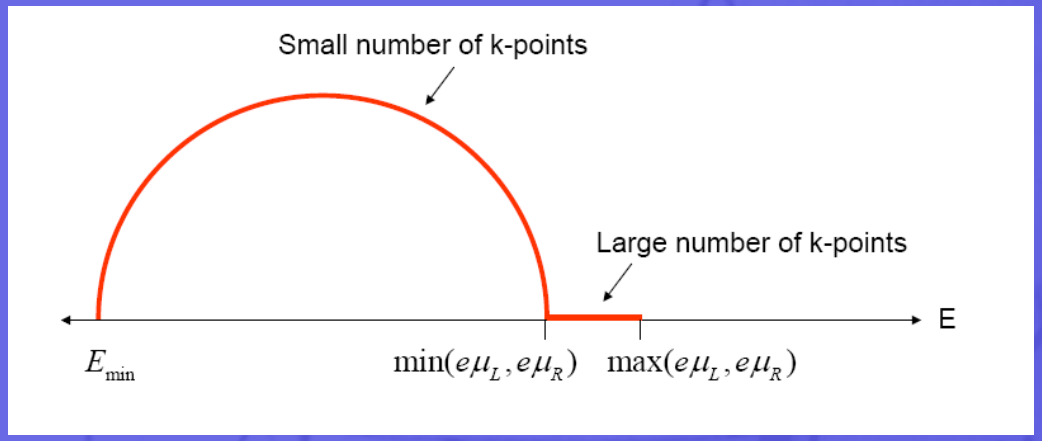




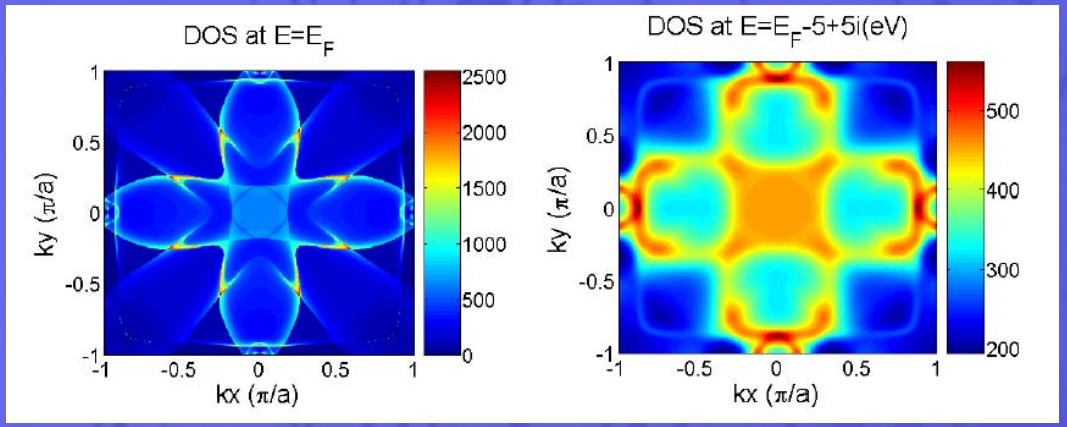


A crucial technical detail for TMR calculations: # of k-points

$$\rho = -\frac{i}{2\pi}\int_{-\infty}^{\infty}\int_{BZ} G^{<}_{\mathbf{k}_{||}}(E)d\mathbf{k}_{||}dE,$$



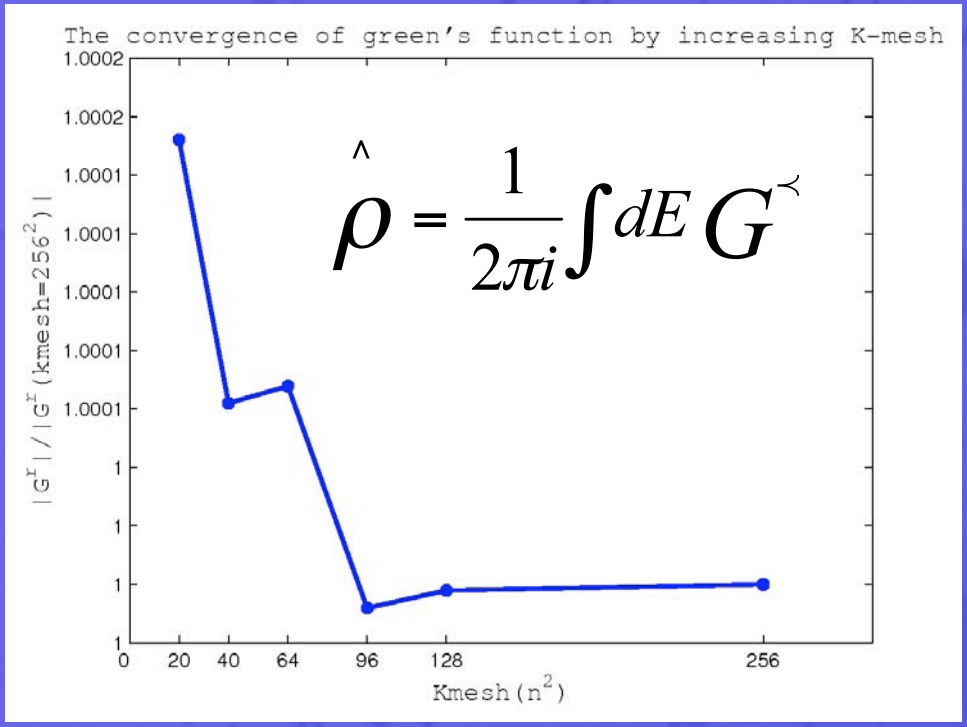
1. In computing density, for some problems a huge number of k-points are needed.
2. In computing transmission coefficient, huge number of k-points are always needed.



Precision control: a very difficult problem

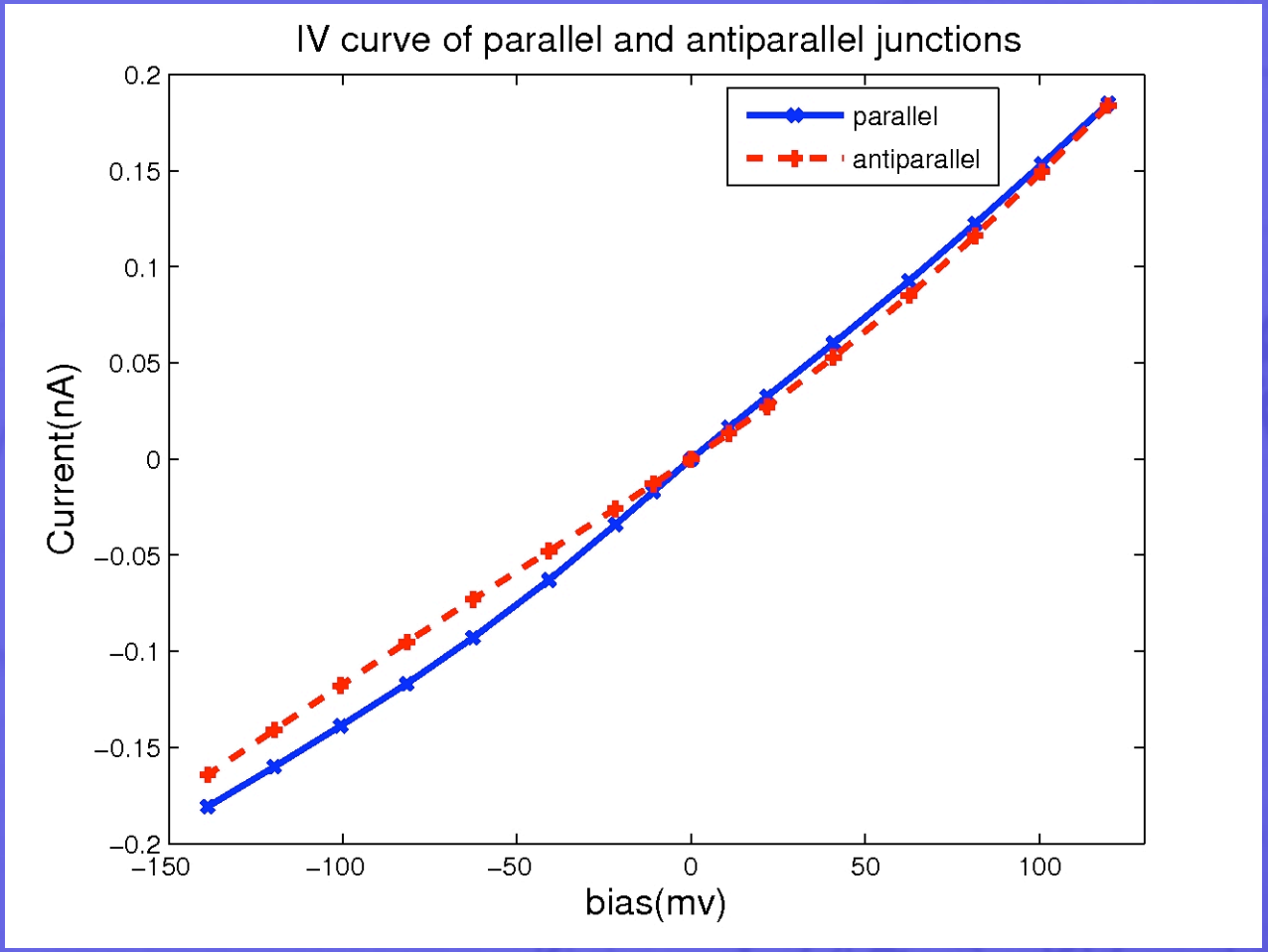
For Ni-Octanethiol-Ni, turns out that a very large k-mesh is needed to converge the density matrix. After thorough test, we used a 96 x 96 k-mesh to obtain converged results.

$$G^R_{\mathbf{k}_{||}} = \begin{pmatrix} \tilde{H}_L^{\mathbf{k}_{||}} + \Sigma_L^{\mathbf{k}_{||}} & \tilde{V}_L^{\mathbf{k}_{||}} & 0 \\ \tilde{V}_L^{\mathbf{k}_{||},\dagger} & \tilde{H}_C^{\mathbf{k}_{||}} & \tilde{V}_R^{\mathbf{k}_{||}} \\ 0 & \tilde{V}_R^{\mathbf{k}_{||},\dagger} & \tilde{H}_R^{\mathbf{k}_{||}} + \Sigma_R^{\mathbf{k}_{||}} \end{pmatrix}^{-1},$$



$$\hat{\rho} = \frac{1}{2\pi i}\int dE\, G^{<}$$

I-V curves of Ni-alkanethiol-Ni

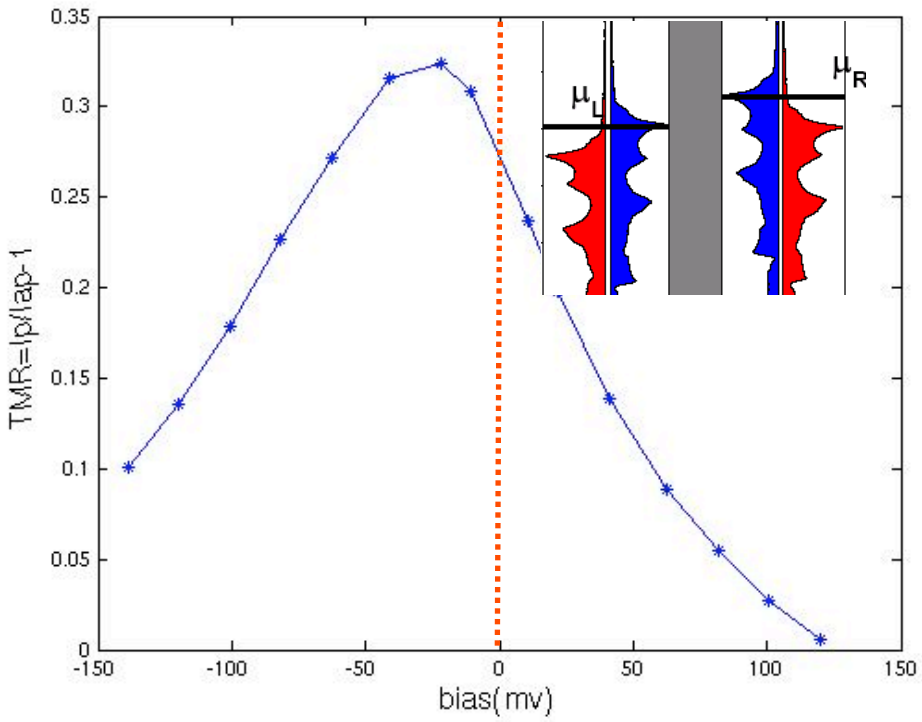


Calculated results:

Peak TMR=33% at -20mv compared with experimental data of 16% at -15mv.

TMR decays to zero around 120mV compared with experimental data of 40mV.

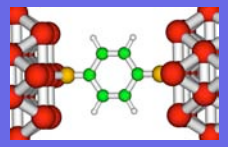
TMR vs bias is asymmetric, also observed in the experiment data.



The level of comparison is very good at this stage of the research.

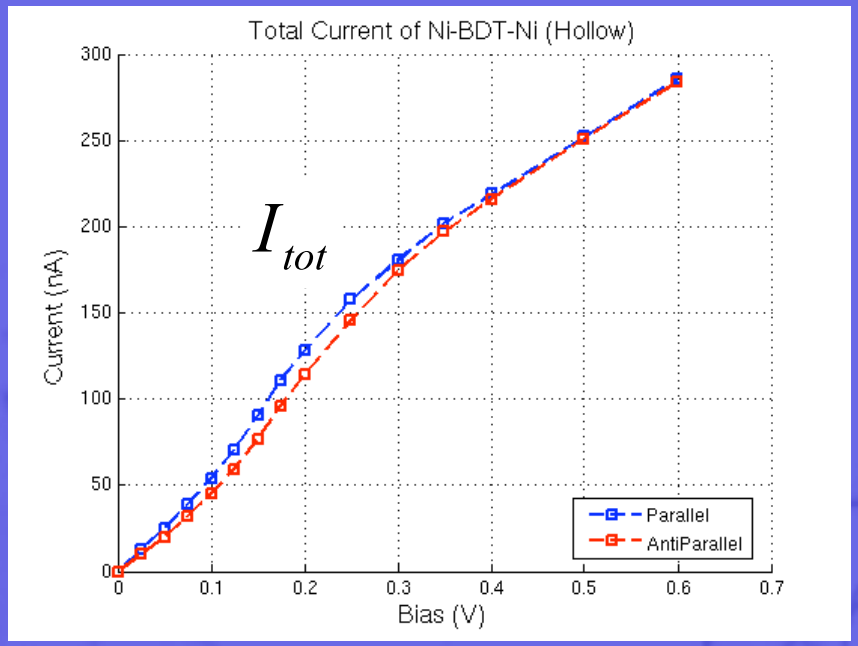
At finite bias voltages, the DOS of Ni leads are shifted and mismatched, leading to the rapid decay of TMR.

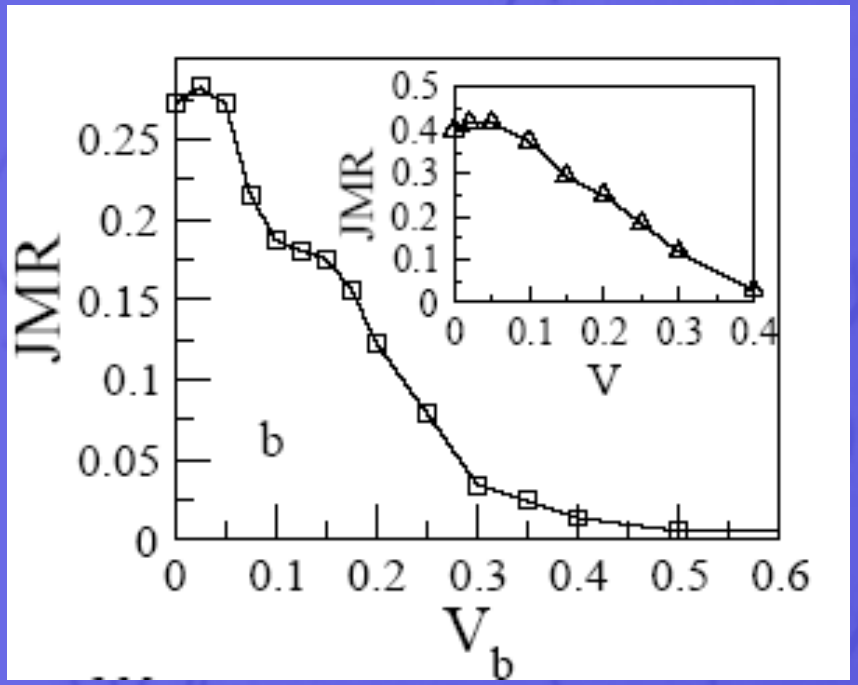
Ning etal, PRL **100**, 056803 (2008).



Ni-BDT-Ni: Magneto-resistance ratio

$$\frac{I_{tot}^{\uparrow\uparrow} - I_{tot}^{\uparrow\downarrow}}{I_{tot}^{\uparrow\downarrow}}$$





There are some molecular signature. For Ni-BDT-NI, the decay voltage scale is about 5 times greater.

Waldron etal PRL 96, 166804 (2006)

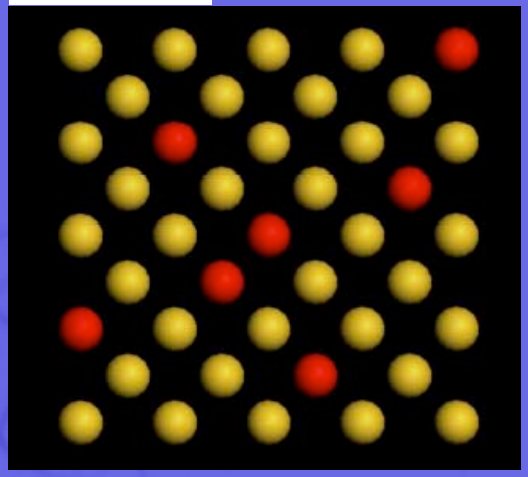
Example 3.

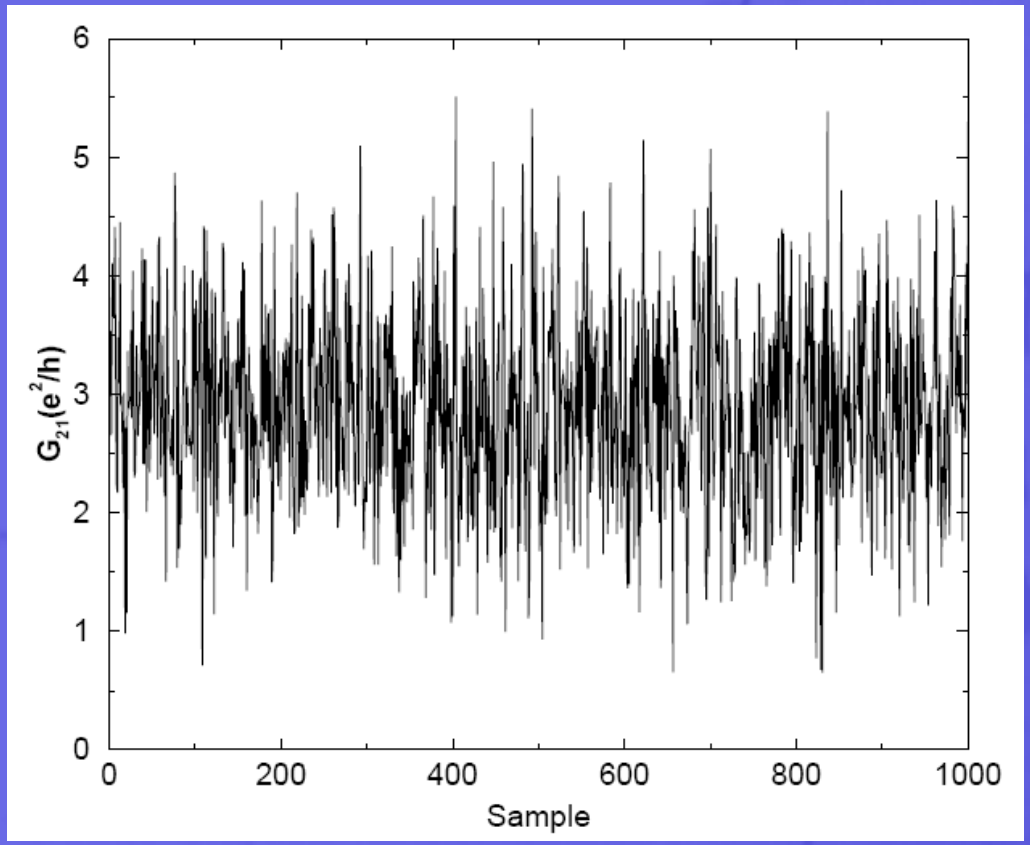
Disorder average: non-equilibrium vertex correction

Youqi Ke, Ke Xia and H.G, Phys. Rev. Lett.**100**, 166805 (2008).

Conductance of wires with impurities

$A_x B_{1-x}$





Huge sample to sample variations. Results must be averaged to be meaningful.
Calculation in this picture was by T. Dejesus, Ph.D thesis, McGill University (2000).

Disorder is a big headache problem in atomic calculations:

Conventional calculation: brute force averaging. One generates many samples and compute for each sample, finally averages over the ensemble.

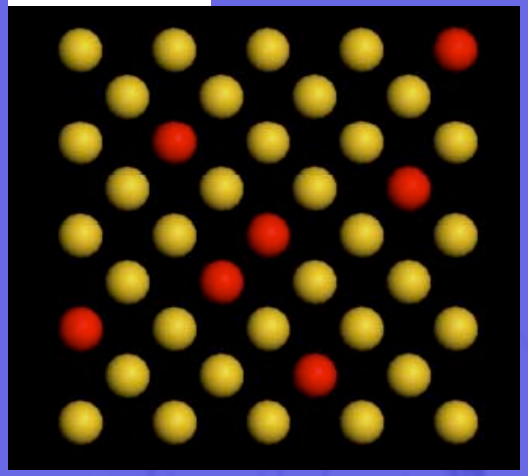
For ab initio calculations, brute force averaging suffers:

- If impurity concentration is low, ~0.1%, too difficult to do *ab initio* calculation.
- Huge number of configurations must be averaged, takes forever to do.

A better method is desired.

Average over H: CPA --- well established method

$A_x B_{1-x}$



When there are impurities, translational symmetry is broken. Coherent Potential Approximation (CPA) is an effective medium theory that averages over the disorder and restores the translational symmetry. So, an atomic site has x% chance to be occupied by A, and (1-x)% chance by B.

PHYSICAL REVIEW VOLUME 184, NUMBER 3 15 AUGUST 1969

Theory of Electronic Transport in Disordered Binary Alloys: Coherent-Potential Approximation*

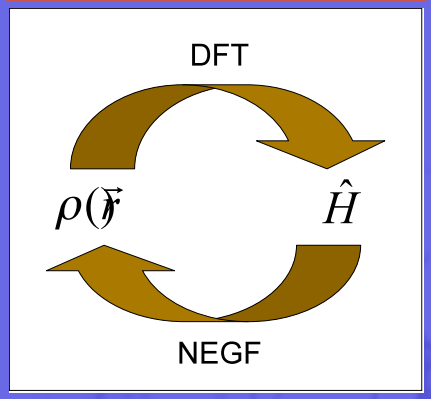
B. VELICKÝ†
Division of Engineering and Applied Physics, Harvard University, Cambridge, Massachusetts 02138

(Received 19 March 1969)

$$T(E,\Delta V) = Tr[G^r \Gamma_L G^a \Gamma_R]$$

Non-equilibrium vertex correction in NEGF-DFT

State-of-the-art 1st principles formalism:

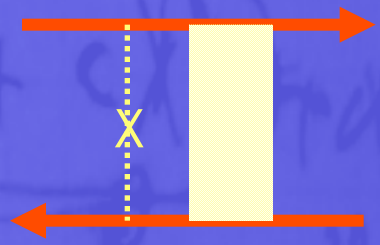


$$\rho \sim \int G^{<}\, dE = \int G^{R}\Sigma^{<}G^{A}\, dE$$

Average over random disorder:

$$\overline{\rho} \sim \int G^{<}\, dE = \int \overline{G^{R}\Sigma^{<}G^{A}}\, dE$$

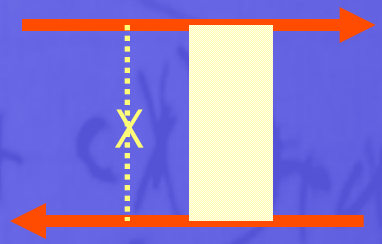
$$\overline{T} = Tr\left(\overline{G^{R}\Gamma_{l}G^{A}\Gamma_{r}}\right)$$



Progress report: we have so far worked out the correlated disorder scattering at the nonequilibrium level, implemented NEGF-DFT-Vertex software, and applied it to several device modeling problems: spin filter by DMS, roughness scattering in magnetic tunnel junctions, and roughness effect for Cu interconnect.

Essence of Nonequilibrium Vertex Correction (NVC)

$$\overline{g}^{\alpha,<} = \overline{g^{\alpha,\mathcal{R}}\Sigma^{\alpha,<}g^{\alpha,\mathcal{A}}}$$
$$= \overline{g}^{\alpha,\mathcal{R}}(\Sigma^{\alpha,<} + \Omega_{NVC})\overline{g}^{\alpha,\mathcal{A}}$$



Conventional vertex correction, for example those appear in computing Kubo formula in metal, is done at equilibrium.

NVC is done at nonequilibrium so that it is related not only to multiple impurity scattering, but also the nonequilibrium statistics of the device scattering region.

Rather messy and complicated in technical details

$$\Omega_{NVC,R} = \sum_{Q=A,B} C_R^Q t_R^{Q,\mathcal{R}} [\overline{g}^{\alpha,\mathcal{R}} \Sigma^{\alpha,<} \overline{g}^{\alpha,\mathcal{A}}]_{RR} t_R^{Q,\mathcal{A}}$$

$$- \sum_{Q=A,B} C_R^Q t_R^{Q,\mathcal{R}} \overline{g}_{RR}^{\alpha,\mathcal{R}} \Omega_{NVC,R} \overline{g}_{RR}^{\alpha,\mathcal{A}} t_R^{Q,\mathcal{A}}$$

$$+ \sum_{Q=A,B} C_R^Q t_R^{Q,\mathcal{R}} [\sum_{R'} \frac{1}{N_{k_\parallel}} \sum_{k_\parallel} \overline{g}_{RR'}^{\alpha,\mathcal{R}}(k_\parallel, E) \Omega_{NVC,R'}$$

$$\times \overline{g}_{R'R}^{\alpha,\mathcal{A}}(k_\parallel, E)] t_R^{Q,\mathcal{A}} \qquad (23)$$

See EPAPS Document No. E-PRLTAO-100-020817 for supplemental material. For more information on EPAPS, see http://www.aip.org/pubservs/epaps.html.

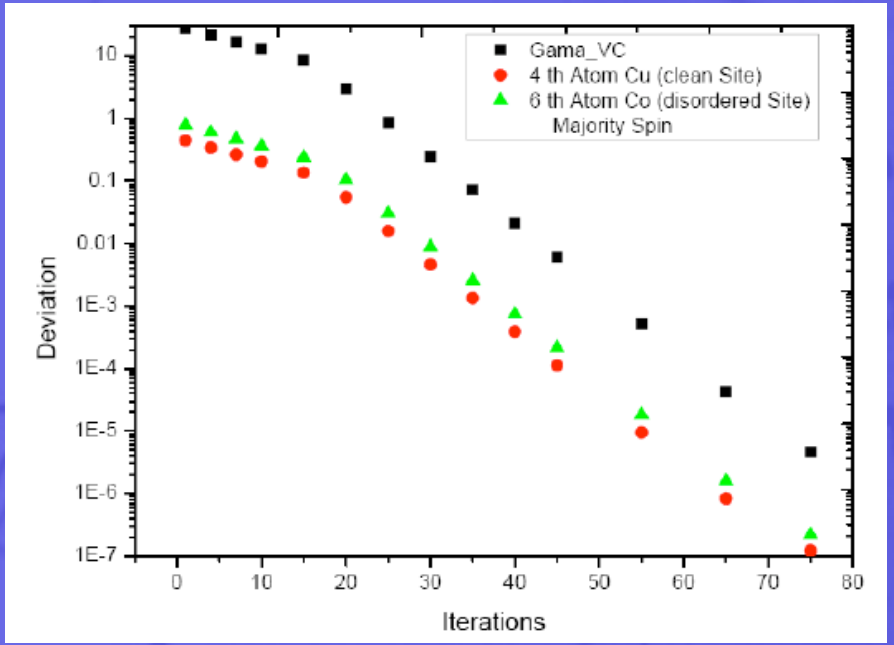
A tough check:

$$\overline{G}^{<,Q}_{RR} = \overline{G}^{A,Q}_{RR} - \overline{G}^{R,Q}_{RR}$$

At equilibrium, fluctuation-dissipation theorem holds.

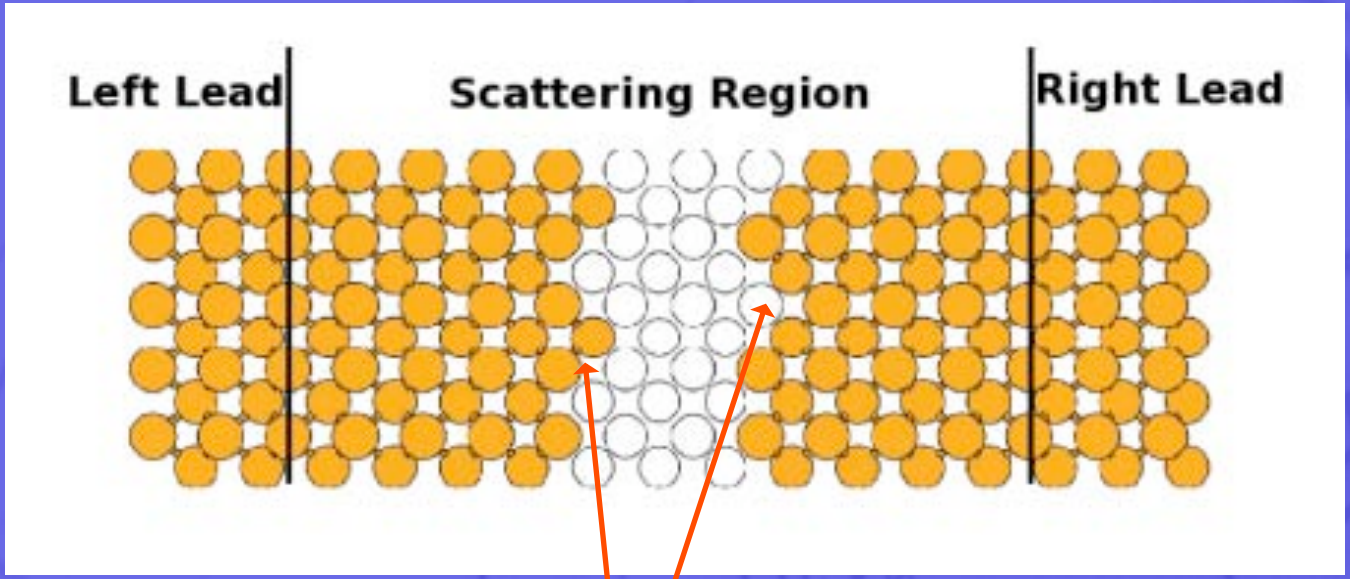
Left hand side has NVC; right hand side does not.

This gives a very tough check to the NVC formalism as well as to the numerical implementation.



5 ML Cu + 5 ML Co0.8Cr0.2 + 20ML Cu + 2ML Co + 5ML Cu

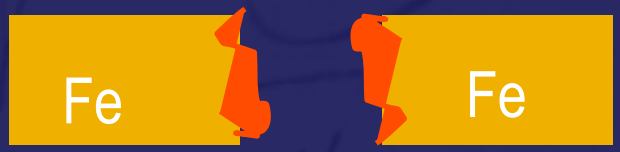
Roughness scattering for MTJ:

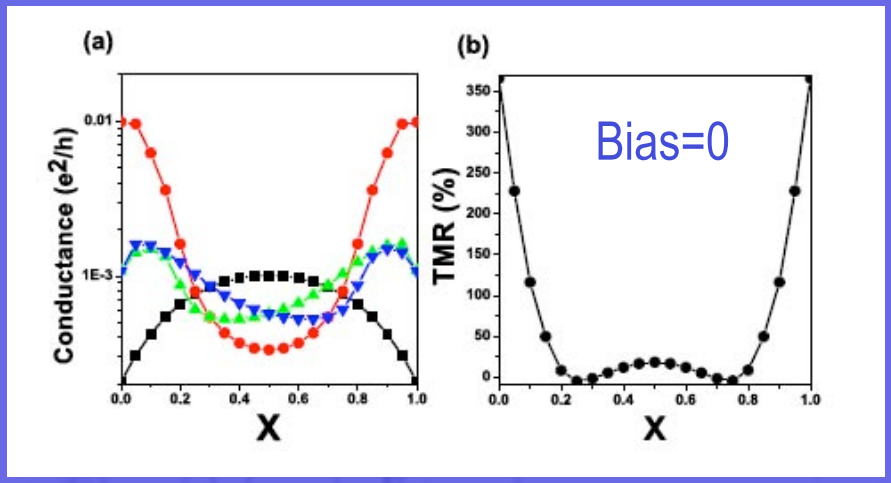


Rough interfaces:

Left (1-x)%-disorder; right x% disorder

Roughness scattering for MTJ:





Red—spin down channel in PC;

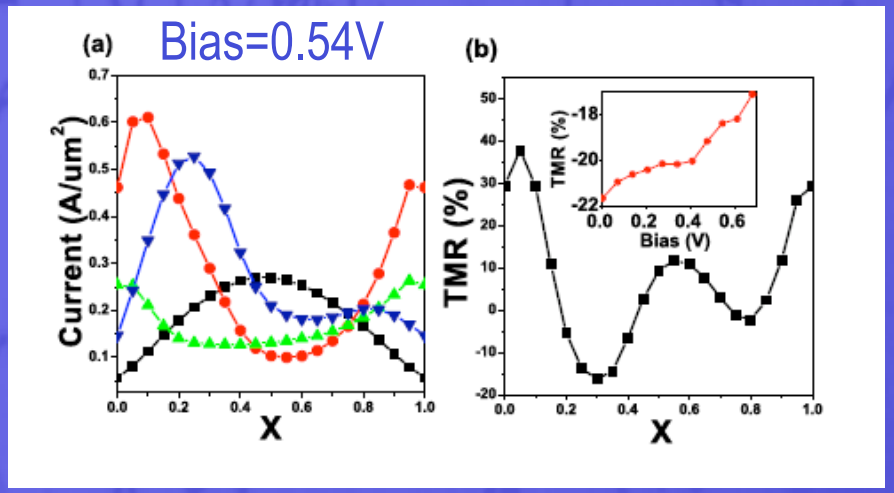
Black-spin up in PC;

Green—spin up in APC;

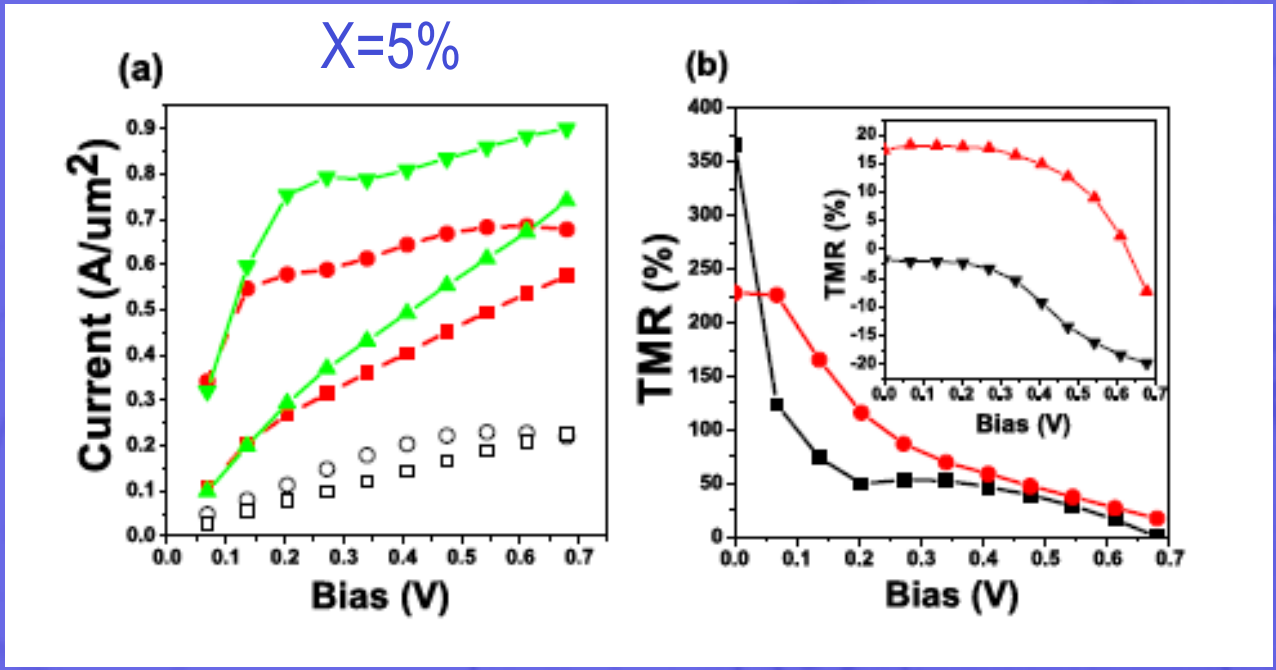
Blue --- spin down in APC

$G_{\uparrow}(x) = G_{\downarrow}(1-x)$ for APC.

Physical quantities vs disorder concentration x for zero or finite bias voltage.



Roughness scattering for MTJ:

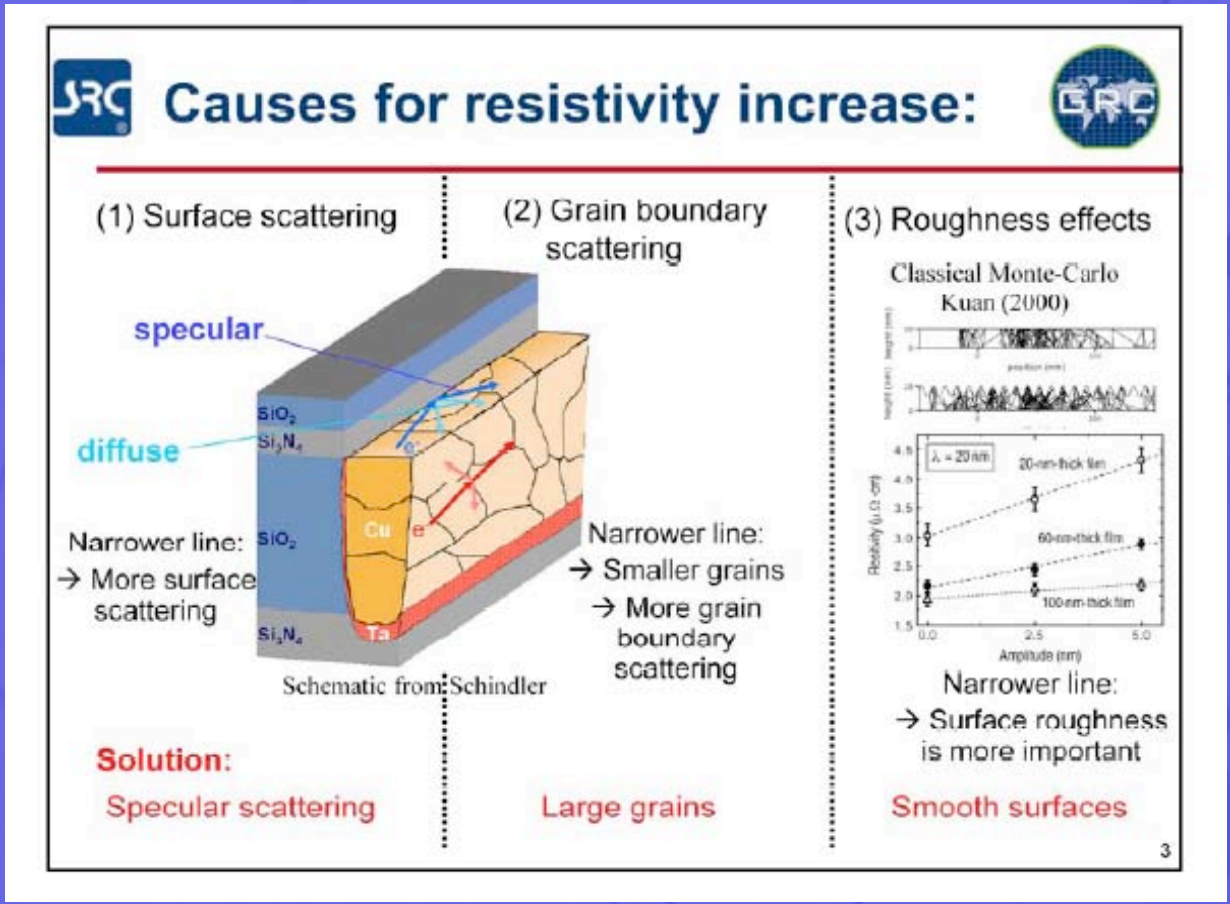


Green dots: PC and APC currents without NVC;

Red dots: full NVC results.

Empty dots: with NVC but without VC in transmission calculation.

Application of NVC: Resistance of Cu interconnects



Collaboration with Prof. Daniel Gall of RPI, funded by SRC.

Example 4.

Transient current driven by a voltage pulse

Progress report

E. Zhu etal PRB 71, 075317 (2004).

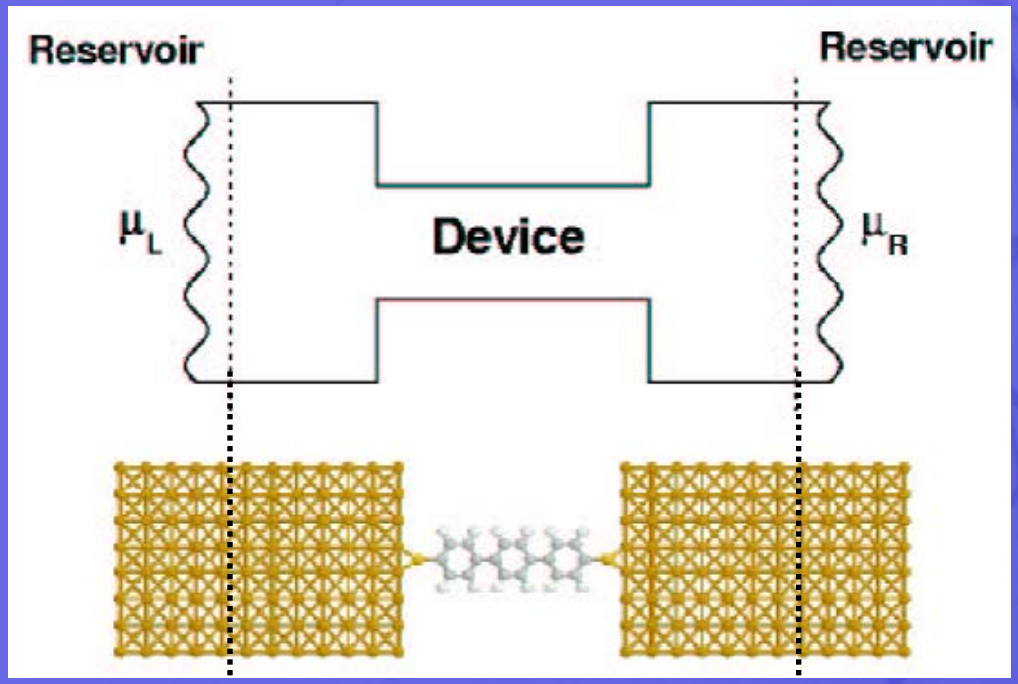
Maciejko etal PRB 74, 085324 (2006).

Ning etal, in progress.

Compute *transient current* from first principles including material details

Time dependent current driven by a voltage pulse applied to a lead:

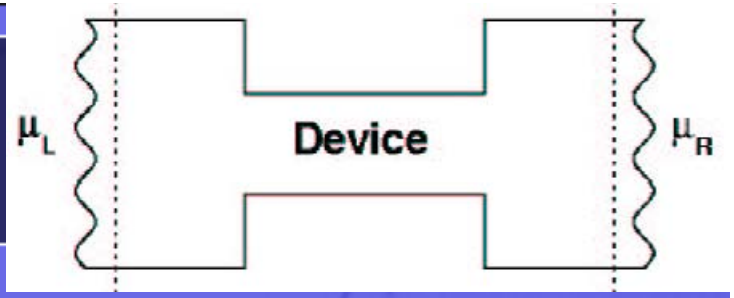
Voltage pulse



Estimating speed: 1. Compute R, L, C, somehow.

2. Send a voltage pulse and directly follow J(t).

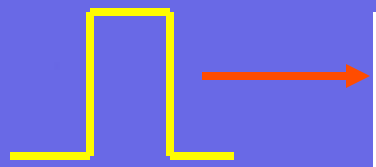
A very hard problem:



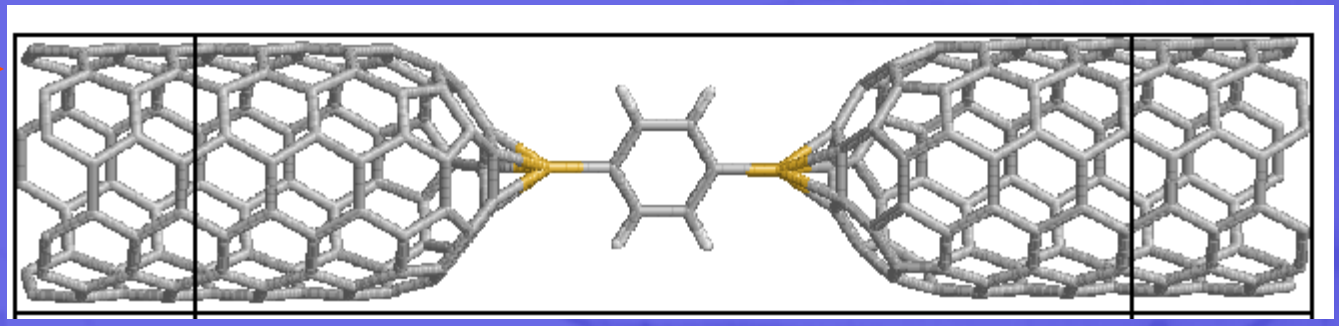
Theoretical model: $H = H_{leads} + H_{device} + H_{coupling} + V(t)$

1. Transfer Hamiltonian: Bardeen formula. Johansson, PRB 41, 9892 (1990).
2. Nonequilibrium Green's functions (NEGF): partitioned approach. C. Caroli etal, J. Phys. C 4, 916 (1971).
3. NEGF: partition free approach. M. Cini, PRB 22, 5887 (1980); G. Stefanucci and Almbladh, PRB 69, 195318 (2004).
4. Theory of Jauho, Wingreen and Meir: PRB 48, 8487 (1993). Partitioned approach plus wideband limit.
5. TD-DFT: Kurth etal PRB 72, 035308 (2005); Stefanucci etal (2004).
6. Time domain docomposition: E. Zhu etal PRB 71, 075317 (2004).
7. Mixed NEGF approach. Maciejko etal PRB 74, 085324 (2006). Full bandstructure of leads and scattering region.

The model: $H = H_{leads} + H_{device} + H_{coupling} + V(t)$



Voltage pulse



1. Include all atomic details: solve $H_0 = H_{leads} + H_{device} + H_{coupling}$ by NEGF-DFT in steady-state;
2. Solve $H = H_0 + V(t)$ by Dyson's equation;
3. For square shaped pulses, transport described by H can be solved *exactly* including full bandstructures of leads and scattering region.

J. Maciejko, J. Wang and H.G, PRB 74, 085324 (2006).

Back to the full time dependent problem:

$H = H_0 + V(t)$, $\quad H_0 = H_{leads} + H_{device} + H_{coupling}$

Solved by NEGF-DFT

Assumption 1: electronic structure follows V(t) adiabatically.

Assumption 2: V(t) is a perfectly step type pulse.

Voltage pulse

Starting from the steady-state solution of H_0, we add V(t) and solve the problem of H by Dyson equation, we get a close form expression for current J(t):

$$J(t) = \frac{dN(t)}{dt}$$

J. Maciejko, J. Wang and H.G. PRB 74, 085324 (2006).

Down-step pulse:

Exact solution:

$$A_\alpha(\epsilon,t)=\tilde{G}^R(\epsilon)+\int\frac{d\omega}{2\pi i}\frac{e^{-i(\omega-\epsilon)t}\tilde{G}^R(\omega)}{\omega-\epsilon-\Delta_\alpha-i0^+}\left[\frac{\Delta_\alpha}{\omega-\epsilon-i0^+}\right.$$

$$\left.+\left(\Delta-\sum_\beta\Delta_\beta\tilde{\Upsilon}^R_{\alpha\beta}(\omega,\epsilon)\right)\bar{G}^R(\epsilon+\Delta_\alpha)\right],\qquad(22)$$

where we have defined

$$\tilde{\Upsilon}^R_{\alpha\beta}(\omega,\epsilon)\equiv\frac{\tilde{\Sigma}^R_\beta(\omega)-\tilde{\Sigma}^R_\beta(\epsilon+\Delta_\alpha-\Delta_\beta)}{\omega-\epsilon-\Delta_\alpha+\Delta_\beta\pm i0^+},\qquad(23)$$

All the right hand side are known functions or stead-state Green's functions which are obtained by NEGF-DFT. We just have to do some integrals to obtain transient current !

$$J_\alpha(t)=-2e\int\frac{d\epsilon}{2\pi}\,\mathrm{Im\,Tr}\{\Gamma_\alpha(\epsilon)[\Psi_\alpha(\epsilon,t)+f(\epsilon)A_\alpha(\epsilon,t)]\},$$

Up-step pulse:

$$A_\alpha(\epsilon,t) = \bar{G}^R(\epsilon+\Delta_\alpha) - \int \frac{d\omega}{2\pi i} \frac{e^{-i(\omega-\epsilon)t}\bar{G}^R(\omega+\Delta_\alpha)}{\omega-\epsilon+\Delta_\alpha-i0^+} \left[\frac{\Delta_\alpha}{\omega-\epsilon-i0^+} + \left(\Delta - \sum_\beta \Delta_\beta \tilde{\Upsilon}^R_{\alpha\beta}(\epsilon,\omega)\right)\tilde{G}^R(\epsilon) \right],$$

Again, all the right hand side are expressed by the steady-state quantities which are calculable from NEGF-DFT.

Square pulse:

$$A_\alpha(\epsilon,t) = \tilde{G}^R(\epsilon) + e^{i\Delta_\alpha s}\int \frac{d\omega}{2\pi i} e^{-i(\omega-\epsilon)t}\tilde{G}^R(\omega)\Bigg\{ \chi_\alpha^{(-)}(\omega,\epsilon)$$

$$+\Bigg[\left(\frac{e^{i(\omega-\epsilon)s}-1}{\omega-\epsilon-i0^+}\right)\Delta + \sum_\beta [\chi_\beta^{(-)}(\omega,\epsilon)\tilde{\Sigma}_\beta^R(\epsilon)$$

$$- e^{-i\Delta_\beta s}\chi_\beta^{(+)}(\omega,\epsilon)\tilde{\Sigma}_\beta^R(\omega)]\Bigg]\tilde{G}^R(\epsilon)$$

$$+\int \frac{d\omega'}{2\pi i}\Bigg[\sum_\beta \frac{\Delta_\beta \chi_\beta^{(-)}(\omega,\omega')\tilde{\Sigma}_\beta^R(\omega')\tilde{G}^R(\epsilon)}{(\omega'-\epsilon+\Delta_\beta+i0^+)(\omega'-\epsilon+i0^+)}$$

$$-\left(\Delta - \sum_\beta \Delta_\beta \tilde{\Upsilon}_{\alpha\beta}^R(\omega,\omega')\right)Q_\alpha(\omega,\omega',\epsilon)\Bigg]\Bigg\}, \quad (58)$$

Still, all quantities on the right hand side are known.

Down-step pulse:

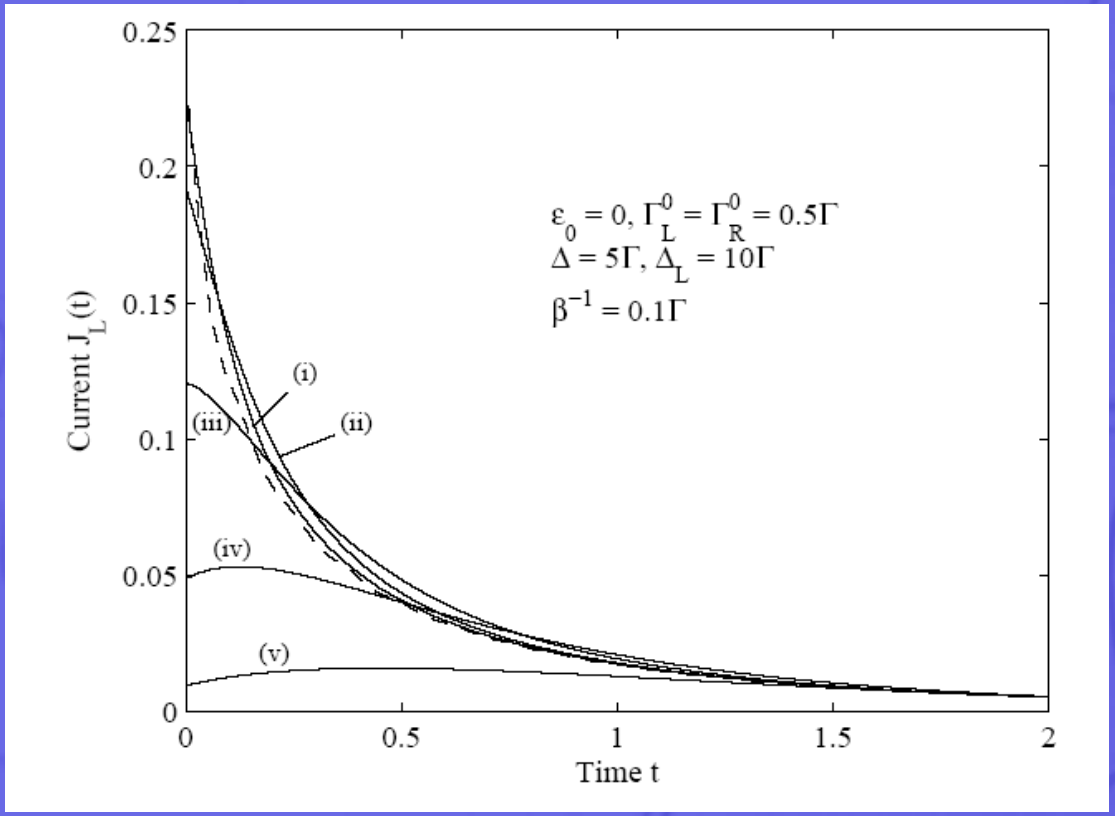
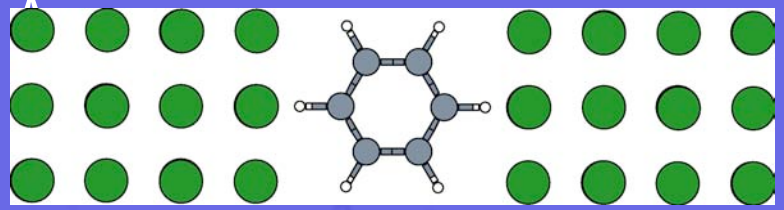
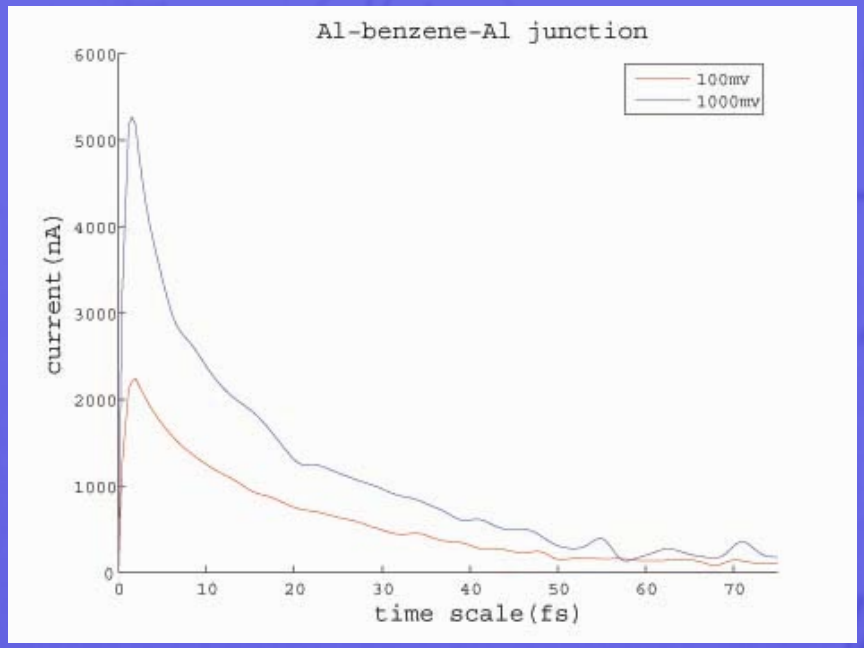


Figure 5.1: Time-dependent current $J_L(t)$ through left lead in response to a downward step pulse for different bandwidths: dashed line, WBL ($W = \infty$); (i) $W = 20\Gamma$, (ii) $W = 10\Gamma$, (iii) $W = 5\Gamma$, (iv) $W = 2.5\Gamma$, and (v) $W = \Gamma$. The current is in units of $e\Gamma/\hbar$ and the time is in units of $\hbar/\Gamma$ where $\Gamma = \Gamma_L^0 + \Gamma_R^0$ is the total linewidth amplitude. Parameters are taken the same as in Ref. [1]. When $W = 100\Gamma$, the result was found (not shown) to be indistinguishable from the $W = \infty$ curve.

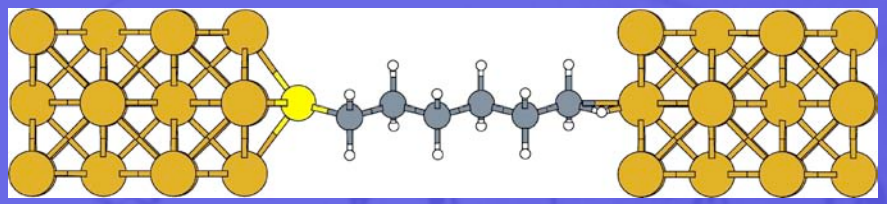
Comparison of different molecular junctions (Downwards pulse)

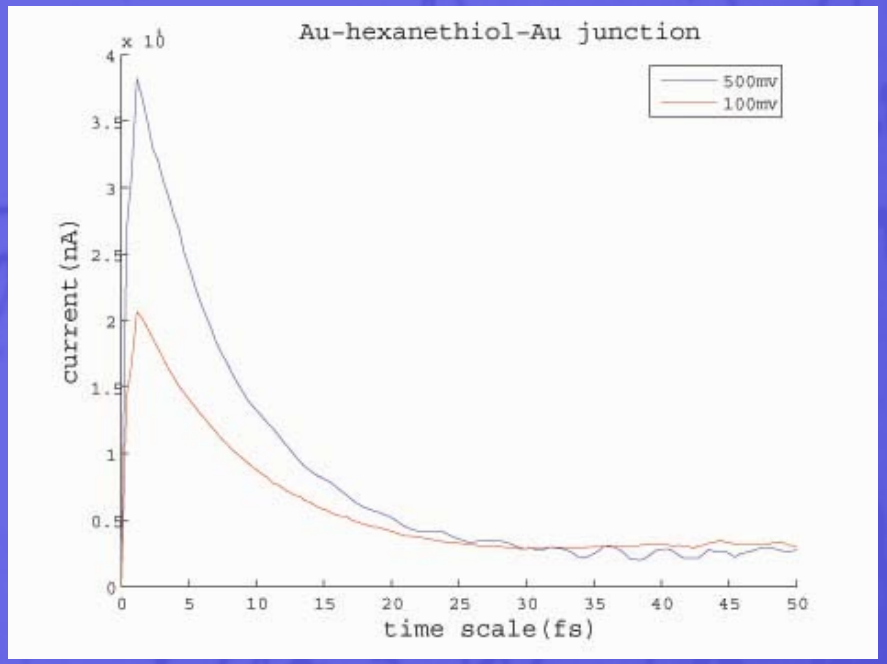
- Al- Benzene- Al





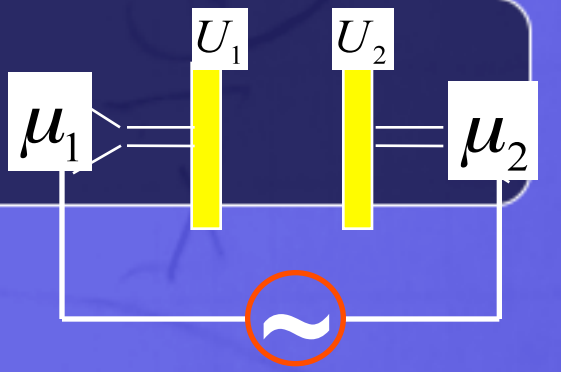
Au-hexanethiol-





Ning etal (2008)

AC transport is full of surprises:



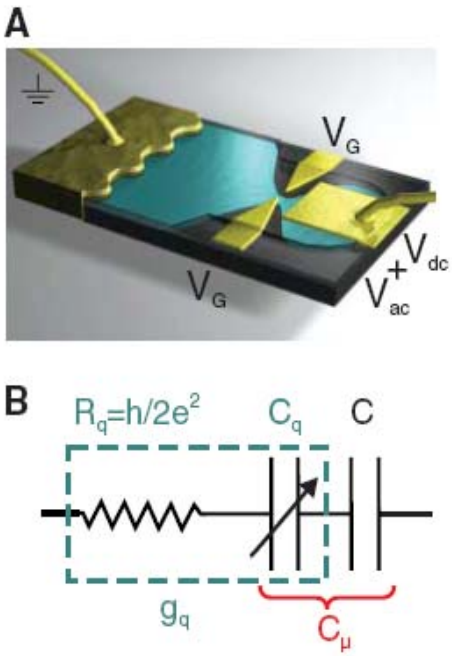
M. Buttiker etal. Phys. Lett. A 180, 364 (1993).
It predicted:

$$G(\omega) = -i\omega C_\mu(\omega)$$

$$G(\omega) = -i\omega C_\mu + \omega^2 C_\mu R_q \,, \qquad R_q = 1/2G_o$$

So, a quantum capacitor behaves as a RC circuit under AC, where $R=R_q$ is of pure quantum origin. If there is one quantum channel connecting the plate, then R_q is half conductance quanta per spin.

Confirmed experimentally in 2006.



Gabelli etal. Science, 313, 499 (2006).

At higher frequency (~5GHz), RLC circuit fits better:

Classical RLC:

$$G(\omega) = -i\omega C_\mu / (1 - \omega^2 L_q C_\mu - i\omega C_\mu R_q)$$

Expand:

$$G(\omega) = -i\omega C_\mu + \omega^2 C_\mu R_q + i\omega^3 C_\mu^2 R_q^2 - i\omega^2 C_\mu^2 L_q$$

Compare to quantum result: $G(\omega) = -i\omega C_\mu(\omega)$

Where we derived:

$$C_\mu(\omega) = C_\mu + i\omega C_\mu^2 \frac{h}{2e^2} - \omega^2 C_\mu^3 \frac{h^2}{4e^4} + \omega^2 C_\mu^2 \frac{h^2}{12\pi\Gamma_L e^2}$$

Wang and Guo, PRB 75, 155336 (2007)

We obtain:

Wang and Guo, PRB 75, 155336 (2007)

$$R_q = \frac{h}{2e^2}$$

$$L_q = \frac{h^2}{12\pi e^2 \Gamma}$$

Because:

$$\tau_d = \frac{4\hbar}{\Gamma}$$

$$L_q = \frac{R_q \tau_d}{12}$$

Conclusion: at high frequency, there is a quantum inductance which is related to the dwell time. This is of pure quantum origin.

Can we calculate these quantities from atomistic point view so real devices can be studied?

Some summary remarks:

- It appears that NEGF-DFT type formalism has emerged as the main theoretical tool for practical atomistic calculations of nanoelectronics. The results for many situations can be directly compared with measured data.
- NEGF-DFT type formalism has direct connection to quantum transport theory which is usually done using NEGF (e.g. molecular superconducting tunnel junction).
- It appears that people in NEGF-DFT are moving toward various practical applications of it, and are not working hard enough for its basic theoretical foundation.
- Technical improvements are more often seen (SIC, GW, CI, etc…).
- A potentially fruitful direction is the TDDFT type approach. There are several recent reports for extending TDDFT to open boundary problems so that transport becomes possible. Only a few real calculations on very small systems exist so far, and even fewer (if at all) compare to any measured data. But this is just the beginning and the real outcome is yet to be seen.
- Size limit so far: a few hundred atoms in the scattering region using LCAO basis; several thousand atoms using LMTO.

Thank you!

$: NSERC, FQRNT, CIFAR, SRC.

Special thanks to RQCHP for computational facility.