Quantum Transport Beyond the Independent-Electron Approximation

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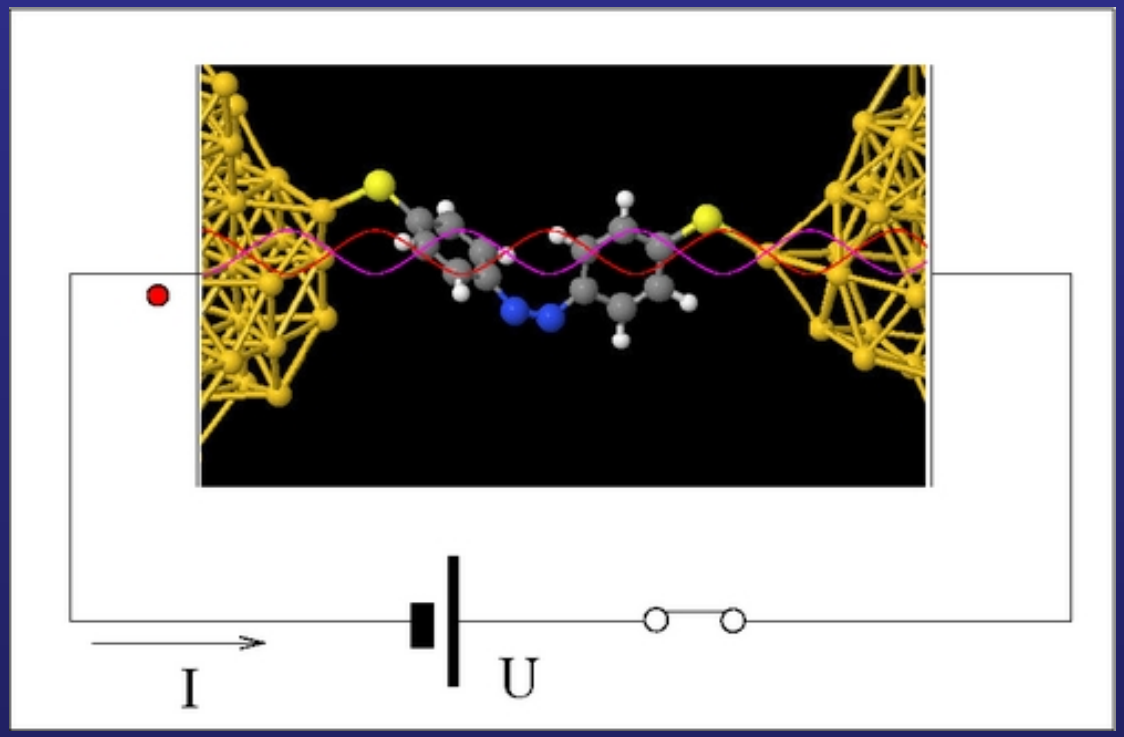
Outline

- Introduction to the quantum transport problem
- *Ab initio* quantum conductance in the presence of e-e interaction (TDDFT / MBPT)
- Stroboscopic wavepacket approach for calculating and interpreting quantum transport

Bothersome aspects of quantum transport



Ab-initio model

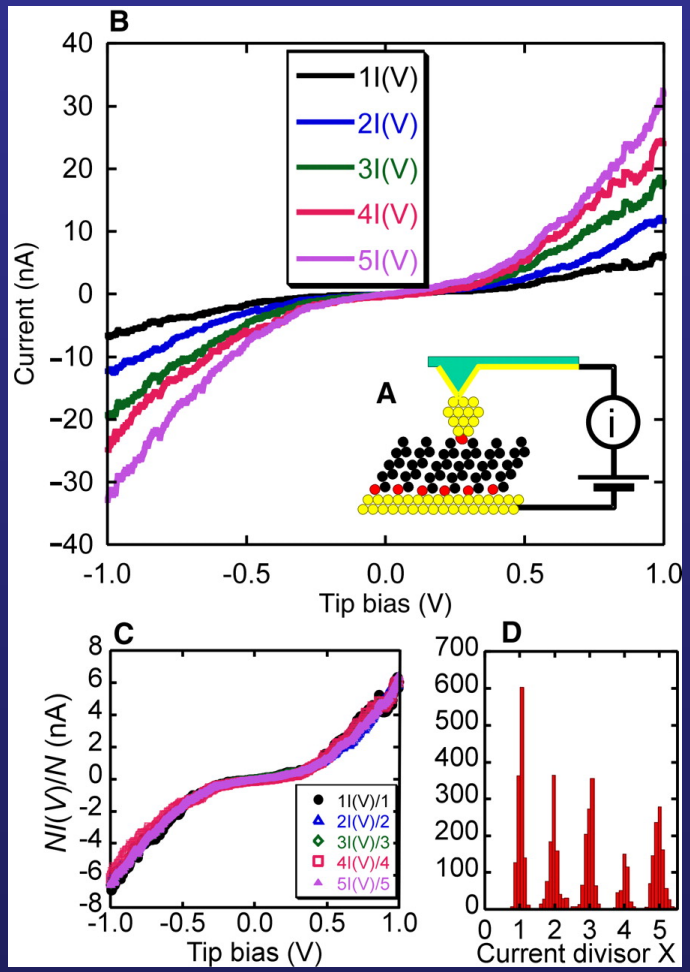
Non-equilibrium

Quantum Mechanics

Many-body problem

$I(t)=I([U])$;
Conductance
$G=I/U$ for
steady state

Experiments & modelling



X. D. Cui et al. Science, **294** 571 (2001)

Experiments:

Octanedithiol/Au: $R \approx 900$ MΩ
[X. D. Cui et al., Science (2001).]
Benzene-di-amin/Au: $R \approx 2$ MΩ
[Quek, Nano Lett. (2007).]
Benzene-di-thiol/Au: $R \approx 18 \pm 12$ MΩ
[M. A. Reed et al. Science (1997).]
H_2/Pt: $G \approx 0.95\ G_0$ ($\approx 1/(13\ \text{k}\Omega)$)
[R.H.M.Smit et al. Nature (2002).]

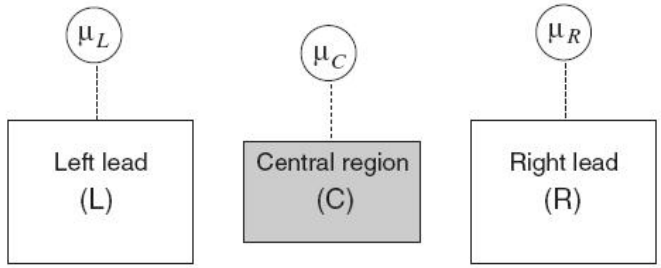
Theory – Density Functional Theory + NEGF:

for $G \approx G_0$ generally good
for $G << G_0$ poor
e.g. $G \approx 0.046\ G_0$ for **Benzene-di-amin/Au**
[Quek, Nano Lett. (2007).]

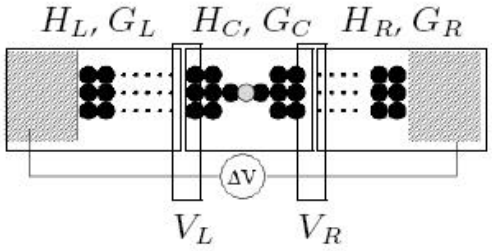
1. Ab Initio Quantum Conductance with e-e Interaction

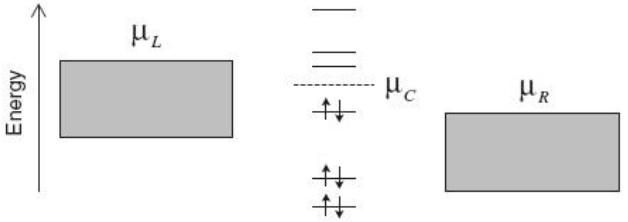
NEGF Laudauer-Büttiker

Mads Brandbyge *et al.* PRB (2002).



- Real difficulty in transport - the system is infinite!!





- Contacting-based formulation

$$G_{CC} = G_C + G_C\,(\delta H_C + \Sigma_{CC}) \cdot G_{CC}, \quad \text{"The Dyson eq. - DFT SCF for centre"}$$

$$\Sigma_{CC} = V_L G_L V_L + V_R G_R V_R, \quad \text{"The leads' self-energies"}$$

$$G_{LC} = G_L V G_{CC} \quad \text{"leads - typically not SCF"}$$

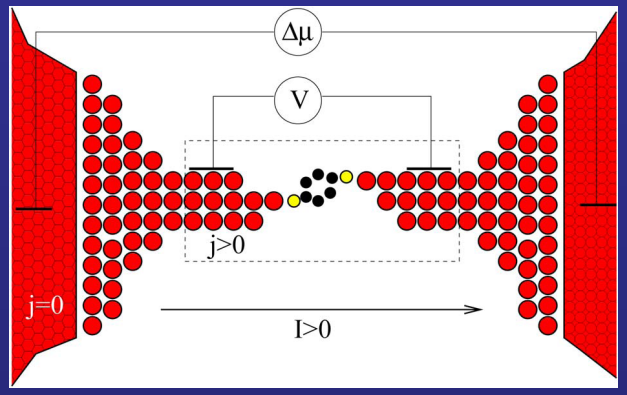
$$I = \frac{2e}{\hbar}\Re\{Tr\left[V G^{<}_{LC}\right]\}$$

Typically leads' SCF neglected ⇒ errors few %

L/r_s	$G_{2P}/G_{\Delta\mu=\Delta\phi}$	$G_{4P}/G_{\Delta\mu=\Delta\phi}$
1.0	0.89	1.24
1.5	0.97	1.06
3.0	1.00	1.00

[Mera, Bokes, Godby PRB 72, 085311 (2005).]

Quantum Transport Theories



$$G = \frac{e^2}{h} T(E_F)$$

- Conductance in 1-electron or mean-field theory given by Landauer formula
- Drawbacks of usual approach:
  - Can be orders of magnitude wrong
  - Difficult to generalise to many-body case
  - Calculation of *T* not readily compatible with periodic bcs

Our Approach

- "Beyond ground-state DFT" description of quantum transport still troublesome
- Formulate the linear-response theory of conductance for rigorous *ab-initio* modelling within a supercell technique:
  - well defined conductance
  - converged basis set
  - realistic e-e interaction

4-point Kubo conductance

Plane-wave basis

GW method

P. Bokes, J. Jung and RWG, PRB 2007

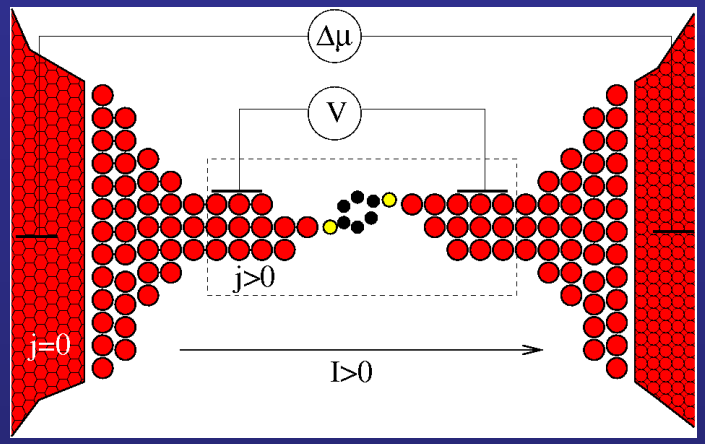
The 4-point conductance

P. Bokes, J. Jung and RWG, PRB 2007

$$\vec{j} = \vec{\sigma} \star \vec{E}^{\text{aux}} = \vec{\sigma}^{\text{irr}} \star (\vec{E}^{\text{aux}} + \vec{E}^{i})$$

?

$$G_{4P} \equiv I/V \; ; G_{2P} \equiv I/\Delta\mu$$



$$G^{4P} = \frac{\mathcal{F}^{\sigma}[\sigma^{\text{irr},e}]}{\mathcal{F}^{\sigma}[\sigma^{\text{irr},e}] - \mathcal{F}^{\sigma}[\sigma^{\text{irr}}]} \times \mathcal{G}^{\sigma}[\sigma^{\text{irr}}]$$

G_{2P}

4-point correction term for conductance of electrode (=1 for constrictions)

$$\mathcal{G}^{\sigma}[\sigma^{\text{irr}}] = \lim_{\alpha \to 0^{+}} \int\int \frac{dq dq'}{2\pi} \sigma^{\text{irr}}(q, q'; i\alpha)$$

$$\mathcal{F}^{\sigma}[\sigma^{\text{irr},e}] = -\lim_{\alpha \to 0^{+}} \int dq \, \sigma^{\text{irr},e}(q, q' = 0; i\alpha)$$

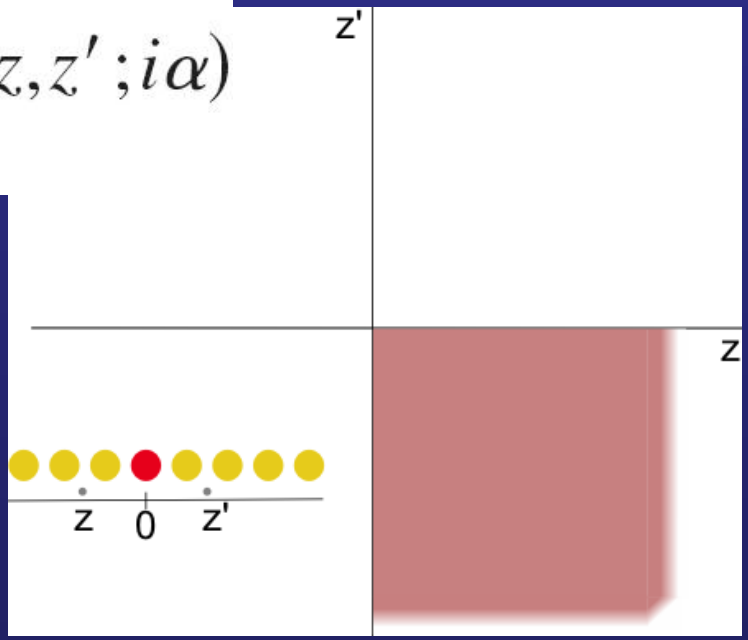
Integrals - real space formulation

"2P" conductance:

$$\mathcal{G}_\alpha[\chi^{\mathrm{irr}}] = \alpha \int_{-L}^{0} \int_{0}^{L} dz dz' \, \chi^{\mathrm{irr}}(z,z';i\alpha)$$

irreducible polarizability:

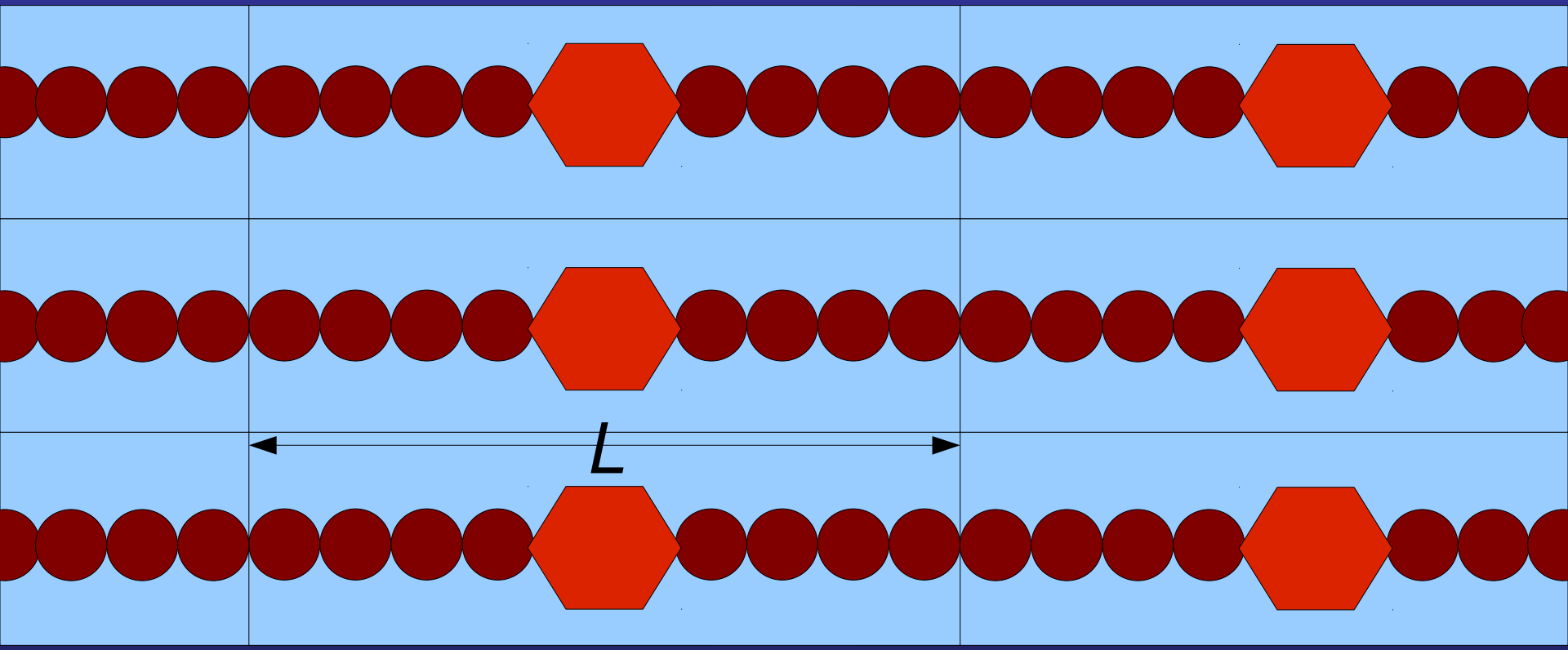
$$\chi^{irr}(z,z') = \frac{\delta n(z)}{\delta V^{tot}(z')}$$



4P Correction factor:

$$\mathcal{F}_\alpha[\chi^{\mathrm{irr}}] = \alpha \int_{-L}^{0} dz \int_{-L}^{L} dz' \, \chi^{\mathrm{irr}}(z,z')z'$$

ω→0 Limit

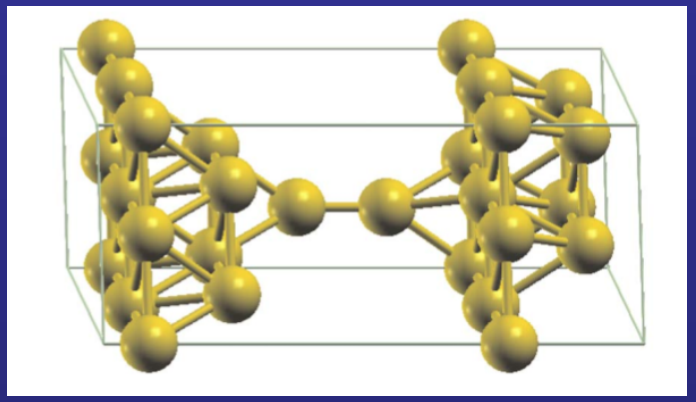


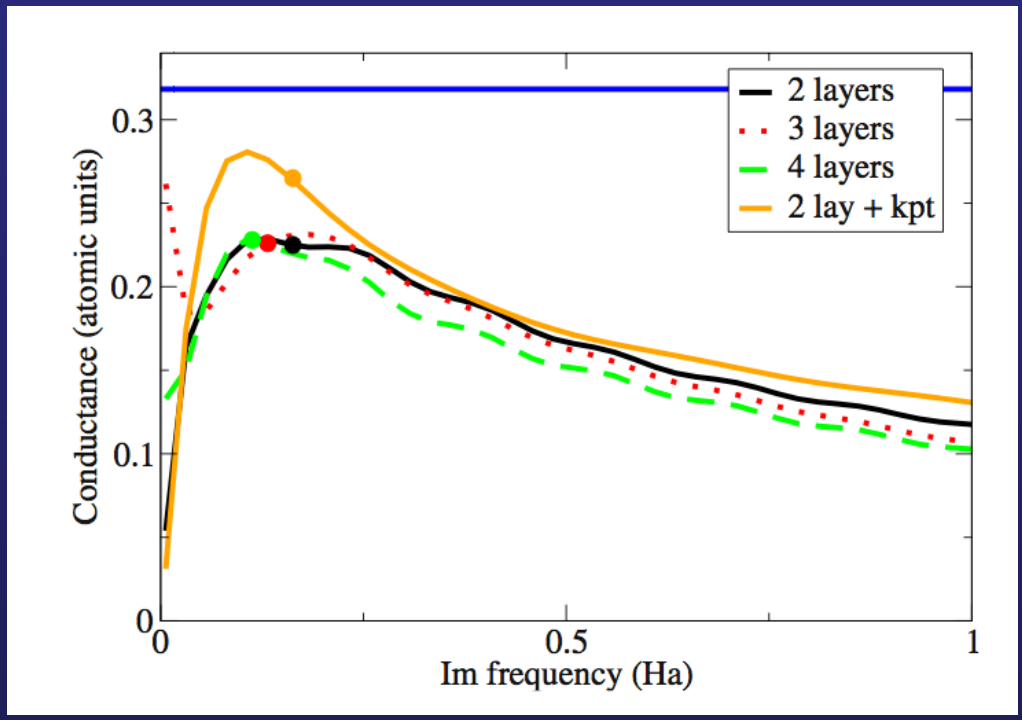
- Moving electron does not “see” neighbouring cell if

$$\frac{2\pi}{\omega} < \frac{L}{v_F}$$

Au wires with structured leads

- 2-atom gold wire between gold electrodes
- Equivalently, a constriction
- HGH-type pseudopotential (6s)
- Convergence w.r.t. electrode thickness





Verstraete, Bokes and Godby, J. Chem. Phys. **130** 124715 (2009)

The GW Approximation

- Iterate Hedin's equations once starting with Σ=0

$$\Sigma(1,2) = \mathrm{i}\int W\left(1^{+},3\right) G(1,4)\,\Gamma(4,2,3)\,\mathrm{d}(3,4)$$

$$P(1,2) = -\mathrm{i}\int G(2,3)\,G(4,2)\,\Gamma(3,4,1)\,\mathrm{d}(3,4)$$

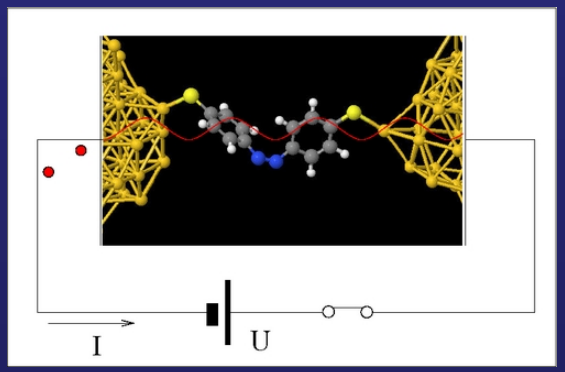
$$W(1,2) = v(1,2) + \int W(1,3)\,P(3,4)\,v(4,2)\,\mathrm{d}(3,4)$$

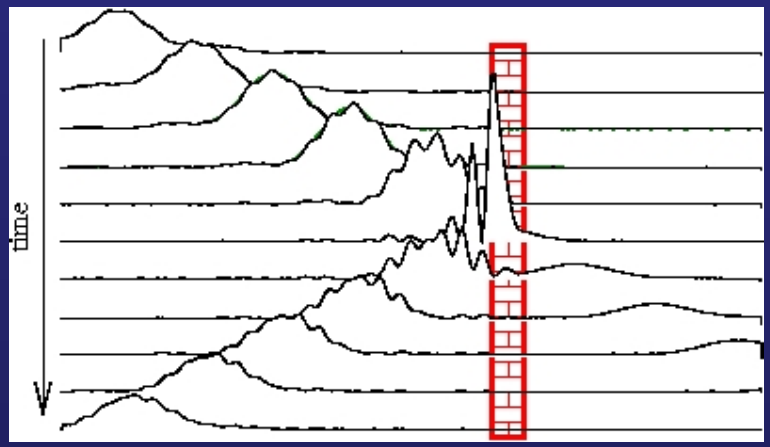
$$\Gamma(1,2,3) = \delta(1,2)\,\delta(1,3) + \int \frac{\delta\Sigma(1,2)}{\delta G(4,5)} G(4,6)\,G(7,5)\,\Gamma(6,7,3)\,\mathrm{d}(4,5,6,7)$$

2. Stroboscopic Wavepacket Approach for Calculating and Interpreting Quantum Transport

Motivation

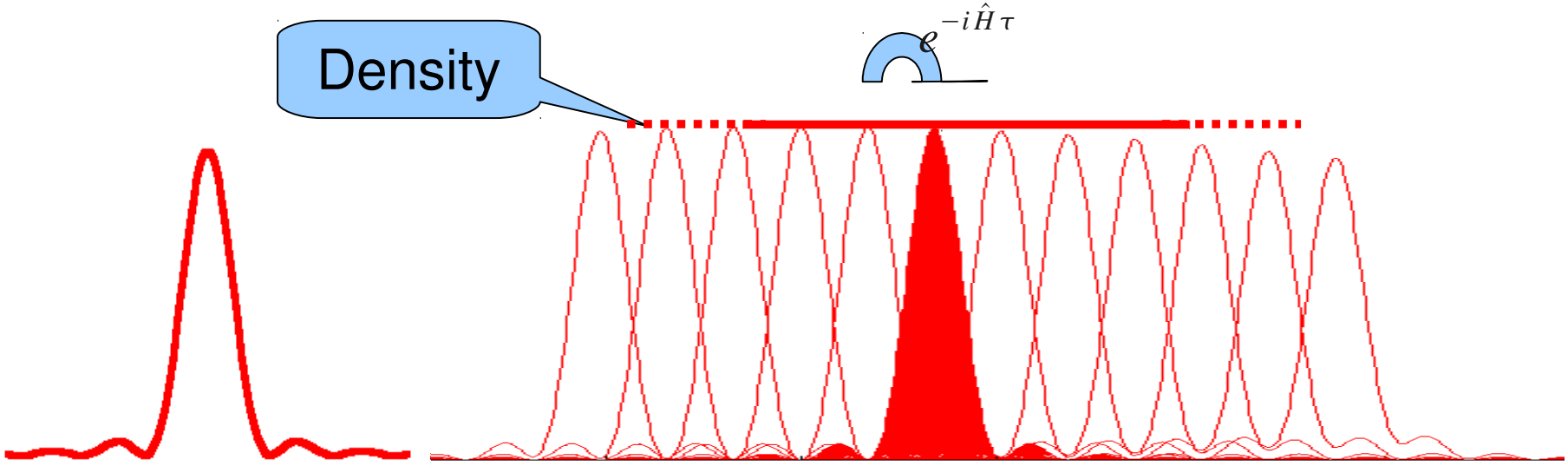
- Non-linear transport?
- Time-dependent transport?
- How about physical insight?





P. Bokes, F. Corsetti and RWG, PRL (July 2008)

Stroboscopic Wavepacket Basis I.



Example: free space in 1D

Stroboscopic Wavepacket Basis II.

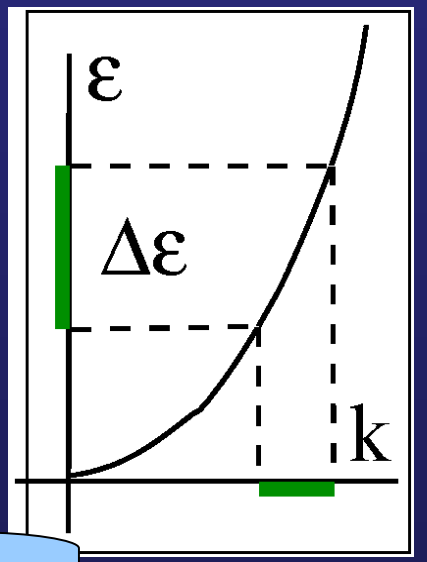
Reference Hamiltonian:

$$\hat{H}\,|\varepsilon,\alpha\rangle = \varepsilon\,|\varepsilon,\alpha\rangle$$

- infinite system
- continuous spectrum
- translational symmetry (locality)

Choice of normalisation of underlying eigenstates:

$$\langle \varepsilon',\alpha' | \varepsilon,\alpha\rangle = \delta(\varepsilon-\varepsilon')\delta_{\alpha,\alpha'}$$



The *initial set of wavepackets* at t=0:

$$|n,0,\alpha\rangle = \frac{1}{\sqrt{\Delta\varepsilon_n}} \int_{\varepsilon_n^\alpha}^{\varepsilon_{n+1}^\alpha} d\varepsilon' U_{\alpha,\alpha'}(\varepsilon')\,|\varepsilon',\alpha'\rangle$$

n – energy-band index

Arbitrary unitary rotation

Stroboscopic Wavepacket Basis III.

Propagation of the initial set to earlier and later times

$$|n,m,\alpha\rangle = e^{-i\hat{H}m\tau_n}|n,0,\alpha\rangle, \quad m = \pm 1, \pm 2, \ldots$$

The time step $\tau_n = 2\pi/\Delta\varepsilon_n$ guarantees orthogonality

$$\langle n,m,\alpha|n,m',\alpha\rangle = \delta_{m,m'}$$

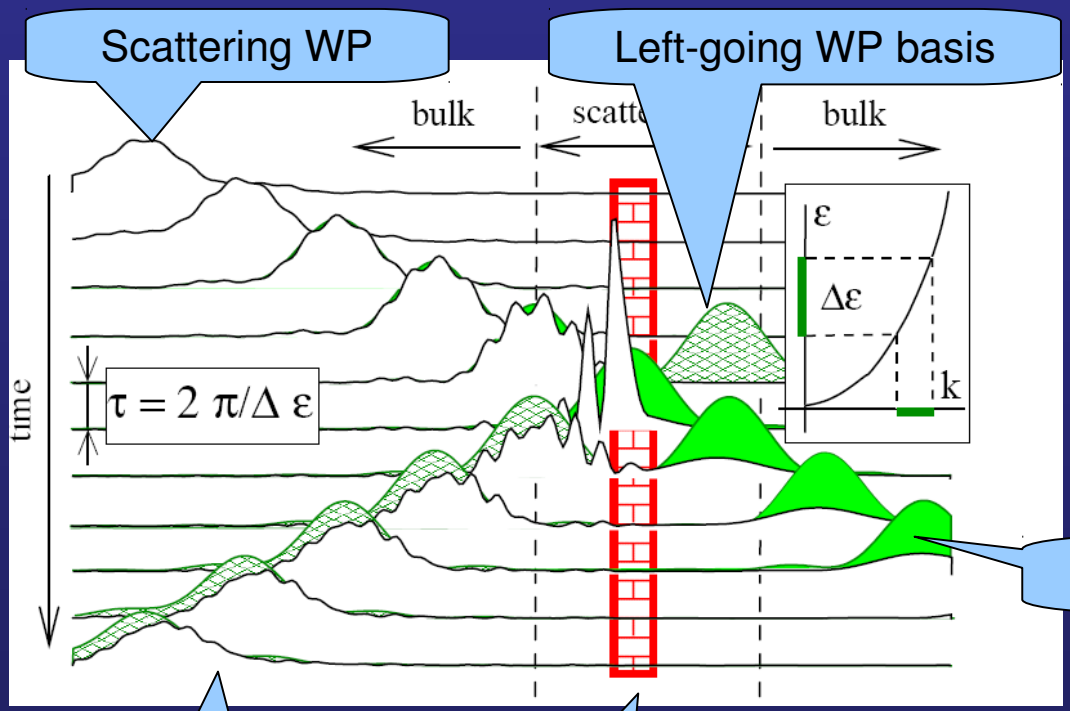
... and completeness

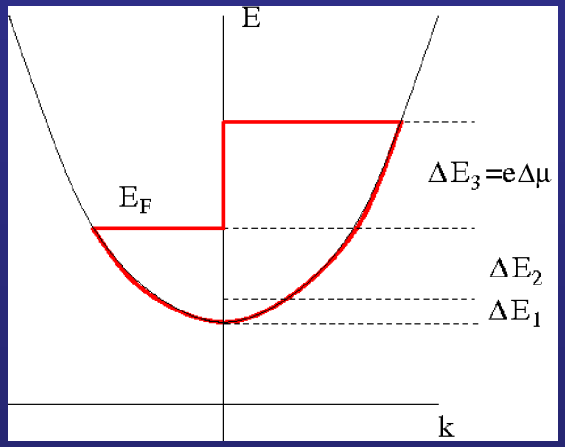
$$\sum_m |n,m,\alpha\rangle\langle n,m,\alpha|\varepsilon,\alpha\rangle = |\varepsilon,\alpha\rangle$$

$$\langle \varepsilon,\alpha|n,m,\alpha\rangle = \frac{1}{\sqrt{\Delta\varepsilon_n}} e^{-i\varepsilon m\tau_n}$$

Stroboscopic propagation

Steady-state transport, Landauer





Right-going WP basis

Reference H

Different H', non-translationally-invariant

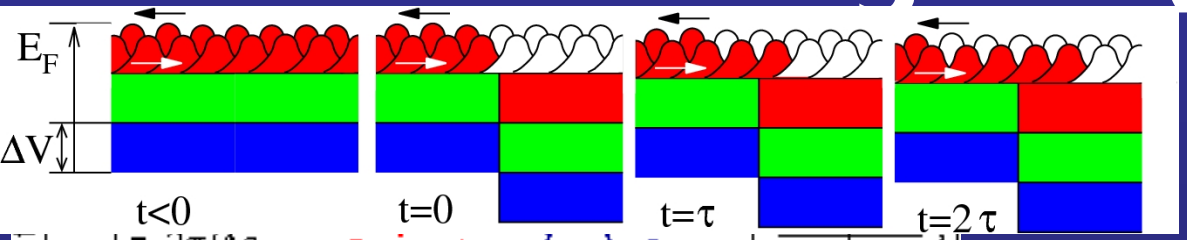
$$I = \frac{(1-R)e}{\Delta\tau} = \frac{1}{2\pi} T \Delta\mu$$

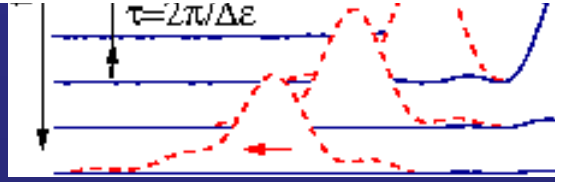
Compactness of basis

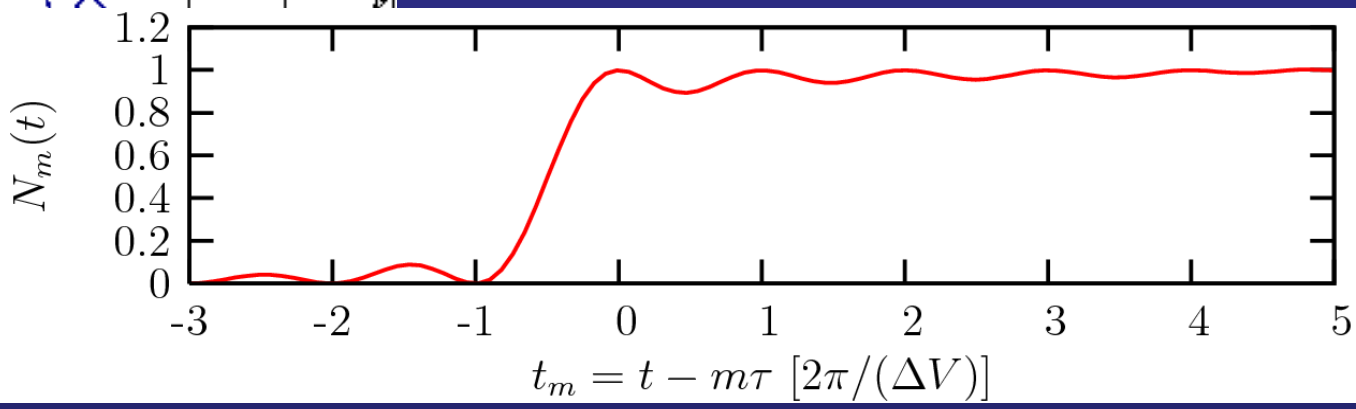
Illustrated for continuous-time propagation

Wavepackets defined for barrier reference potential

Time-dependence: switching-on (1DEG)







$$I_m(t) = \frac{e}{\tau} N_m(t)$$

$$N_m(t) = 2 \sum_{l=0}^{-\infty} |\, (l, +; t \,|m, +\rangle\, |^2$$

In agreement with model td-NEGF from G. Stefanucci and C. O. Almbladh PRB (2004).

$$N_m(t) = \frac{\sin^2(\pi\xi)}{\pi^2} \sum_{l=0}^{\infty} \frac{1}{(l+m-\xi)^2}, \qquad \xi = t/\tau$$

Electron-electron interaction

- Stroboscopic wavepackets automatically apply boundary condition in leads:
  - Incoming current(s)
  - Bias
- Only a small central region needs to be treated using TDDFT or MBPT
- Explicit time propagation exploits compact basis

Summary

- 4-point conductance PRB 2007 JCP 2009
  - well defined for interacting systems
  - numerically feasible in supercell geometry
  - e-e interactions via TDDFT or MBPT
- Stroboscopic wavepacket basis PRL 2008
  - particularly suited for transport problems
  - applications for TD transport and spin Hall effect

http://www-users.york.ac.uk/~rwg3

Collaborators

- Peter Bokes
- Matthieu Verstraete
- Jeil Jung
- Fabiano Corsetti

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