GW-like approaches to quantum transport

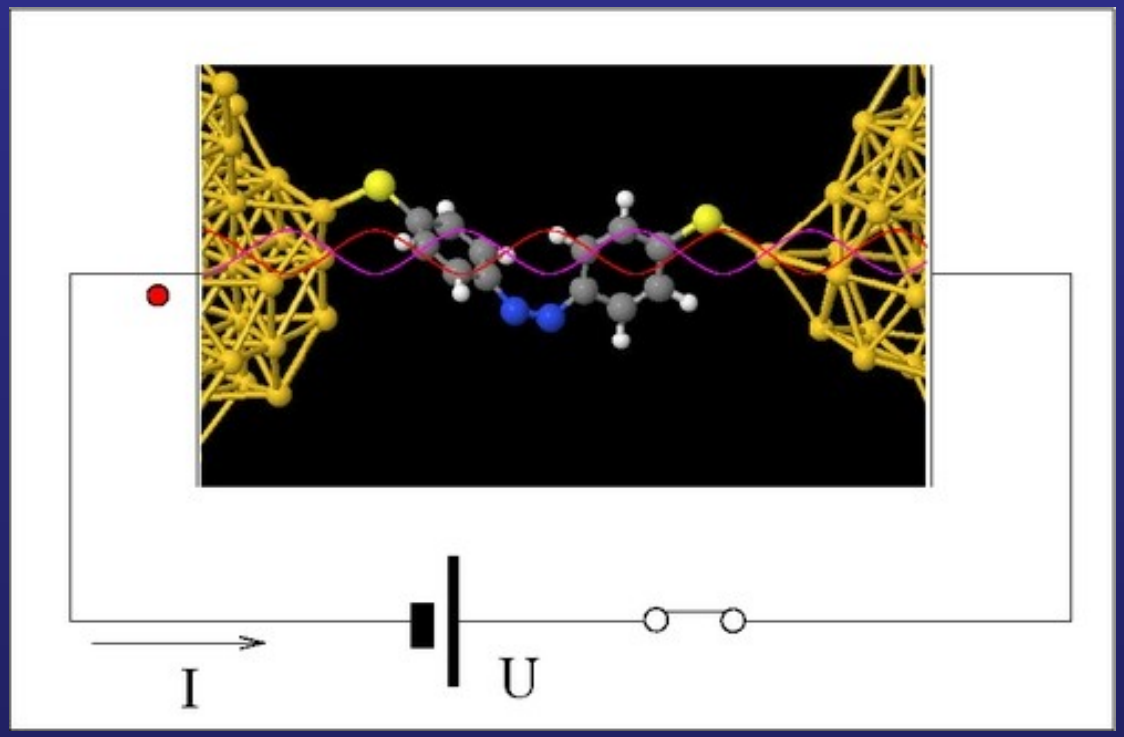
Rex Godby

THE UNIVERSITY of York

Outline

- Introduction to the quantum transport problem
- *Ab initio* quantum conductance in the presence of e-e interaction (TDDFT / MBPT)

Bothersome aspects of quantum transport



Ab-initio model

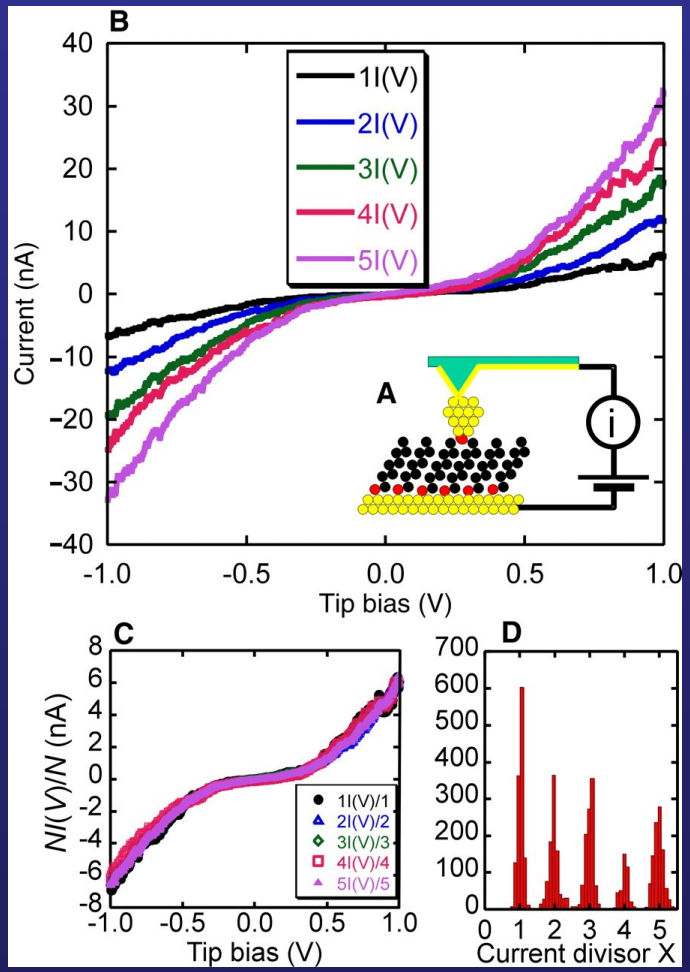
Non-equilibrium

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: $R \approx 900\ \mathrm{M}\Omega$
[X. D. Cui et al., Science (2001).]

Benzene-di-amin/Au: $R \approx 2\ \mathrm{M}\Omega$
[Quek, Nano Lett. (2007).]

Benzene-di-thiol/Au: $R \approx 18 \pm 12\ \mathrm{M}\Omega$
[M. A. Reed et al. Science (1997).]

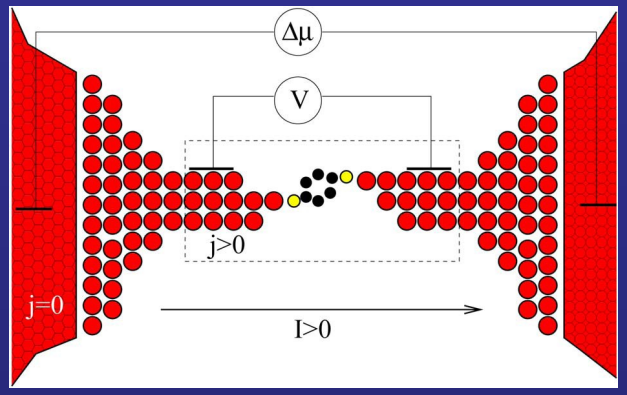
H_2/Pt: $G \approx 0.95\ G_0$ ($\approx 1/(13\ \mathrm{k}\Omega)$)
[R.H.M.Smit et al. Nature (2002).]

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 - Formalism

Quantum Transport Theories

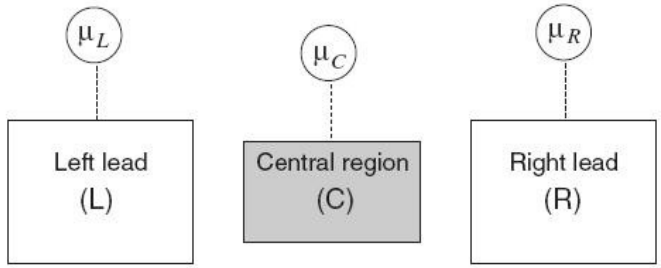


$$G = \frac{e^2}{h} T(E_F)$$

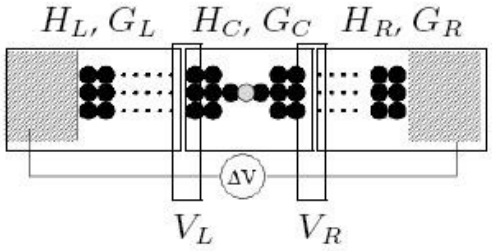
- Conductance in 1-electron or mean-field theory given by Landauer formula
- Drawbacks of usual approach:
  - Can be orders of magnitude wrong
  - Difficult to generalise to many-body case
  - Calculation of *T* not readily compatible with periodic bcs

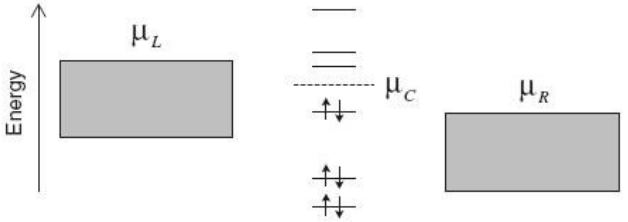
NEGF Laudauer-Büttiker

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).]

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

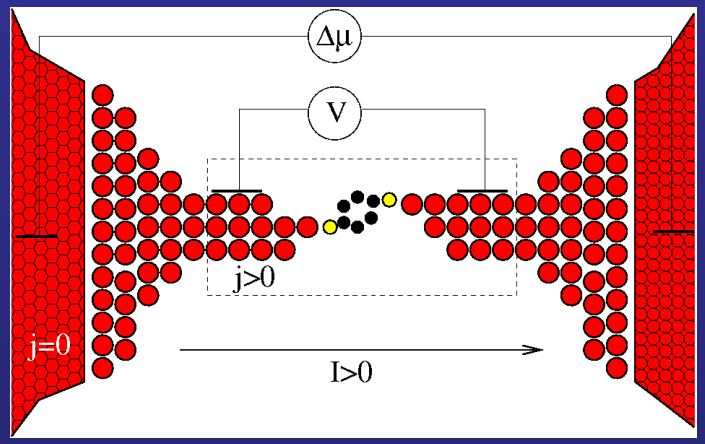
The 4-point conductance

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)

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

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

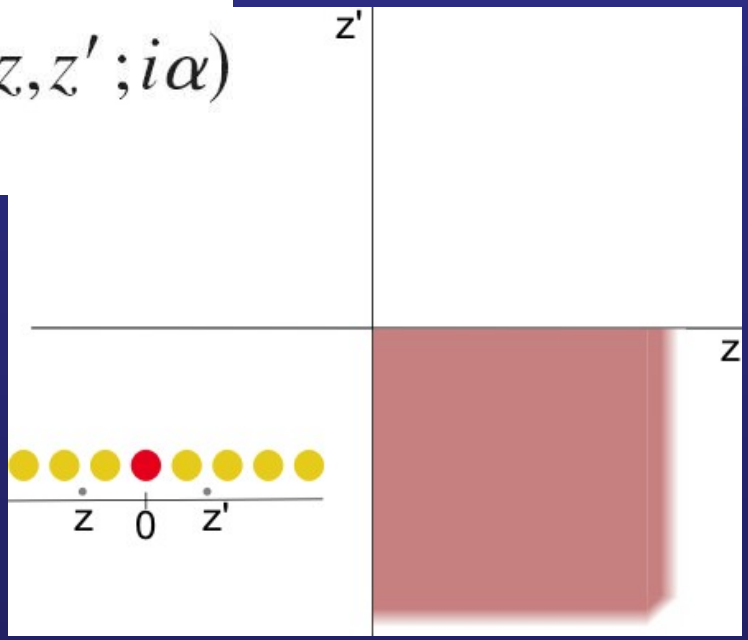
Integrals - real space formulation

"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:

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



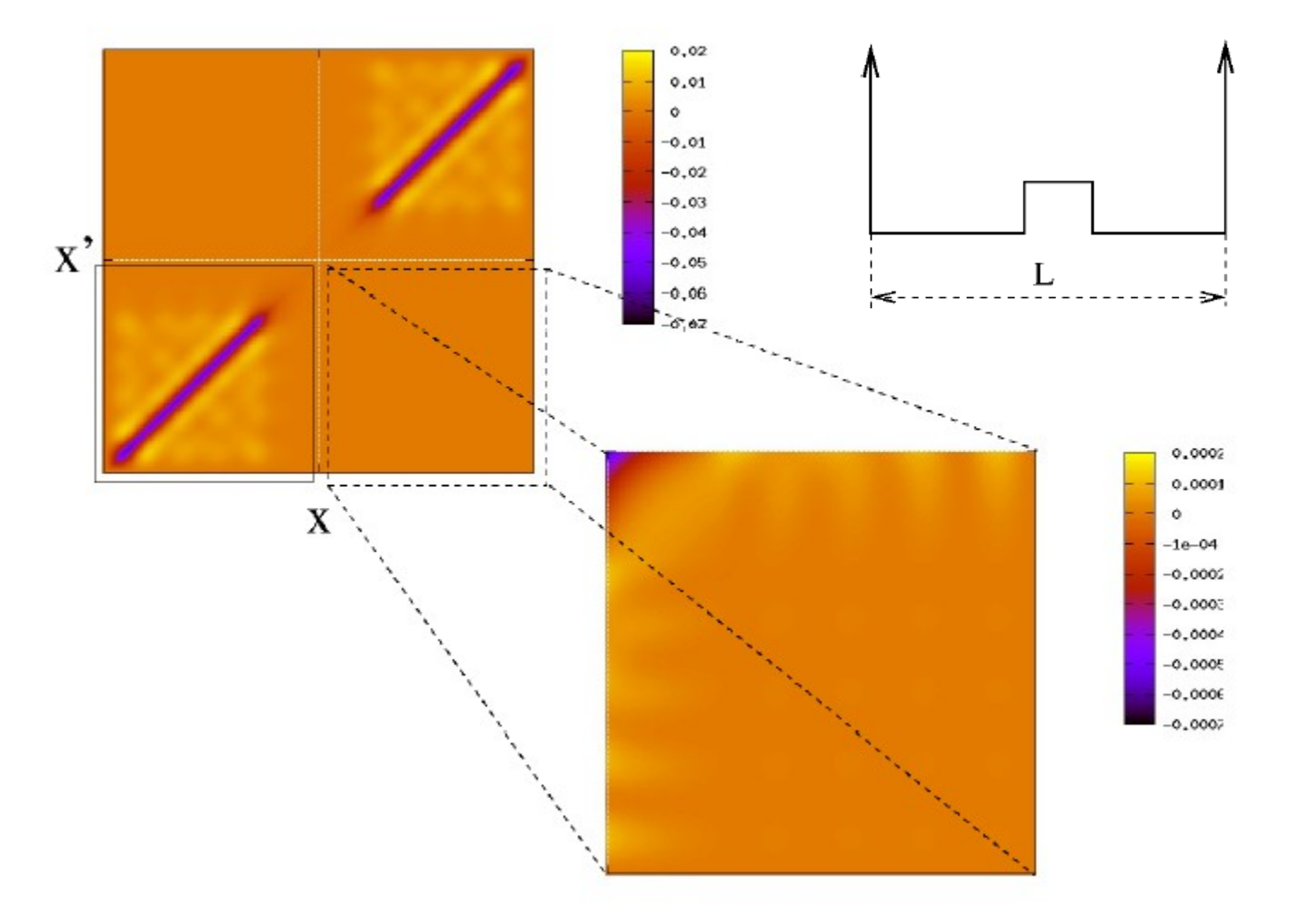
4P Correction factor:

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

2. Implementation and Convergence

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

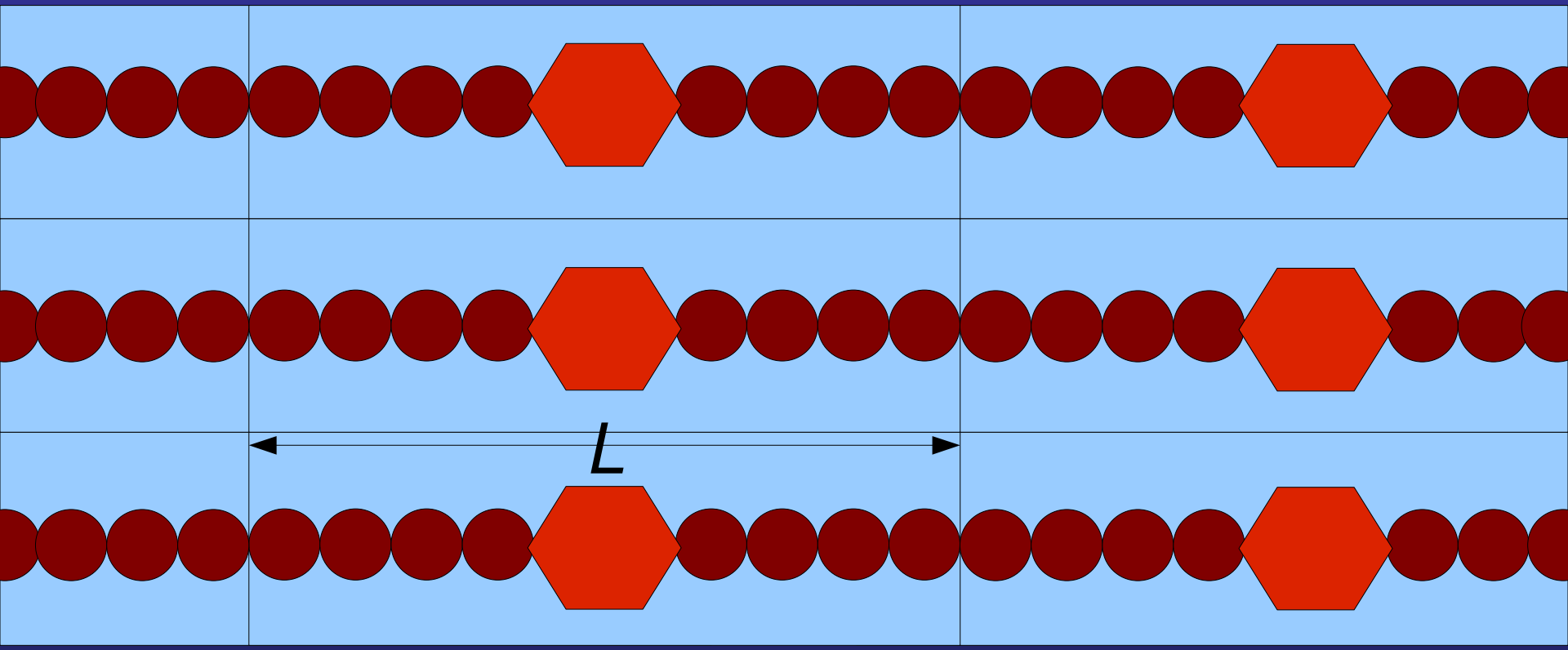
Real-space integrals



Finite-size convergence tests

- Study two simple systems for which G can be calculated analytically as a function of system size (also for infinite size)
  - 1D jellium wire
  - 1D tight-binding model
- Calculate at T=0 and $T \sim E_F$ or bandwidth

ω→0 Limit

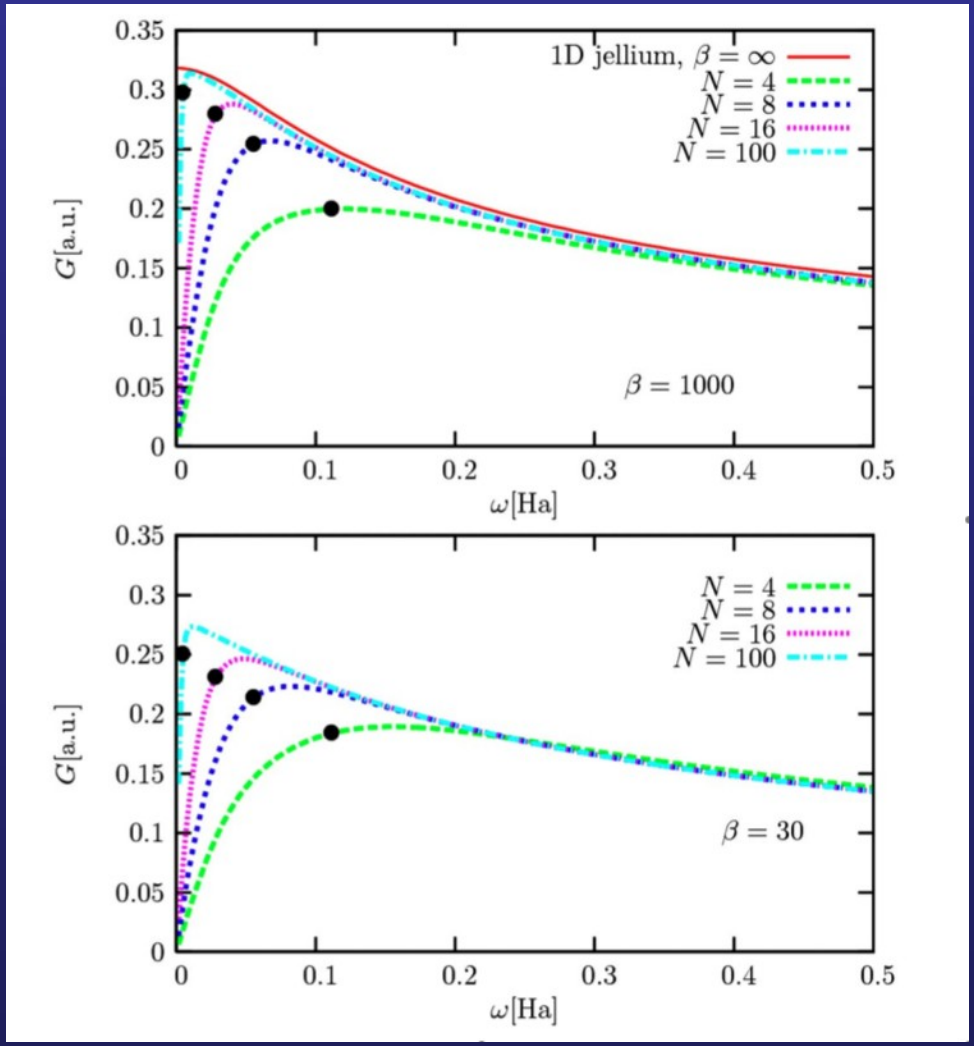


- Moving electron does not “see” neighbouring cell if

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

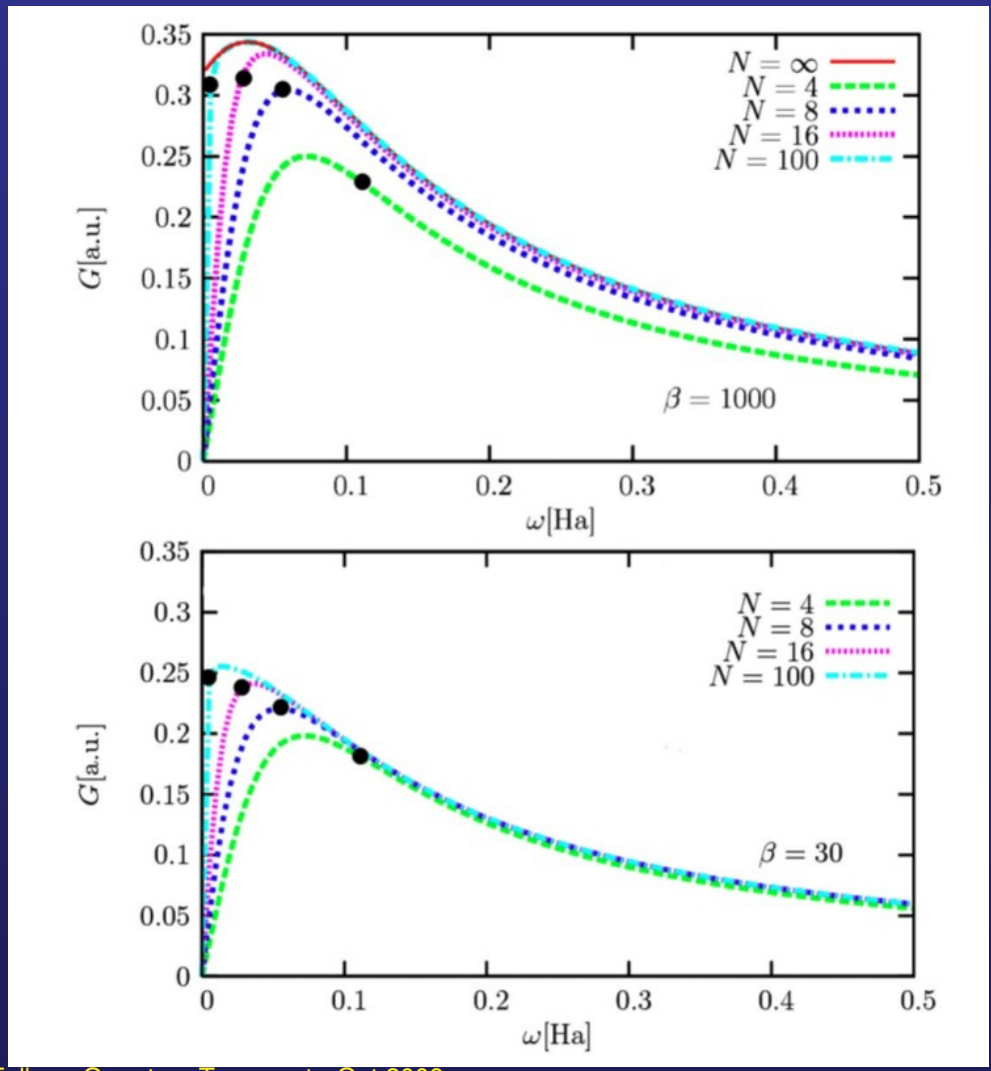
Finite-size convergence tests - Jellium wire

- N = no. of Na-equivalent atoms
- Dot indicates $\omega_{min} = 2\pi v_F / L$
- Convergence with N



Finite-size convergence tests – Tight-binding wire

- N = no. of tight-binding atoms
- Dot indicates $\omega_{min} = 2\pi v_F / L$
- Convergence with N

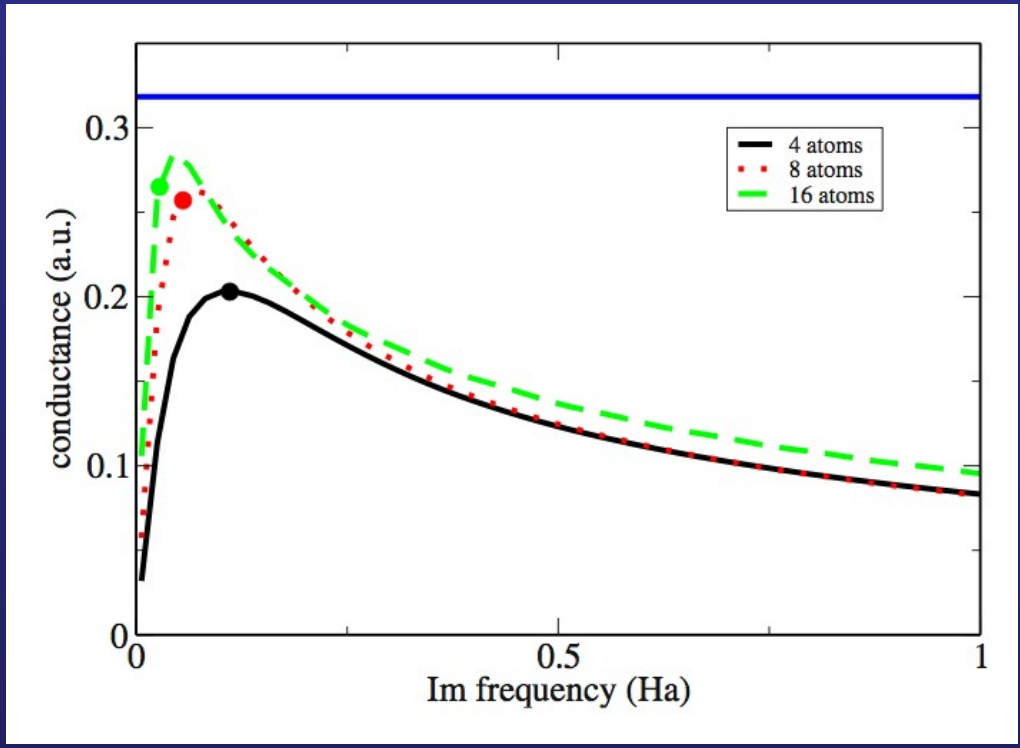


3. Results for Real Atomic Wires

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

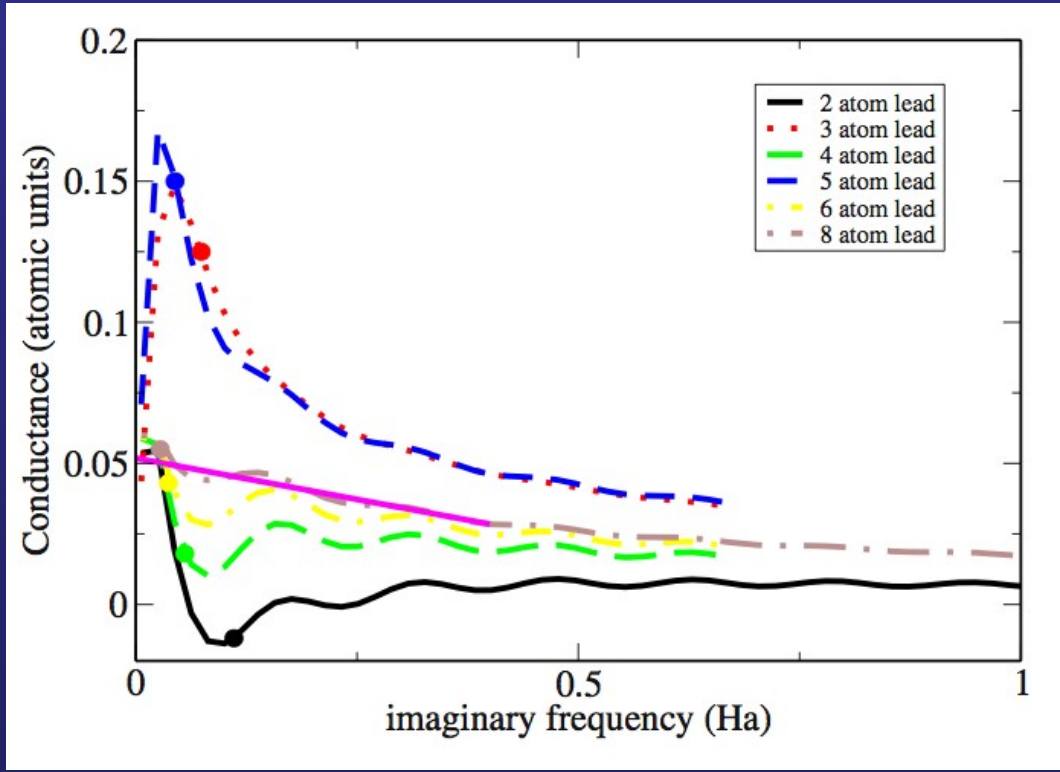
Uniform Na monowires

- Troullier-Martins pseudopotential
- RPA, equivalent to Landauer-Büttiker
- Plane-wave basis set
- Good convergence of extrapolated value with 8-atom cell, to expected 2.0 quanta (shown in blue)



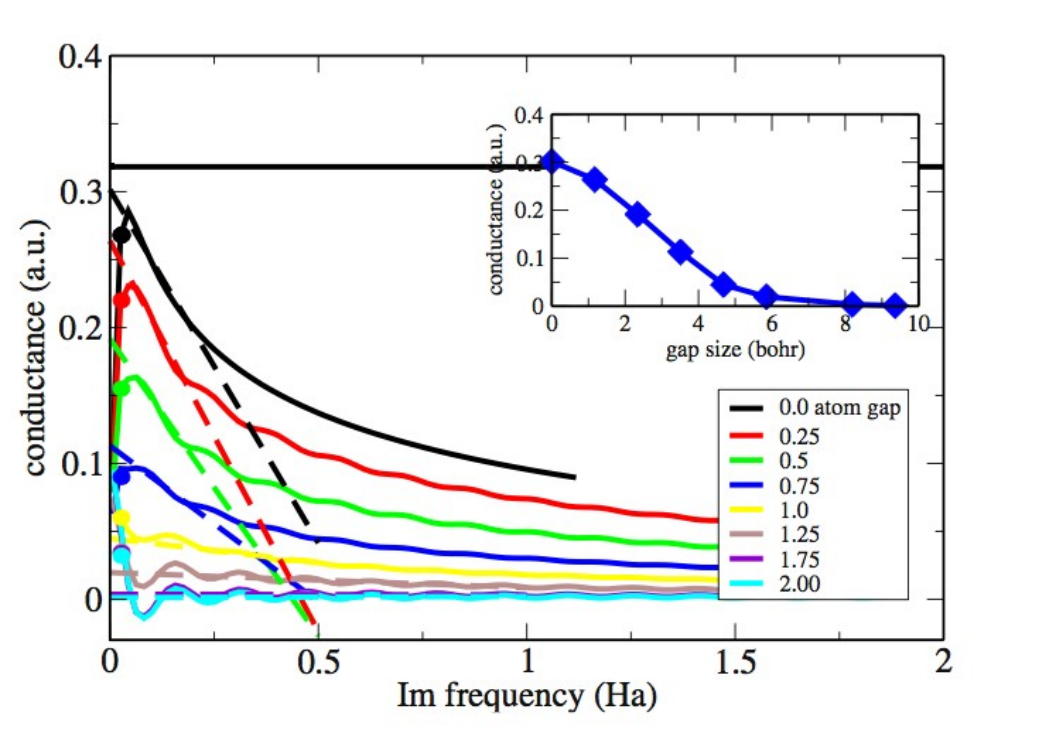
Na monowires with a gap

- Missing atom (e.g. 2+gap+2)
- Good extrapolation available for even nos. (odd has wrong Fermi energy)



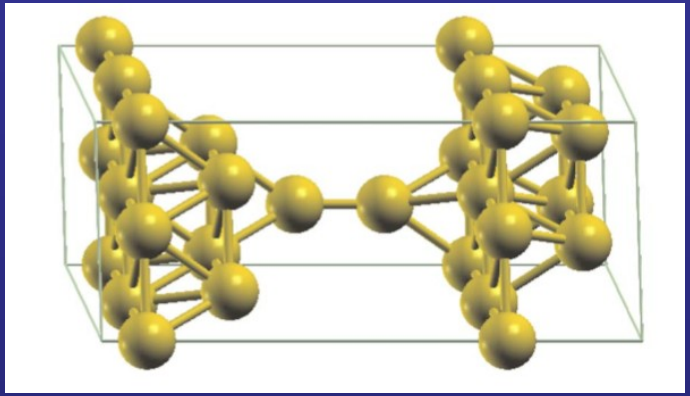
Na monowires with a gap (2)

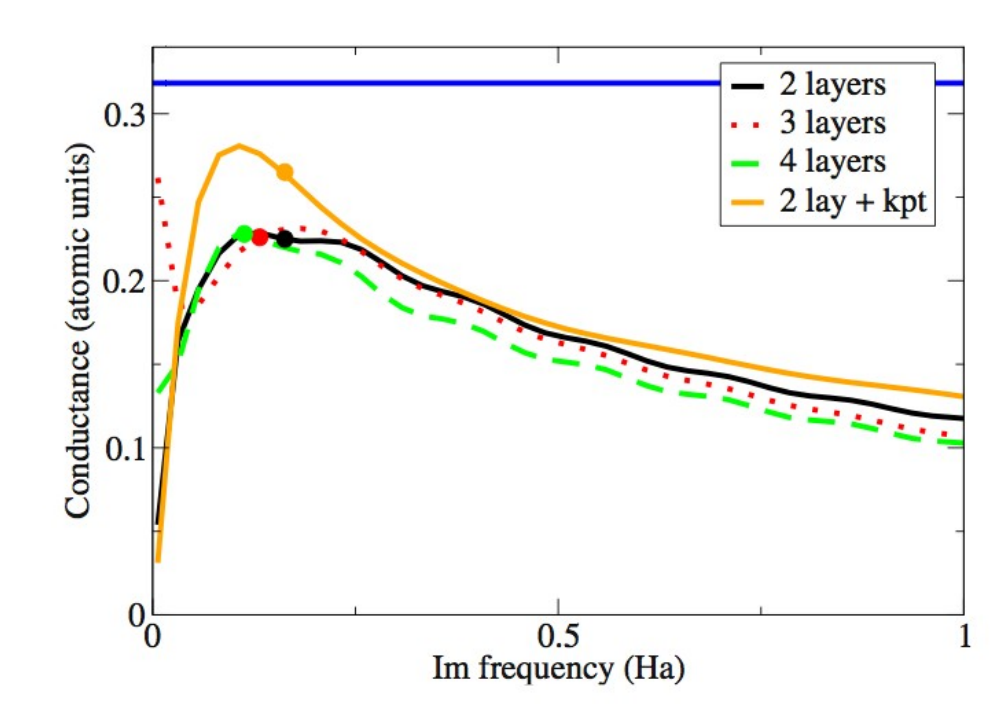
- As a function of gap size (8-atom leads)
- Good extrapolation



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





4. Beyond RPA: Many-Body Effects

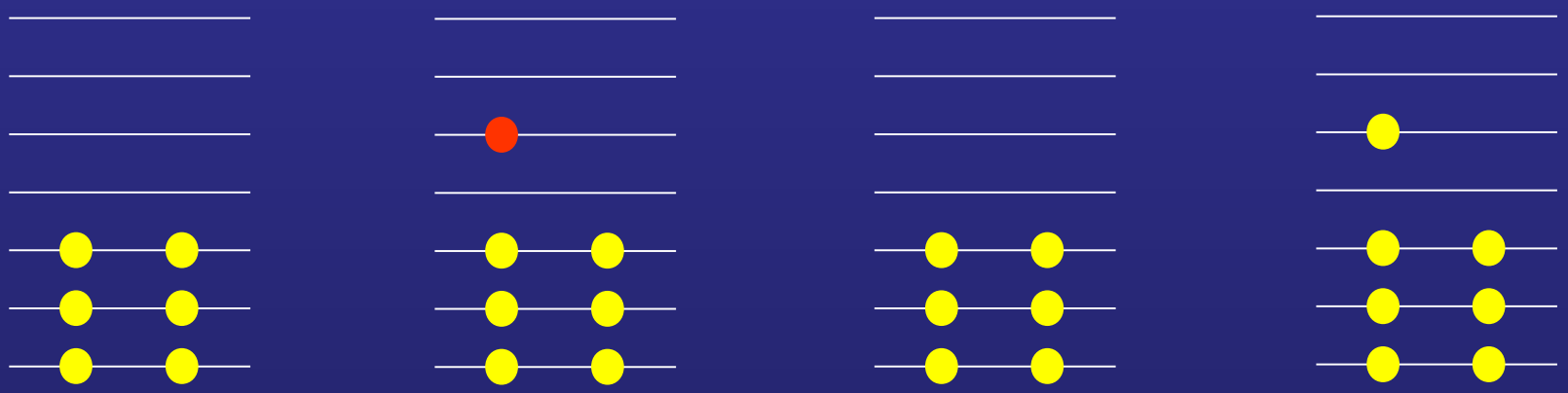
Calculating χ including interactions

- Time-dependent density-functional theory
  - e.g. J. Jung, P. Bokes, and RWG, PRL 2007
  - *but* the form of the XC kernel is delicate
    - Na Sai, PRL(2005), Koentopp PRB(2006), Toher PRL(2005), Burke PRL(2005)
- *or,* Many-body perturbation theory

$$\chi(r,r';i\tau)=-2G(r,r';i\tau)G(r',r;-i\tau)$$

+ vertex diagrams

Electronic Excitations



$|\, N,0\rangle$ $|\, N+1,s\,\rangle$ $|\, N-1,s\,\rangle$ $|\, N,s\,\rangle$

Hedin's Equations

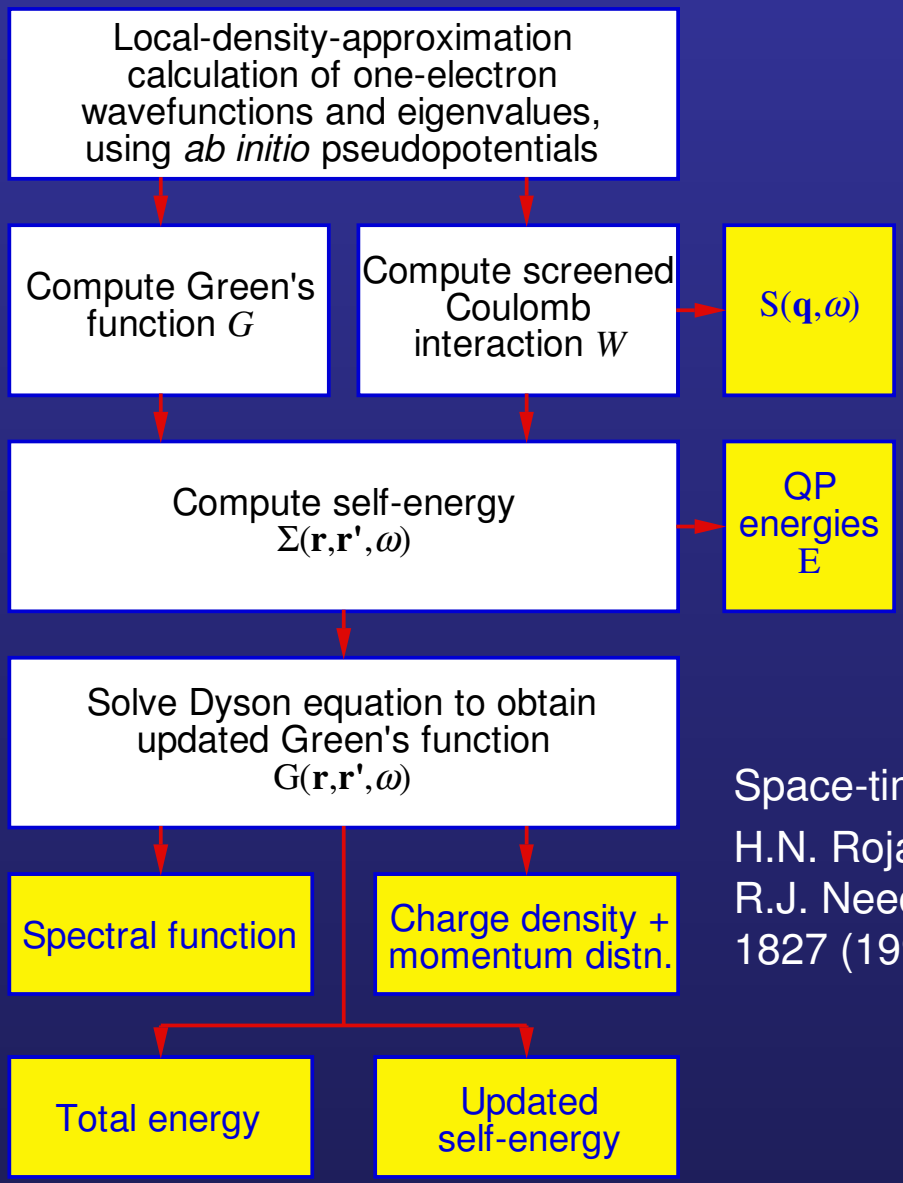
$$
\begin{aligned}
\Sigma(1,2) &= \mathrm{i}\int W\left(1^{+},3\right) G(1,4)\,\Gamma(4,2,3)\,\mathrm{d}(3,4) \\
P(1,2) &= -\mathrm{i}\int G(2,3)\,G(4,2)\,\Gamma(3,4,1)\,\mathrm{d}(3,4) \\
W(1,2) &= v(1,2)+\int W(1,3)\,P(3,4)\,v(4,2)\,\mathrm{d}(3,4) \\
\Gamma(1,2,3) &= \delta(1,2)\,\delta(1,3) \\
&+ \int \frac{\delta\Sigma(1,2)}{\delta G(4,5)} G(4,6)\,G(7,5)\,\Gamma(6,7,3)\,\mathrm{d}(4,5,6,7)
\end{aligned}
$$

- With Σ/G relation, exact closed equations for G, Σ etc.

The GW Approximation

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

$$
\begin{aligned}
\Sigma(1,2) &= \mathrm{i}\int W\left(1^{+},3\right) G(1,4)\,\Gamma(4,2,3)\,\mathrm{d}(3,4) \\
P(1,2) &= -\mathrm{i}\int G(2,3)\,G(4,2)\,\Gamma(3,4,1)\,\mathrm{d}(3,4) \\
W(1,2) &= v(1,2)+\int W(1,3)\,P(3,4)\,v(4,2)\,\mathrm{d}(3,4) \\
\Gamma(1,2,3) &= \delta(1,2)\,\delta(1,3) \\
&+ \int \frac{\delta\Sigma(1,2)}{\delta G(4,5)} G(4,6)\,G(7,5)\,\Gamma(6,7,3)\,\mathrm{d}(4,5,6,7)
\end{aligned}
$$



Space-time method:

H.N. Rojas, RWG and R.J. Needs, Phys. Rev. Lett. **74** 1827 (1995)

GW(ST) method

- Corrects band-gaps and band alignment
- Introduces finite lifetime
- *GW* and quantum transport:
  - Thygesen JCP (2007), Darancet PRB (2007), Neaton PRL (2007), ...
- Convenient implementation: real-space / imaginary-time:
  - Rojas, RWG, Needs PRL (1995)
- Finite temperatures (metals): Verstraete (2008)

[Conference Talk 3 November: Stroboscopic Wavepacket Approach for Calculating and Interpreting Quantum Transport]

Acknowledgements and Summary

Collaborators

- Peter Bokes
- Matthieu Verstraete
- Jeil Jung

European Theoretical Spectroscopy Facility

an initiative of the Nanoquanta Network of Excellence

Summary

- Response-function approach to conductance PRB 2007, JCP 2009
  - well defined for interacting systems
  - numerically feasible in supercell geometry
  - results for Na and Au nanowire systems
  - future: e-e interactions via MBPT or TDDFT

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