Quantum Transport: electron-electron and electron-phonon effects

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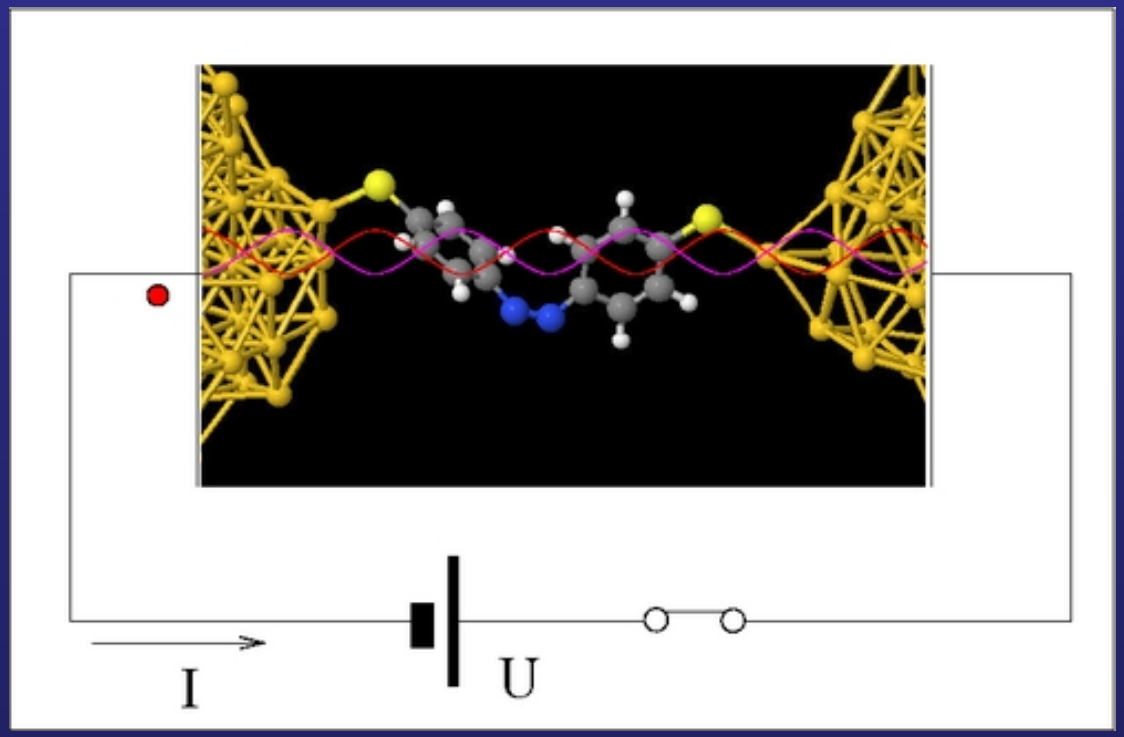
Outline

- Introduction to the quantum transport problem
- *Ab initio* quantum conductance in the presence of e-e interaction (TDDFT / MBPT)
- Beyond conductance: *I*(*V*), spectral functions with e-ph interaction

Bothersome aspects of quantum transport



Ab-initio model

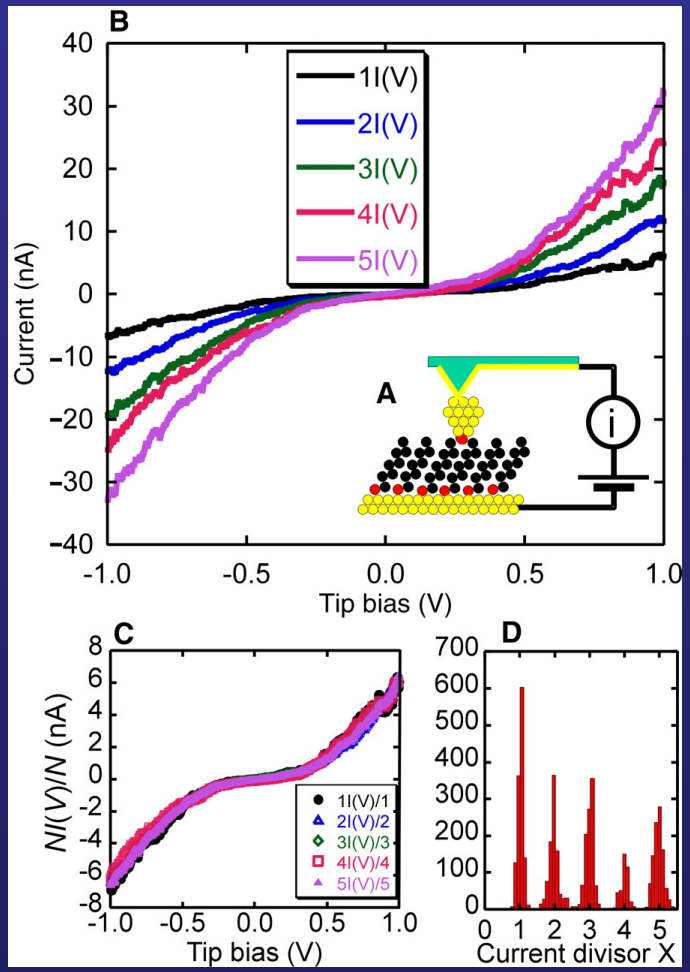
Non-equilibrium

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:

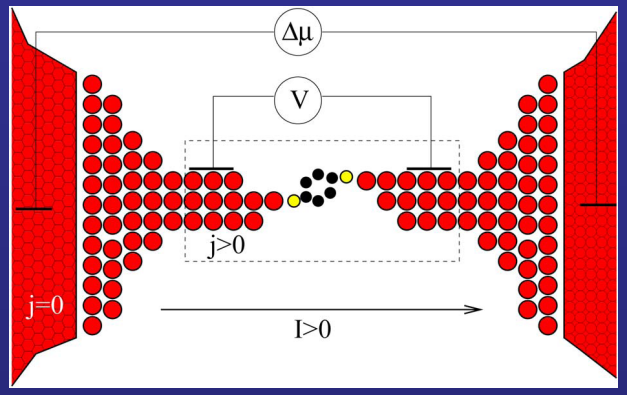
Octanedithiol/Au: $R \approx 900$ MΩ
[X. D. Cui et al., Science (2001).]
Benzene-di-amin/Au: $R \approx 2$ MΩ
[Quek, Nano Lett. (2007).]
Benzene-di-thiol/Au: $R \approx 18 \pm 12$ MΩ
[M. A. Reed et al. Science (1997).]
H_2/Pt: $G \approx 0.95\ G_0$ ($\approx 1/(13\ k\Omega)$)
[R.H.M.Smit et al. Nature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

Quantum Transport Theories

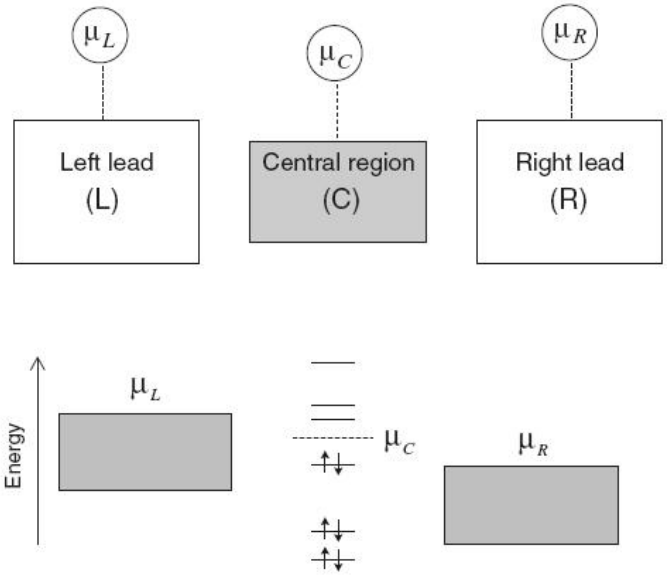


$$G = \frac{e^2}{h} T(E_F)$$

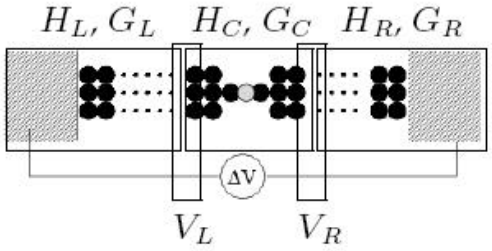
- Conductance in 1-electron or mean-field theory given by Landauer formula
- Drawbacks of usual approach:
  - Can be orders of magnitude wrong
  - Difficult to generalise to many-body case
  - Calculation of *T* not readily compatible with periodic bcs

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

- TDDFT functionals for quantum transport still problematic
- 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

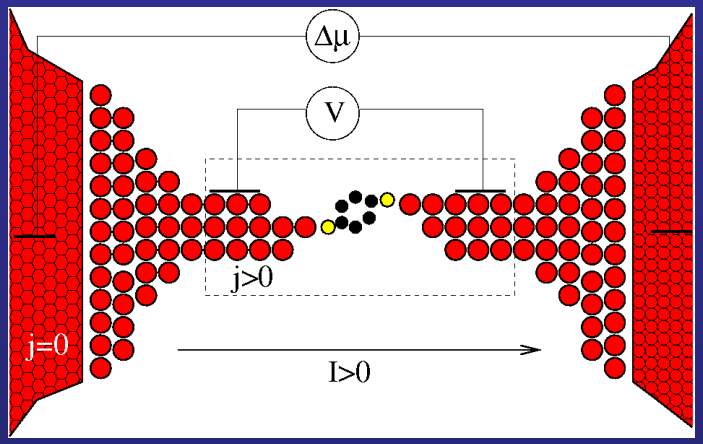
The 4-point conductance

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)

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

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

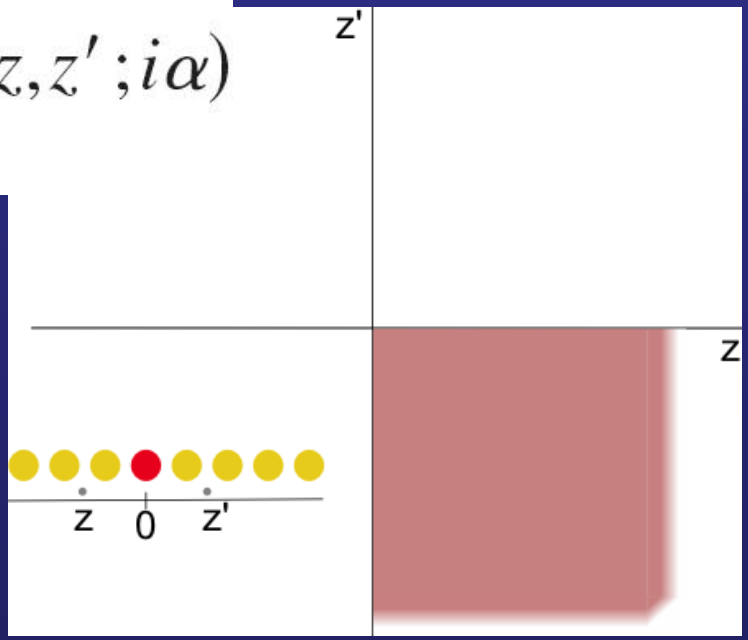
Integrals - real space formulation

"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:

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



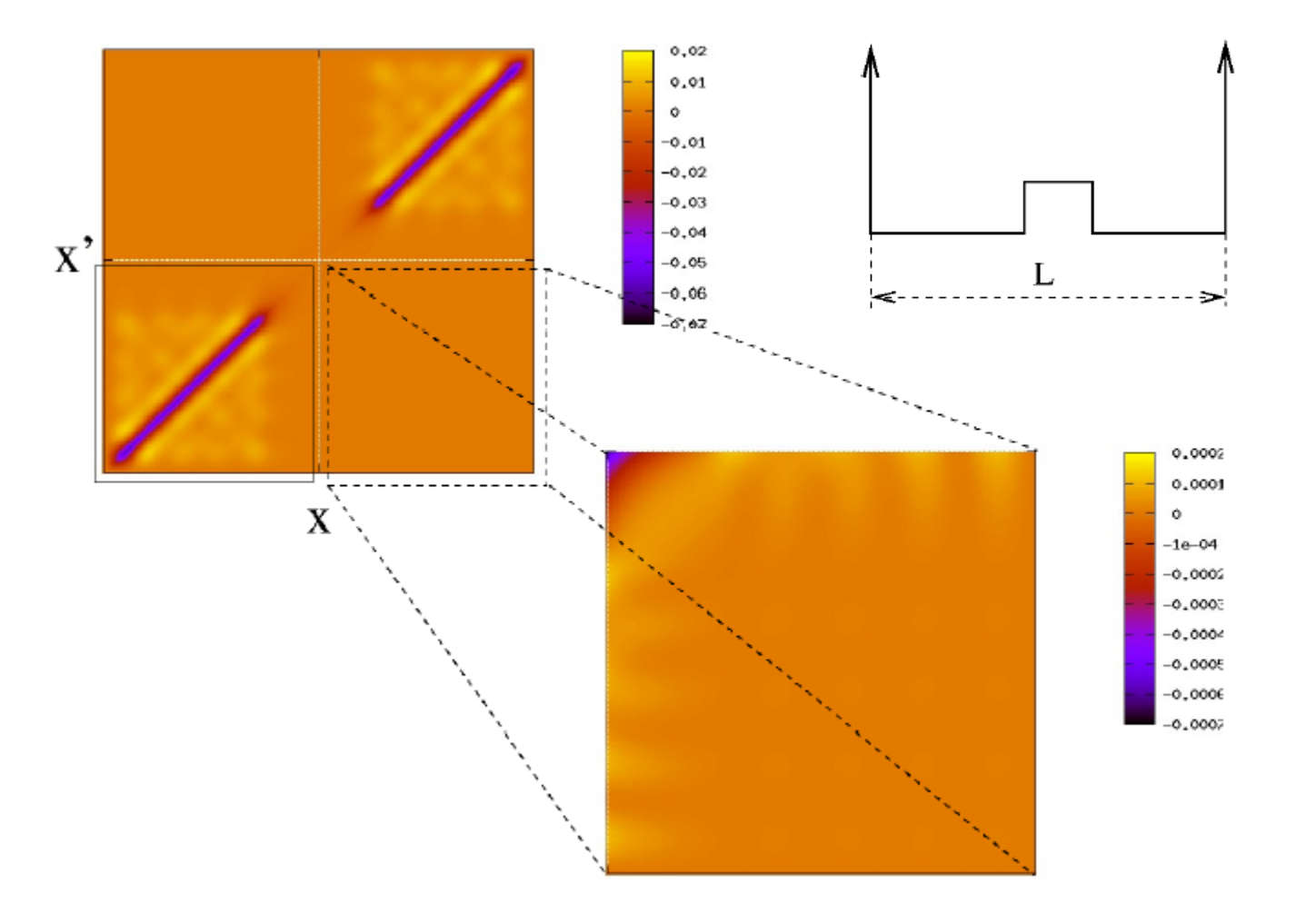
4P Correction factor:

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

Implementation and Convergence

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

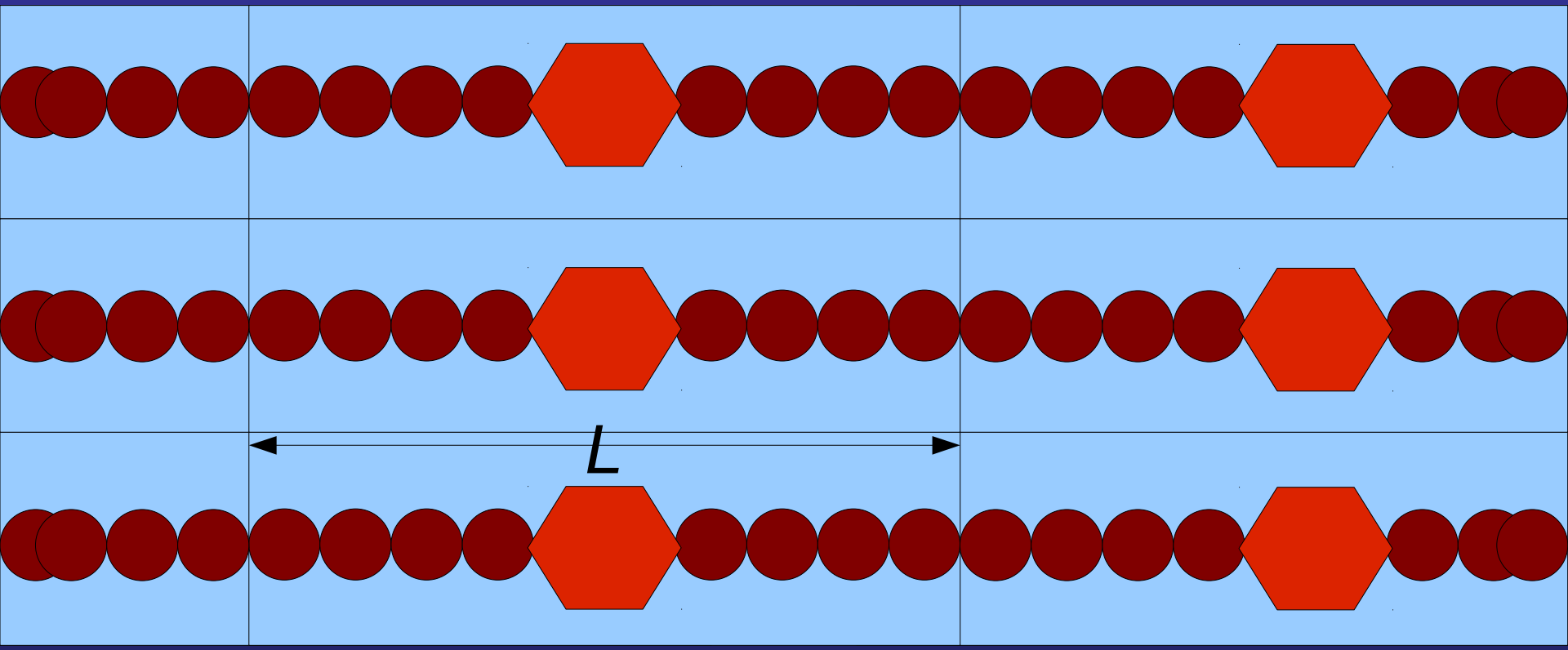
Real-space integrals



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

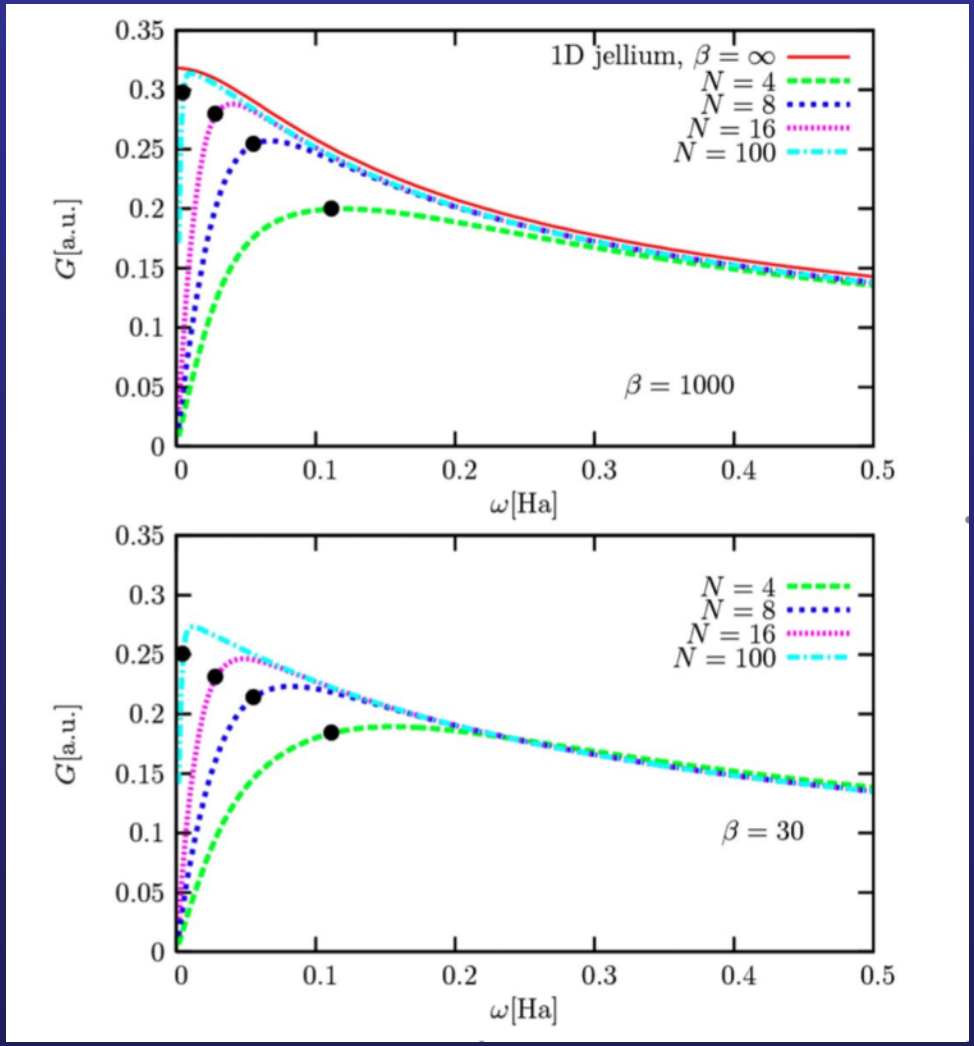
$$\frac{2\pi}{\omega} < \frac{L}{v_F}$$

Finite-size convergence tests – Jellium wire

- N = no. of Na-equivalent atoms
- Dot indicates

$$\omega_{min} = 2\pi v_F / L$$

- Convergence with N

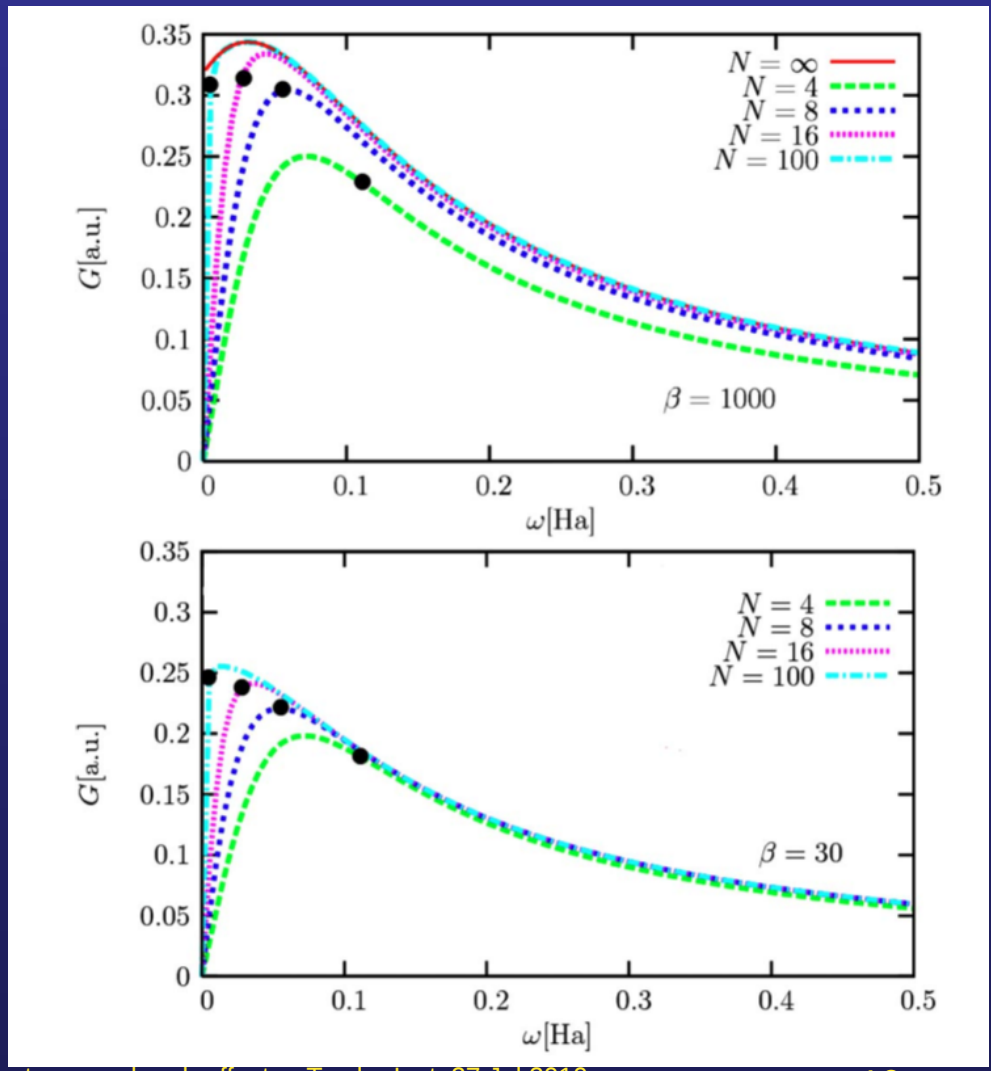


Finite-size convergence tests – Tight-binding wire

- N = no. of tight-binding atoms
- Dot indicates

$$\omega_{min} = 2\pi v_F / L$$

- Convergence with N

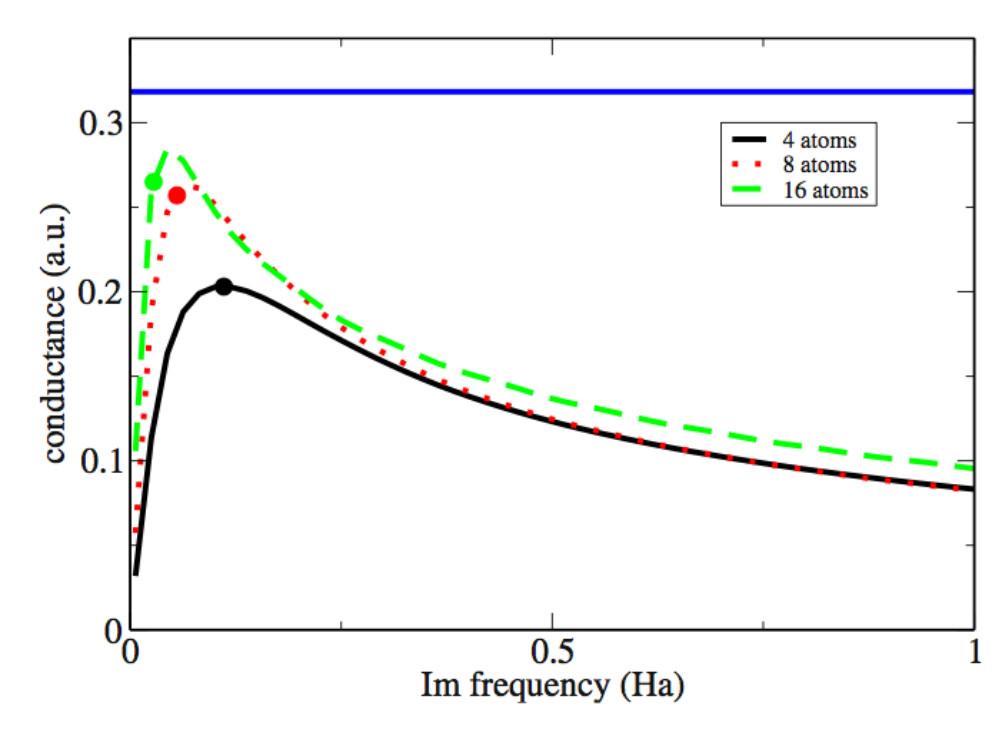


Results for Real Atomic Wires

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

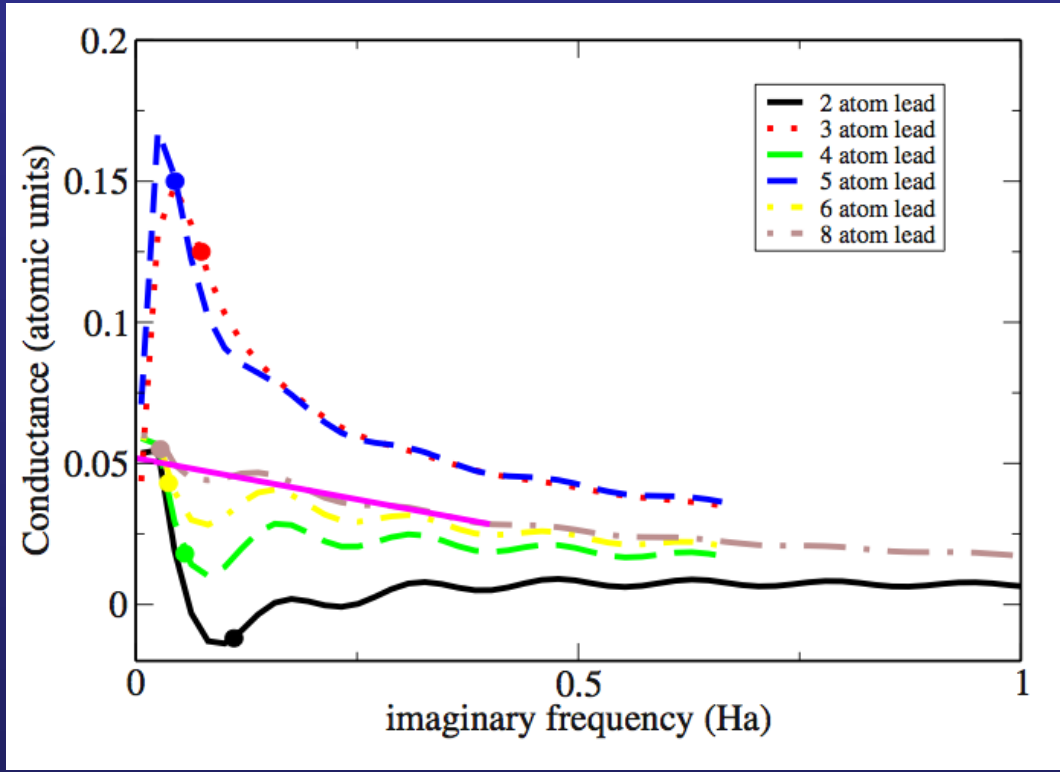
Uniform Na monowires

- 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)



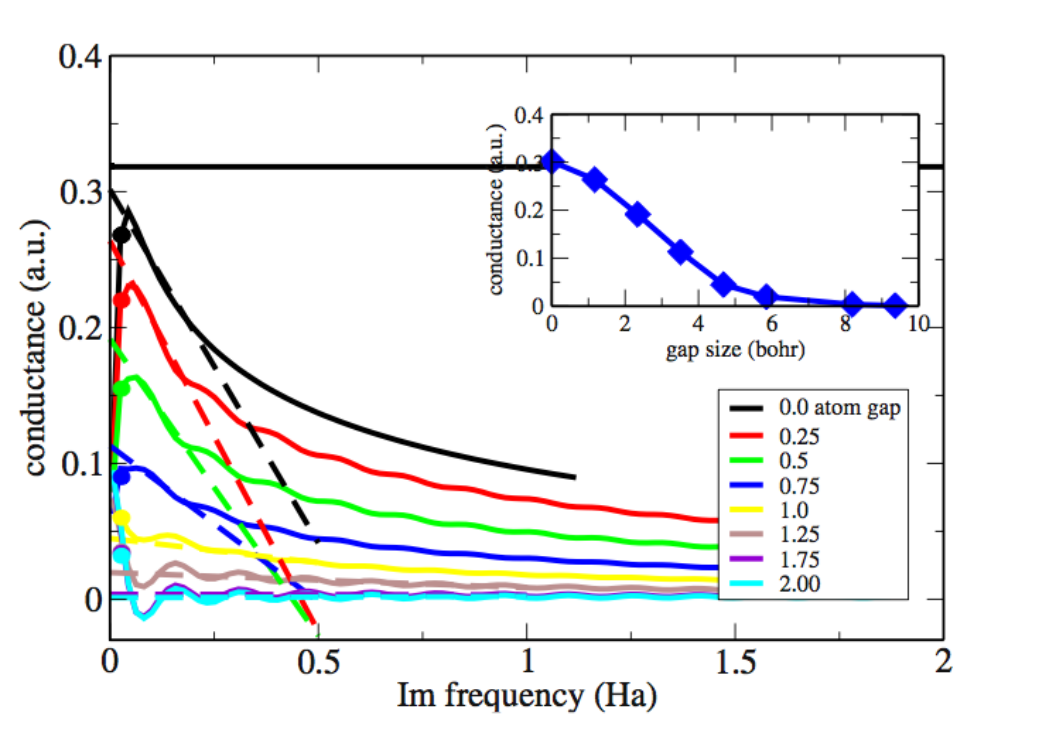
Na monowires with a gap

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



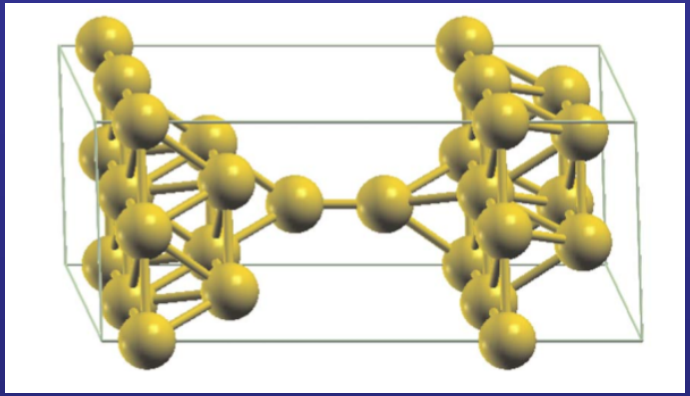
Na monowires with a gap (2)

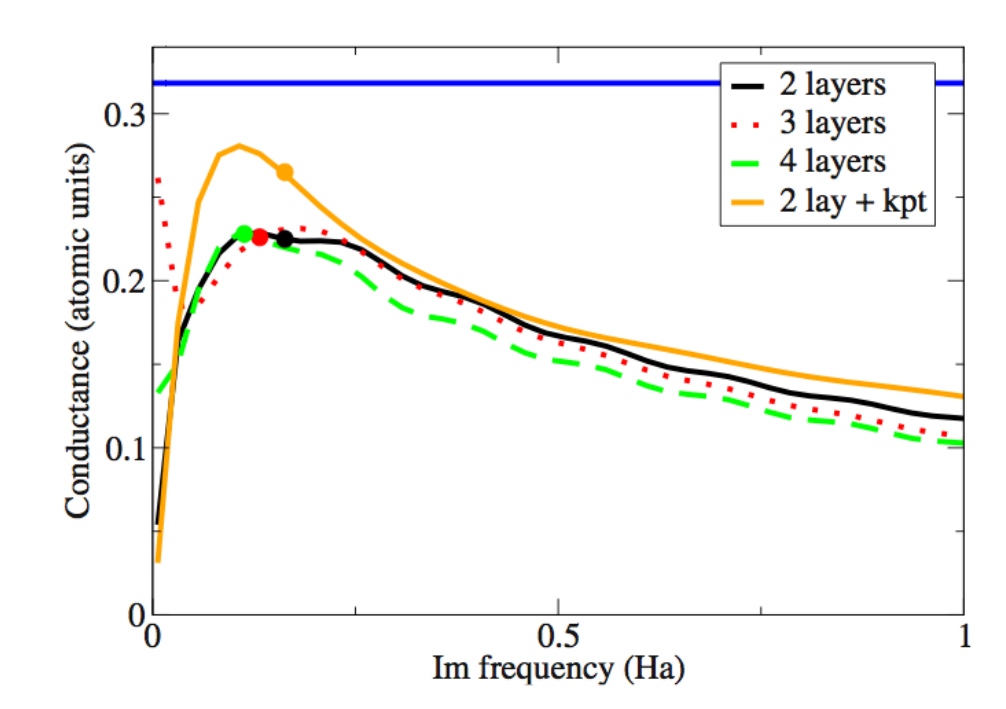
- As a function of gap size (8-atom leads)
- Good extrapolation



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





Beyond RPA: Many-Body Effects

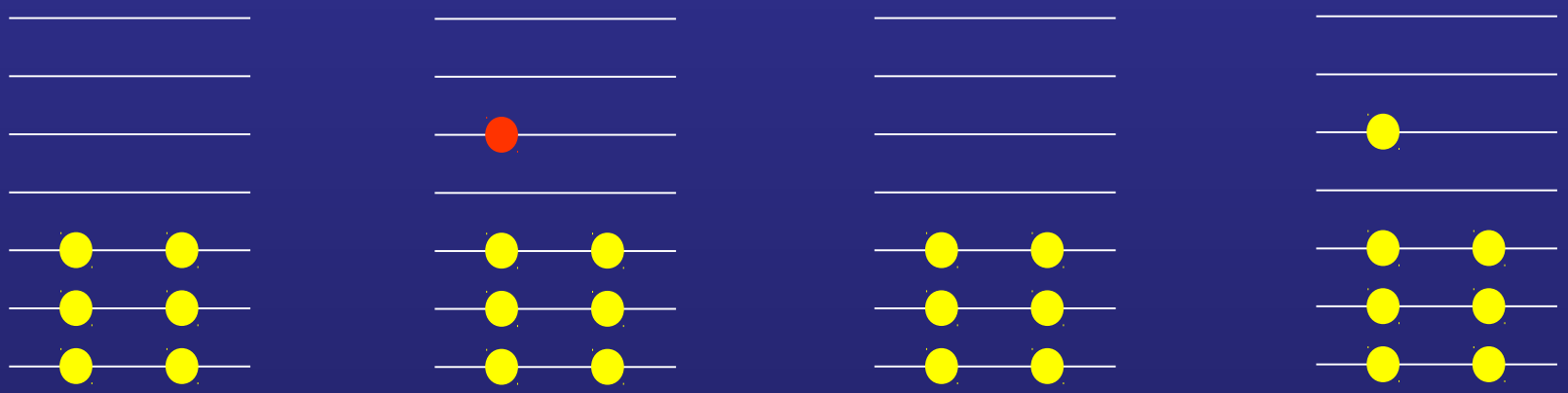
Calculating χ including interactions

- 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

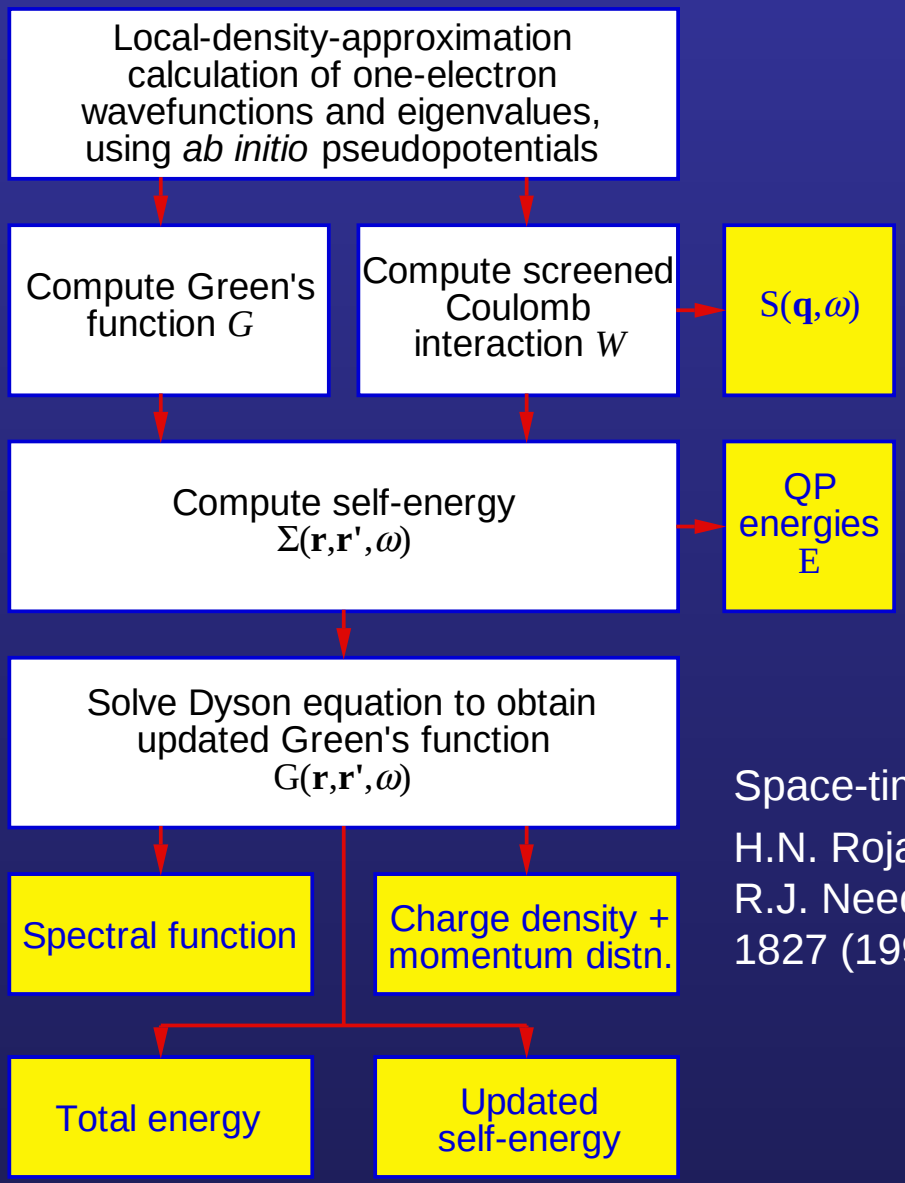
$$
\begin{aligned}
\Sigma(1,2) &= \mathrm{i}\int W\left(1^{+},3\right) G(1,4)\,\Gamma(4,2,3)\,\mathrm{d}(3,4) \\
P(1,2) &= -\mathrm{i}\int G(2,3)\,G(4,2)\,\Gamma(3,4,1)\,\mathrm{d}(3,4) \\
W(1,2) &= v(1,2)+\int W(1,3)\,P(3,4)\,v(4,2)\,\mathrm{d}(3,4) \\
\Gamma(1,2,3) &= \delta(1,2)\,\delta(1,3) \\
&+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

- 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

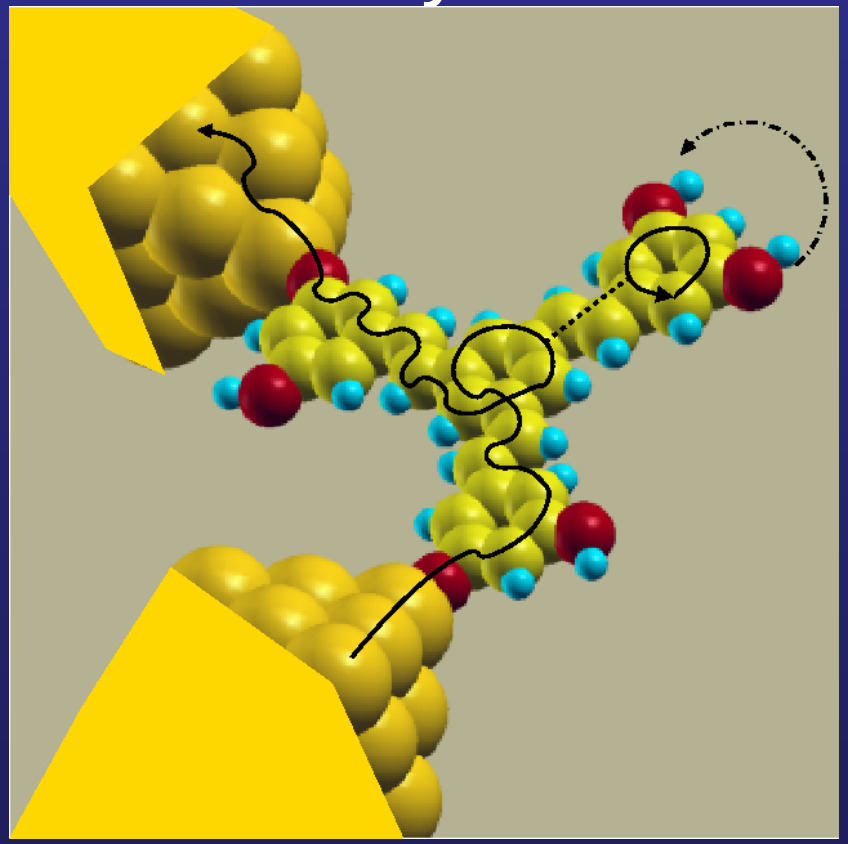
- Corrects band-gaps and band alignment
- Introduces finite lifetime
- *GW* and quantum transport:
  - Thygesen JCP (2007), Darancet PRB (2007), Neaton PRL (2007), ...
- Convenient implementation: real-space / imaginary-time:
  - Rojas, RWG, Needs PRL (1995)
- Finite temperatures (metals): Verstraete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

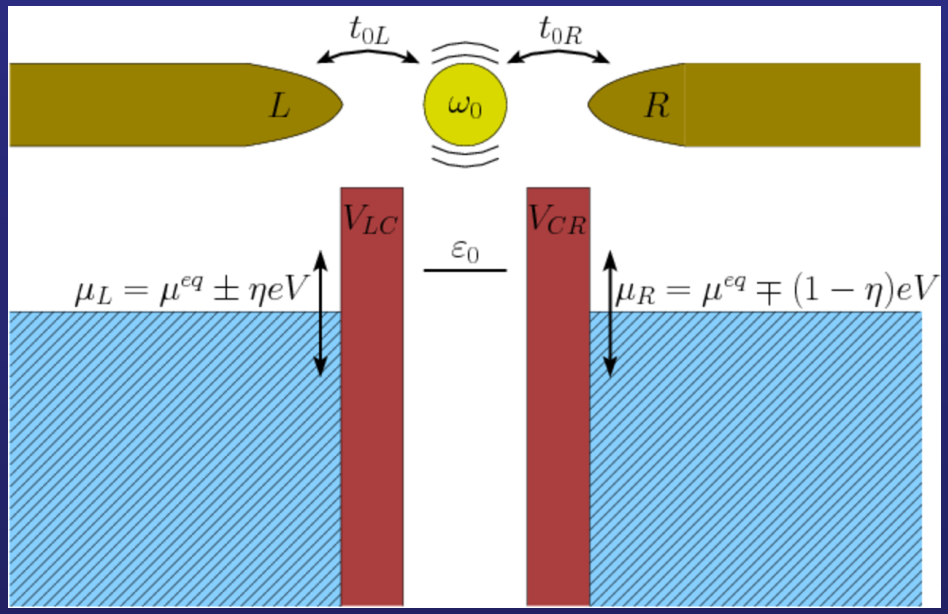
2. Beyond Conductance: I(V), Spectral Functions

L.K. Dash, H. Ness and RWG,
J. Chem. Phys. 2010;
H. Ness, L.K. Dash and RWG,
PRB (in press)/arXiv 2010

Single-site single-mode (SSSM) model

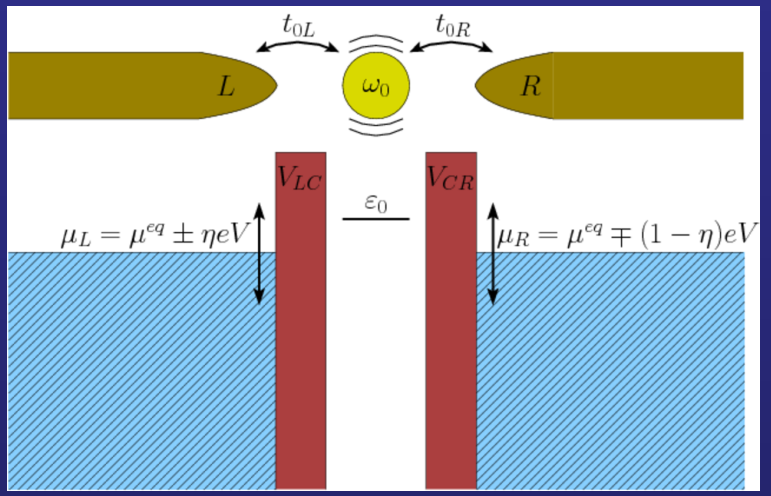
- SSSM system used to illustrate results





$$H_L + H_R = \sum_{\alpha=L,R} \varepsilon_\alpha c_\alpha^\dagger c_\alpha,$$

$$H_C^e = \sum_n \varepsilon_n d_n^\dagger d_n.$$



$$H_{\text{vib}} = \sum_\lambda \hbar\omega_\lambda a_\lambda^\dagger a_\lambda,$$

$$H_{e\text{-vib}} = \sum_{\lambda,n,m} \gamma_{\lambda nm}(a_\lambda^\dagger + a_\lambda) d_n^\dagger d_m,$$

MBPT calculations for SSSM model

- Non-equilibrium Green's function (NEGF) formalism, steady current
- E-ph interaction included perturbatively
- $$G^{>,<} = (1 + G^r\Sigma^r)g_C^{>,<}(1 + \Sigma^a G^a) + G^r\Sigma^{>,<}G^a.$$

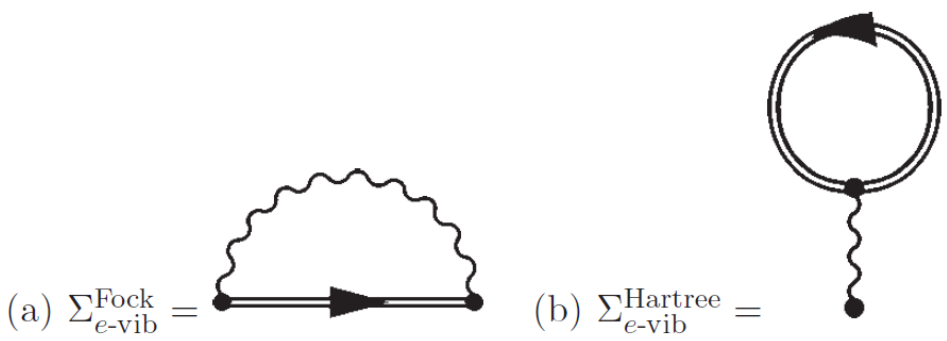


FIG. 2. The (a) Fock and (b) Hartree diagrams.

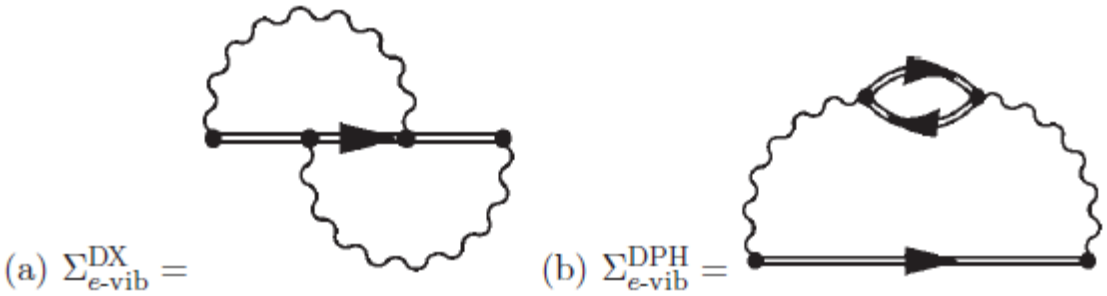


FIG. 3. The (a) double exchange DX and (b) vibron propagator dressed by the e-h bubble diagrams (dressed phonon, DPH).

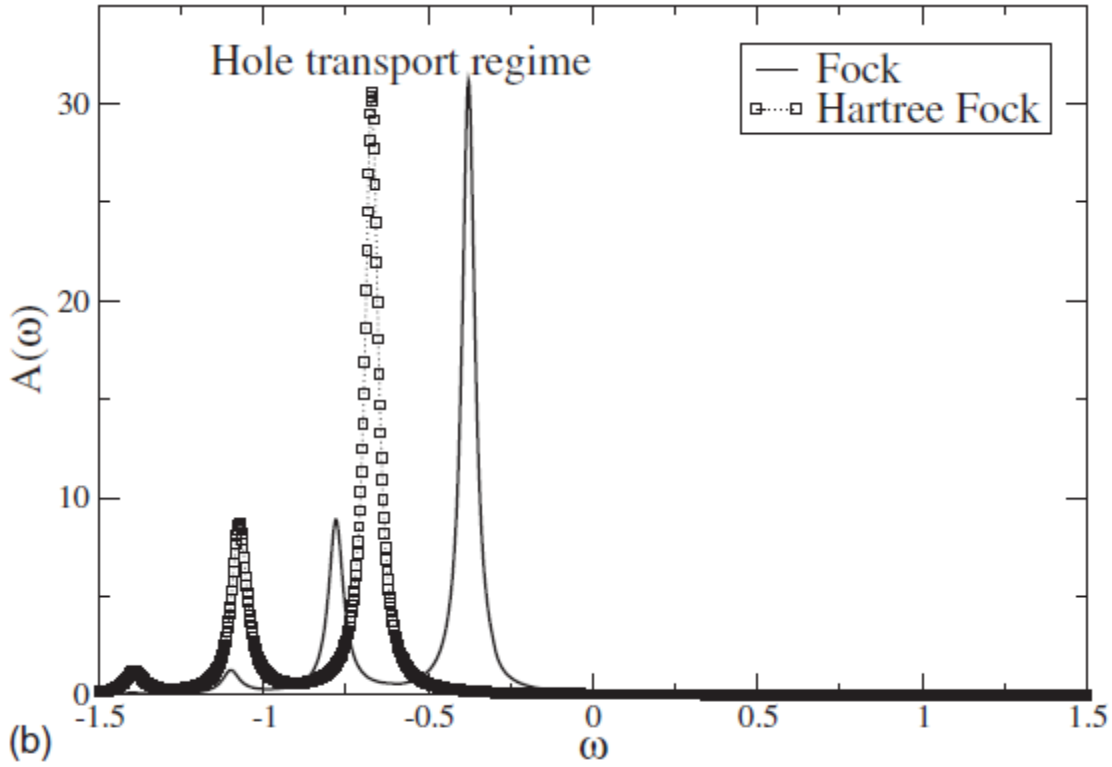


FIG. 4. Equilibrium (zero bias) spectral functions $A(\omega)$ for the off-resonant (a) electron and (b) hole transport regime. Calculations were performed with the Fock-like electron-vibron diagram (solid line) and with both the Hartree and Fock-like diagrams (symbols and dotted line). For the electron transport regime the inclusion of the Hartree self-energy has no effect, but for the hole transport regime it shifts the entire spectral function to lower energies. The parameters are $\varepsilon_0=+0.5(-0.5)$ for electron (hole) transport $\gamma_0=0.21$, $\omega_0=0.3$, $t_{0L,R}=0.15$, $\eta=0.005$, and $\mu_L=\mu_R=\mu^{eq}=0$.

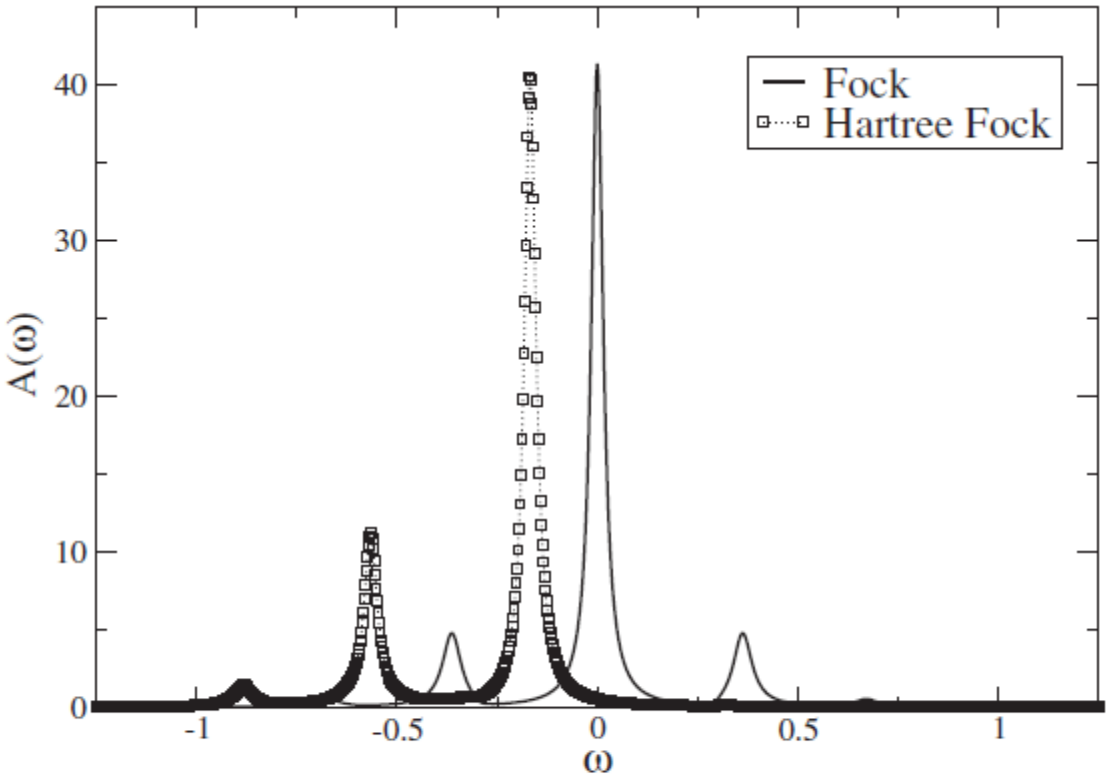


FIG. 5. Equilibrium spectral function $A(\omega)$ for the resonant transport regime. Calculations were performed with the Fock-like (solid line) and with both the Hartree and Fock-like diagrams (dashed line). The parameters are identical to those used in Figure 4, except for the value of the electron level which is ε_0=0 for the resonant regime.

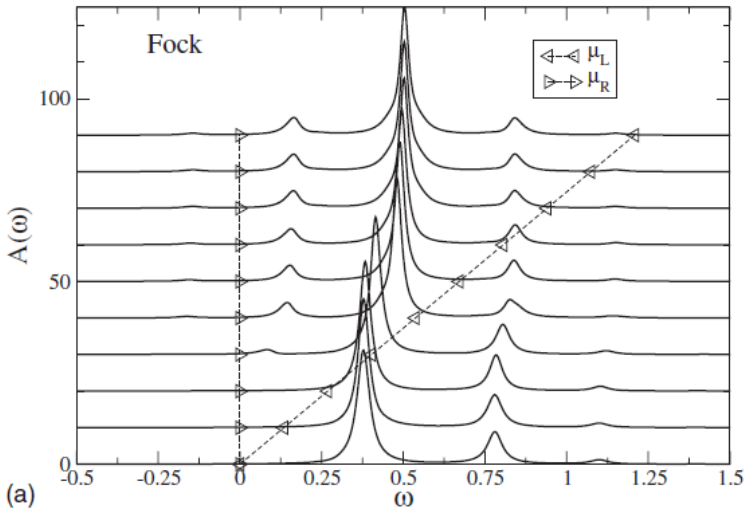


FIG. 6. Nonequilibrium spectral function for the off-resonant transport regime with applied bias for (a) the Fock diagram only and (b) both Fock and Hartree diagrams. The curves are offset vertically for clarity. The applied bias is given by the chemical potentials of the left (left-pointing arrows) and right (right-pointing arrows) leads, respectively. The Hartree potential has a strong effect on the peak positions. The other parameters are ε_0=+0.5, γ_0 =0.21, ω_0=0.3, $t_{0L,R}$=0.15, η=0.005, and η_V=1.

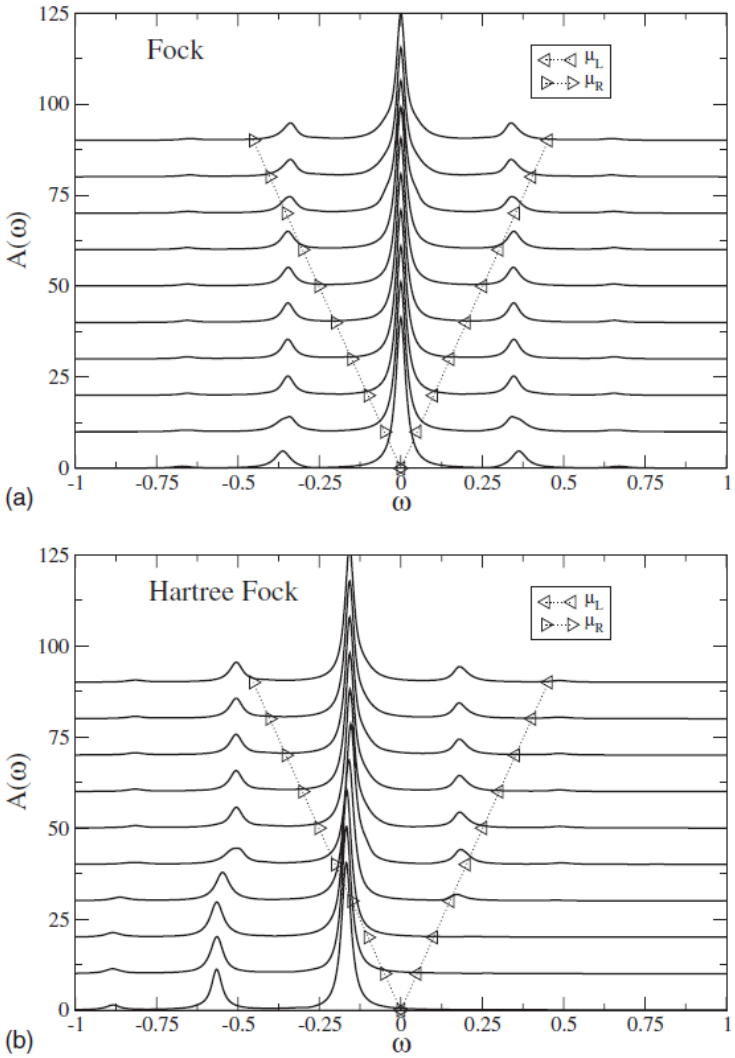


FIG. 8. Nonequilibrium spectral function $A(\omega)$ for the resonant transport regime for different applied biases for (a) the Fock diagram only, and (b) the Fock and Hartree diagrams. The curves are shifted vertically (+10 in the y-axis) for clarity, with the values of the left and right chemical potentials given by the triangular symbols, here we have a symmetric potential drop. Adding the Hartree diagram breaks the electron-hole symmetry. The other parameters are the same as for Fig. 6 except for ε_0=+0 and η_V=0.5.

Landauer vs Exact

- Can Landauer formula be generalised to interacting case?
- Ness, Dash and RWG, PRB in press / arXiv
- See also Meir and Wingreen PRL 1992

$$I = \frac{2e}{\hbar}\int \frac{\mathrm{d}\omega}{2\pi}\Big((f_L - f_R)\mathrm{Tr}\left[\Gamma_L G^r \Gamma_R G^a\right] + \mathrm{Tr}\Big[\Big((f_L - f_{\mathrm{int}}^{\mathrm{NE}})\Gamma_L - (f_R - f_{\mathrm{int}}^{\mathrm{NE}})\Gamma_R\Big) \times G^r \frac{i(\Sigma_{\mathrm{int}}^{>} - \Sigma_{\mathrm{int}}^{<})}{2} G^a\Big]\Big) . \quad (5)$$

The first term in Eq. (4) and (5) looks like a Landauer-like (LL) expression for the current,

$$I^{\mathrm{LL}} = \frac{2e}{h}\int \mathrm{d}\epsilon (f_L(\epsilon) - f_R(\epsilon)) T_{\mathrm{eff}}(\epsilon) = \frac{2e}{\hbar}\int \frac{\mathrm{d}\omega}{2\pi}(f_L - f_R)\ \mathrm{Tr}\left[\Gamma_L G^r \Gamma_R G^a\right], \quad (6)$$

with an effective transmission

$$T_{\mathrm{eff}}(\epsilon) = \mathrm{Tr}\left[\Gamma_L G^r \Gamma_R G^a\right](\epsilon) = \mathrm{Tr}[t^\dagger(\epsilon) t(\epsilon)], \quad (7)$$

Landauer vs Exact

- In fact the *full* current can be written in form similar to Landauer formula, with renormalisation of contacts:

$$I_L = \frac{2e}{\hbar}\int \frac{\mathrm{d}\omega}{2\pi}(f_L(\omega) - f_R(\omega))\mathrm{Tr}\left[\Gamma_L G^r \Upsilon_R G^a\right] \quad (8)$$

with the coupling to the right contact Υ_R being renormalized as

$$\Upsilon_R(\omega) = \Gamma_R(\omega)\Lambda(\omega), \quad (9)$$

and

$$\Lambda(\omega) = 1 + \Gamma_R^{-1}\frac{f_L(\omega) - f_{\mathrm{int}}^{\mathrm{NE}}(\omega)}{f_L(\omega) - f_R(\omega)}\, i(\Sigma_{\mathrm{int}}^{>} - \Sigma_{\mathrm{int}}^{<})(\omega), \quad (10)$$

- Ness, Dash and RWG, PRB in press / arXiv

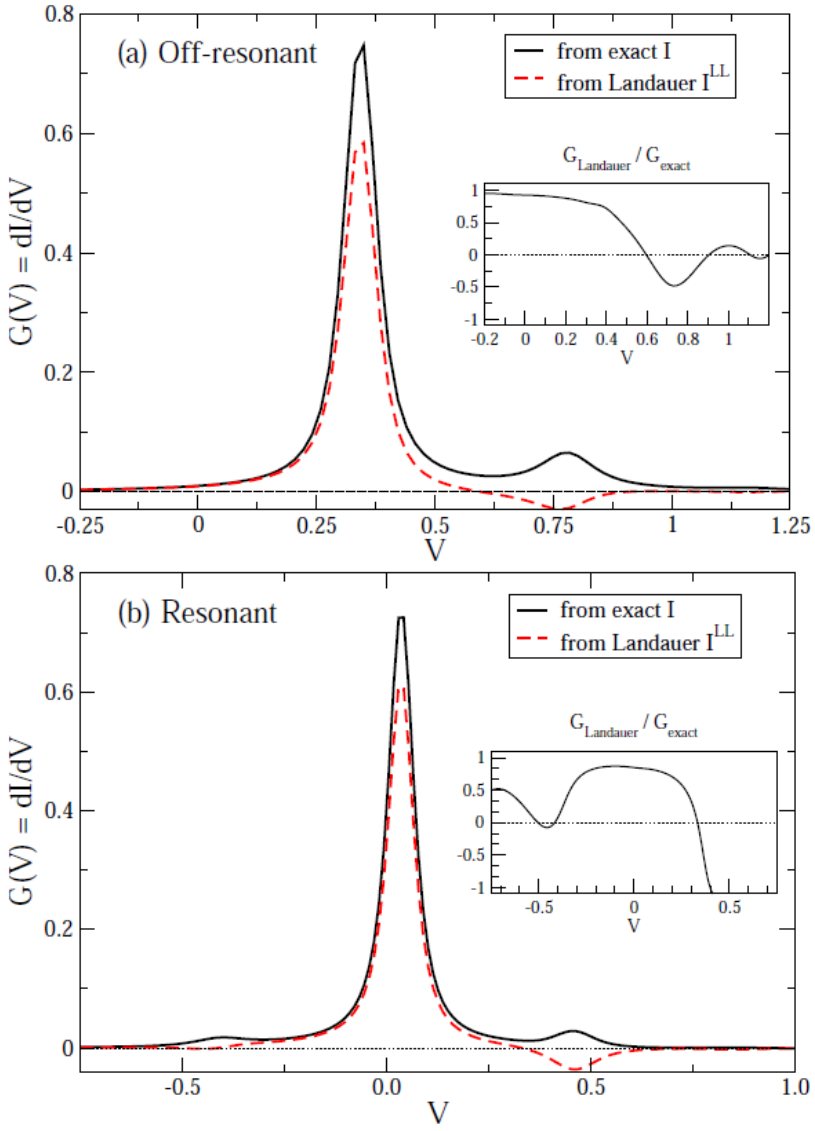


FIG. 2: Dynamical conductance dI/dV from the exact expression for the current $I(V)$ and the corresponding Landauer-like current $I^{\mathrm{LL}}(V)$. Green's functions calculations are performed

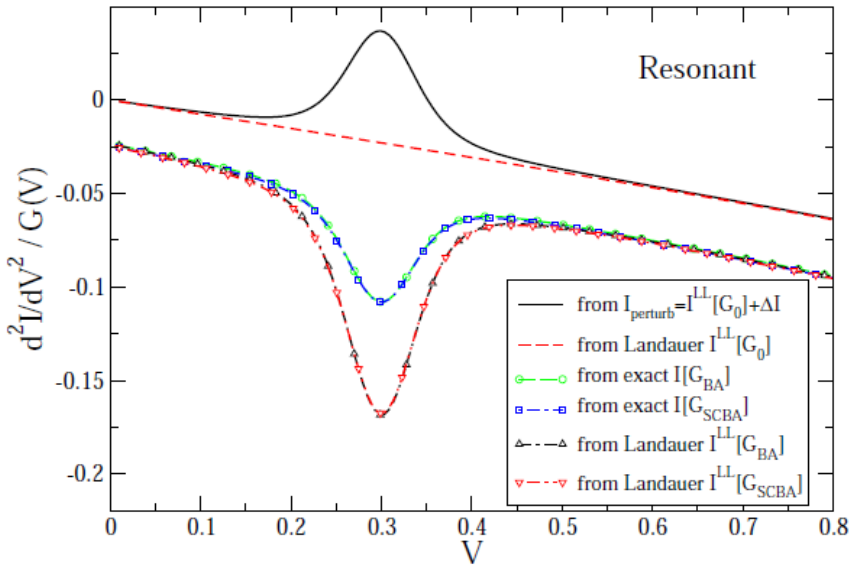


FIG. 4: IETS signal d^2I/dV^2, normalized by $G(V)$, obtained from the exact current $I(V)$ and from the corresponding Landauer-like $I^{\mathrm{LL}}(V)$ term. Calculations are for the resonant transport regime, intermediate electron-vibron coupling $\gamma_0/\omega_0 = 0.65$ and strong coupling to the leads. The different approximations used to calculate the Green's functions are shown in the legend (see main text for detail). The inelastic vibron excitation is present in the IETS signal derived from the exact $I(V)$ and is located around the vibron energy ω_0. It corresponds to a negative contribution to the baseline, as expected for mostly transparent junctions (metallic-like behavior). Interestingly, this feature is also present in the IETS derived from $I_{\mathrm{LL}}(V)$ in contrast to what is obtained for the off-resonant case. Hence, for resonant transport, it seems that Landauer-like approach can reproduce the inelastic IETS features at $V = \omega_0$. The parameters for the calculations are $\varepsilon_0 = 0, \omega_0 = 0.3, \gamma_0 = 0.195, t_{0L,R} = 1.50, T_{L,R} = 0.011, \eta = 0.025, \eta_V = 1$.

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ETSF

European Theoretical Spectroscopy Facility

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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
- Beyond conductance: *I*(*V*), spectral functions JCP 2010 PRB(in press)/arXiv 2010
  - Effect of interactions beyond Landauer and Landauer-like formulae

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

Challenges for QMC

- Need benchmark calculations for non-equilibrium systems
- Conductance: linear response at $i\omega \to 0$ suffices
- Better: steady current vs. bias $I(V)$
- Better: time-dependent $\boldsymbol{j}(t)$ given some applied voltage $V(t)$