TIME-DEPENDENT QUANTUM TRANSPORT IN NANOSYSTEMS

A nonequilibrium Green's function approach

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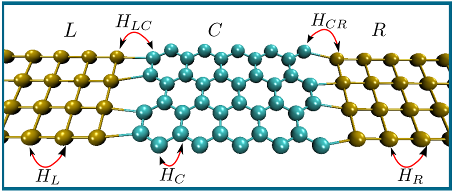

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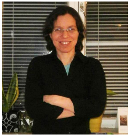

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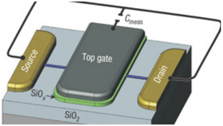
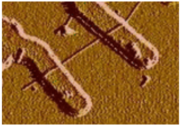
OUTLINE

INTRODUCTION TO QUANTUM TRANSPORT



1



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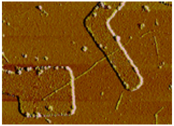

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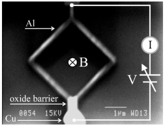

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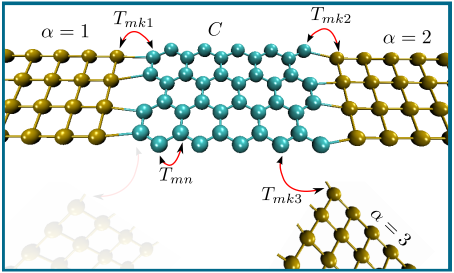
- Possibility to manufacture and control nanoscale junctions
- Ultrafast transient responses experimentally reachable
- Nanoscale development of electronic devices
  - Transistor count, switching times, sensors, etc.
- Need theory for describing the full time dependence
- Transient spectroscopy: “seeing” how the systems operate

[1] S. Ilani, et. al: Nature Physics **2**, 687 (2006)
[2] M. Ahlskog, et. al: Phys. Rev. B **79**, 155408 (2009)
[3] D. Talukdar, et. al: Phys. Rev. B **88**, 125407 (2013)
[4] K. Arutyunov, et. al: Phys. Rev. B **70**, 064514 (2004)

SYSTEM SETUP AND THE MODEL HAMILTONIAN



$$
\begin{aligned}
\hat{H} &= \sum_{mn,\sigma} T_{mn} \hat{d}^{\dagger}_{m,\sigma} \hat{d}_{n,\sigma} + \sum_{k\alpha,\sigma} \epsilon_{k\alpha} \hat{d}^{\dagger}_{k\alpha,\sigma} \hat{d}_{k\alpha,\sigma} \\
&+ \sum_{mk\alpha,\sigma} \left(T_{mk\alpha} \hat{d}^{\dagger}_{m,\sigma} \hat{d}_{k\alpha,\sigma} + T_{k\alpha m} \hat{d}^{\dagger}_{k\alpha,\sigma} \hat{d}_{m,\sigma} \right) \\
&+ \text{interactions}
\end{aligned}
$$

Partition-free approach: Initially coupled systems in equilibrium (unique μ and β)

GREEN'S FUNCTIONS AND THE EQUATIONS OF MOTION

- One-particle Green's function (propagator): Ensemble average of the contour-ordered product of particle creation and annihilation operators (Heisenberg picture)

$$G_{mn}(t,t') = -\mathrm{i}\langle \mathcal{T}_\gamma[\hat{d}_{m,\mathrm{H}}(t)\hat{d}^\dagger_{n,\mathrm{H}}(t')]\rangle$$

- Equations of motion for the Green's function

$$\left[\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t} - H\right] G(t,t') = \delta(t,t') + \int_\gamma \mathrm{d}\bar{t}\,\Sigma(t,\bar{t})G(\bar{t},t')$$

- Embedding (retarded) self-energy: purely imaginary constant $\sim$ level-width matrix Γ (in wide-band limit)

$$\Sigma^{\mathrm{R}}_{\alpha,mn}(\omega) = \sum_k T_{mk\alpha}\frac{1}{\omega - \epsilon_{k\alpha} - V_\alpha + \mathrm{i}\eta} T_{k\alpha n} \approx -\frac{\mathrm{i}}{2}\Gamma_{\alpha,mn}$$

SOLUTION TO THE EQUATIONS OF MOTION

- Explicit expression for the time-dependent density matrix[1]

Steady-state value

Molecule–lead transitions

$$
\begin{aligned}
-\mathrm{i}G^{<}_{mn}(t,t) = \rho_{mn}(t) &= \sum_{\alpha} \Gamma_{\alpha,mn} \Lambda_{\alpha,mn} \\
&+ \sum_{\alpha} V_{\alpha} \Gamma_{\alpha,mn} \left[\Pi_{\alpha,mn}(t) + \Pi^{*}_{\alpha,nm}(t) \right] \\
&+ \sum_{\alpha} V_{\alpha}^{2} \Gamma_{\alpha,mn} \mathrm{e}^{-\mathrm{i}(\epsilon_m - \epsilon_n^{*})t} \Omega_{\alpha,mn}
\end{aligned}
$$

Intramolecular transitions

- Densities and currents
  - Diagonal elements $\sim$ site-localized charge densities
  - Off-diagonal elements $\sim$ local bond currents
- Extensions to local perturbations (EM fields)[2], superconductivity (NSN junctions) and arbitrary temperatures[3]

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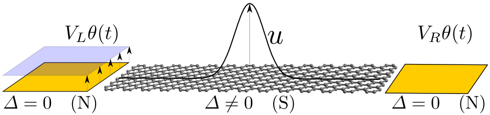
[1] **RT**, R. van Leeuwen, E. Perfetto, and G. Stefanucci, J. Phys.: Conf. Ser. **427**, 012014 (2013)

[2] **RT**, E. Perfetto, G. Stefanucci, and R. van Leeuwen, Phys. Rev. B **89**, 085131 (2014)

[3] **RT**, R. van Leeuwen, E. Perfetto, and G. Stefanucci, J. Phys.: Conf. Ser. **696**, 012016 (2016)

TIME-DEPENDENT LANDAUER–BÜTTIKER FORMULA [1]



$$\begin{aligned}
I_\alpha(t) &= 2\int \frac{\mathrm{d}\omega}{2\pi} f(\omega-\mu)\sum_\beta \mathrm{Tr}\Big\{ \\
&\varGamma_\alpha G^{\mathrm{R}}(\omega+V_\beta)\varGamma_\beta G^{\mathrm{A}}(\omega+V_\beta) - \varGamma_\alpha G^{\mathrm{R}}(\omega+V_\alpha)\varGamma_\beta G^{\mathrm{A}}(\omega+V_\alpha) \\
&+ V_\beta\left[\varGamma_\alpha \mathrm{e}^{\mathrm{i}(\omega+V_\beta-h_{\mathrm{eff}})t} G^{\mathrm{R}}(\omega)\left(-\mathrm{i}\delta_{\alpha\beta}G^{\mathrm{R}}(\omega+V_\beta)+A_\beta(\omega+V_\beta)\right)+\mathrm{h.c.}\right] \\
&+ V_\beta^2 \varGamma_\alpha \mathrm{e}^{-\mathrm{i}h_{\mathrm{eff}}t} G^{\mathrm{R}}(\omega)A_\beta(\omega+V_\beta)G^{\mathrm{A}}(\omega)\mathrm{e}^{\mathrm{i}h^\dagger_{\mathrm{eff}}t}\Big\}, \\
A_\alpha(\omega) &= G^{\mathrm{R}}(\omega)\varGamma_\alpha G^{\mathrm{A}}(\omega)
\end{aligned}$$

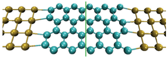
- Time-dependent bias voltages and local perturbations[2]
- Temperature bias, heat currents and local temperatures[3]

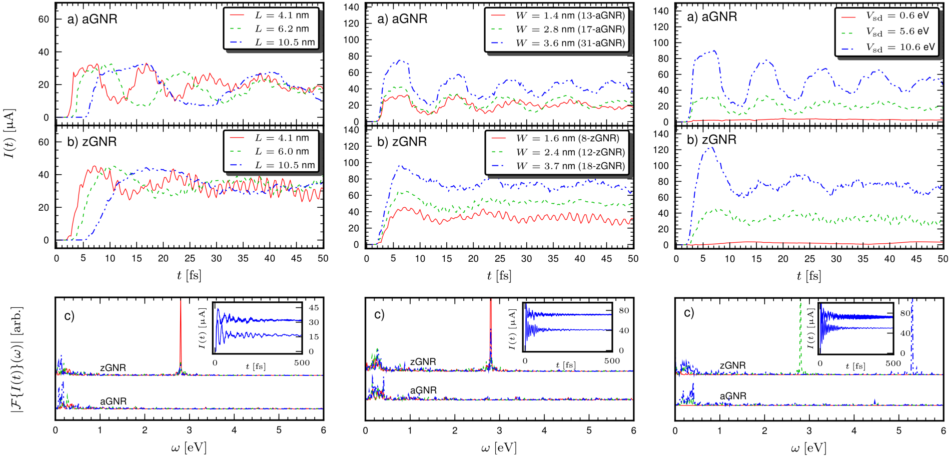
[1] **RT**, R. van Leeuwen, E. Perfetto, and G. Stefanucci, J. Phys.: Conf. Ser. **427**, 012014 (2013)

[2] M. Ridley, A. MacKinnon, and L. Kantorovich, Phys. Rev. B **91**, 125433 (2015)

[3] F. G. Eich, M. Di Ventra, and G. Vignale, Phys. Rev. B **93**, 134309 (2016)

GEOMETRY AFFECTS THE TRANSIENTS[1]

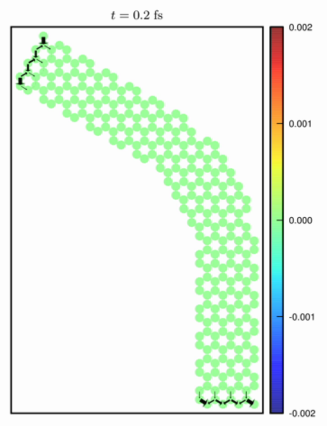


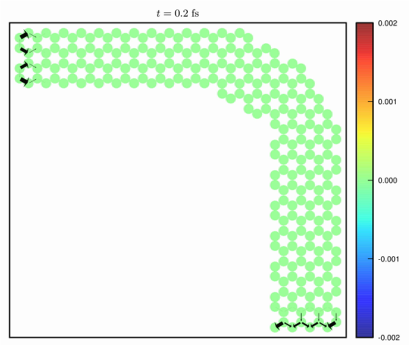


- ▶ Edge-state transition only visible in zigzag ribbons

[1] **RT**, E. Perfetto, G. Stefanucci, and R. van Leeuwen, Phys. Rev. B **89**, 085131 (2014)

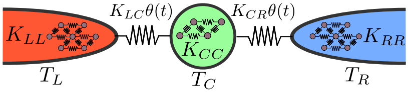
CURVATURE TRIGGERS SPATIALLY AND TEMPORALLY FOCUSED CURRENTS[1]





[1] C. G. Rocha, **RT**, R. van Leeuwen, and P. Koskinen, Nanoscale **7**, 8627 (2015)

TIME-DEPENDENT LANDAUER–BÜTTIKER FORMALISM FOR PHONONS[1]



- Harmonic lattice: $\hat{H} = \sum_j \frac{\hat{p}_j^2}{2} + \sum_{jk} \frac{1}{2}\hat{u}_j K_{jk} \hat{u}_k = \frac{1}{2}\sum_{jk} \hat{\boldsymbol{\phi}}_j \boldsymbol{\Omega}_{jk} \hat{\boldsymbol{\phi}}_k$
- Spinor representation: $\hat{\boldsymbol{\phi}}_j = \begin{pmatrix} \hat{u}_j \\ \hat{p}_j \end{pmatrix}$; $\boldsymbol{\Omega}_{jk} = \begin{pmatrix} K_{jk} & 0 \\ 0 & \delta_{jk} \end{pmatrix}$
- Commutation relations: $\left[\hat{\boldsymbol{\phi}}_j, \hat{\boldsymbol{\phi}}_k\right] = \delta_{jk}\boldsymbol{\alpha}$; $\boldsymbol{\alpha} = \begin{pmatrix} 0 & \mathrm{i} \\ -\mathrm{i} & 0 \end{pmatrix}$
- Phonon Green's function: $\boldsymbol{D}_{jk}(z,z') = -\mathrm{i}\langle \mathcal{T}_\gamma[\hat{\boldsymbol{\phi}}_j(z)\hat{\boldsymbol{\phi}}_k(z')]\rangle$
- Due to the spinor representation the equations of motion will be 1st order in time! (Similar to the electronic case)
- Partitioned approach (subsystems initially uncoupled)

[1] **RT**, N. Säkkinen, D. Karlsson, G. Stefanucci, and R. van Leeuwen, Phys. Rev. B **93**, 214301 (2016)

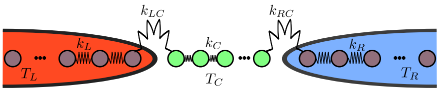
TIME-DEPENDENT LANDAUER–BÜTTIKER FORMALISM FOR PHONONS[1]

Explicit expression for the time-dependent density matrix

$$
\begin{aligned}
\mathrm{i}\boldsymbol{D}^{<}(t,t) &= \mathrm{e}^{-\mathrm{i}\boldsymbol{\Omega}_{\mathrm{eff}}t}\boldsymbol{\alpha} f_C(\boldsymbol{\Omega}\boldsymbol{\alpha})\mathrm{e}^{\mathrm{i}\boldsymbol{\Omega}^{\dagger}_{\mathrm{eff}}t} \\
&+ \sum_{\lambda}\int_{-\omega_c}^{\omega_c}\frac{\mathrm{d}\omega}{2\pi}f_{\lambda}(\omega)\left[\mathbf{1}-\mathrm{e}^{\mathrm{i}(\omega-\boldsymbol{\Omega}_{\mathrm{eff}})t}\right]\boldsymbol{B}_{\lambda}(\omega)\left[\mathbf{1}-\mathrm{e}^{-\mathrm{i}(\omega-\boldsymbol{\Omega}^{\dagger}_{\mathrm{eff}})t}\right] \\
\boldsymbol{\Omega}_{\mathrm{eff}} &= \frac{1}{\boldsymbol{\alpha}+\frac{\mathrm{i}}{2}\boldsymbol{\Gamma}'_0}(\boldsymbol{\Omega}+\boldsymbol{\Lambda}_0) \qquad \text{(non-hermitian)} \\
\boldsymbol{B}_{\lambda}(\omega) &= \frac{1}{\omega(\boldsymbol{\alpha}+\frac{\mathrm{i}}{2}\boldsymbol{\Gamma}'_0)-\boldsymbol{\Omega}-\boldsymbol{\Lambda}_0}\,\omega\boldsymbol{\Gamma}'_{0,\lambda}\,\frac{1}{\omega(\boldsymbol{\alpha}-\frac{\mathrm{i}}{2}\boldsymbol{\Gamma}'_0)-\boldsymbol{\Omega}-\boldsymbol{\Lambda}_0}
\end{aligned}
$$

[1] **RT**, N. Säkkinen, D. Karlsson, G. Stefanucci, and R. van Leeuwen, Phys. Rev. B **93**, 214301 (2016)

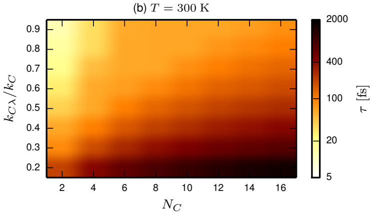
TIME-DEPENDENT LANDAUER–BÜTTIKER FORMALISM FOR PHONONS[1]



- 1D harmonic chains of atoms
- Time-dependent heat currents
- $T_C = (T_L + T_R)/2$

Thermalization time τ

$$\kappa = \frac{J^Q(t=\tau) - J^Q_{SS}}{J^Q_{SS}} = 10\%$$



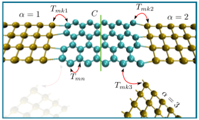
Further (future) applications could involve studying local heat currents and temperatures in, e.g., graphene-based phononic junctions

[1] **RT**, N. Säkkinen, D. Karlsson, G. Stefanucci, and R. van Leeuwen, Phys. Rev. B **93**, 214301 (2016)

SUMMARY AND OUTLOOK

Time-dependent quantum transport in nanosystems

Noninteracting particles within WBA $\Rightarrow$ Analytic solution



$$\hat{H} = \sum_{mn,\sigma} T_{mn}\hat{d}^\dagger_{m,\sigma}\hat{d}_{n,\sigma} + \sum_{k\alpha,\sigma}\epsilon_{k\alpha}\hat{d}^\dagger_{k\alpha,\sigma}\hat{d}_{k\alpha,\sigma} + \sum_{mk\alpha,\sigma}\left(T_{mk\alpha}\hat{d}^\dagger_{m,\sigma}\hat{d}_{k\alpha,\sigma} + T_{k\alpha m}\hat{d}^\dagger_{k\alpha,\sigma}\hat{d}_{m,\sigma}\right)$$

Explicit expression for the equal-time *lesser* Green's function $\sim$ time-dependent density matrix

Steady-state value

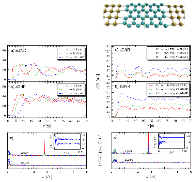
$$-\mathrm{i}G^<_{mn}(t,t) = \rho_{mn}(t) = \sum_\alpha \Gamma_{\alpha,m}\Lambda_{\alpha,mn} + \sum_\alpha V_\alpha \Gamma_{\alpha,mn}\left(\Pi_{\alpha,mn}(t) + \Pi^*_{\alpha,nm}\right. + \sum_\alpha V_\alpha^2 \Gamma_{\alpha,m} e^{-\mathrm{i}(\epsilon_m-\epsilon_n^*)t}\Omega_{\alpha,mn}$$

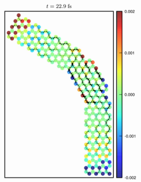
Molecule–lead t

Intramolecular transiti

Densities and currents

- Diagonal elements ρ_{mm} $\sim$ site-localized charge densitie
- Off-diagonal elements $\sim$ local bond currents





How to deal with interactions (e-e, e-ph)?

- Generalized Kadanoff–Baym Ansatz: $G^{\lessgtr}(t,t') = G^{\mathrm{R}}(t,t')\rho^{\lessgtr}(t') - \rho^{\lessgtr}(t)G^{\mathrm{A}}(t,t')$
- Diagonal propagation vs. (t,t')-plane $\Rightarrow$ computable to more realistic systems?

More on quantum transport and the nonequilibrium Green's functions:

R. Tuovinen (PhD thesis)
Time-dependent quantum transport in nanosystems: A nonequilibrium Green's function approach, University of Jyväskylä, 2016

G. Stefanucci and R. van Leeuwen
Nonequilibrium Many-Body Theory of Quantum systems: A Modern Introduction, Cambridge University Press, 2013