Thermal density-functional theory: Towards the ab-initio description of thermoelectric transport at the nano scale

F. G. Eich

Max Planck Institute
for the Structure and Dynamics of Matter

7th Time-dependent Density-Functional Theory:
Prospects and Applications
Benasque, Spain, September 22, 2016

References and Acknowledgements

Max Di Ventra

PRL 112, 196401 (2014) PHYSICAL REVIEW LETTERS week ending 16 MAY 2014

Density-Functional Theory of Thermoelectric Phenomena

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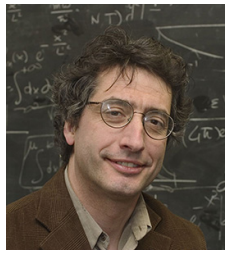

Giovanni Vignale

PHYSICAL REVIEW B 90, 115116 (2014)

Luttinger-field approach to thermoelectric transport in nanoscale conductors

F. G. Eich,[1,*] A. Principi,[1] M. Di Ventra,[2] and G. Vignale[1]
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Temperature-driven transient charge and heat currents in nanoscale conductors

F. G. Eich,[1,*] M. Di Ventra,[2] and G. Vignale[1]
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Recent review → arXiv:1607.05464

DFG Deutsche Forschungsgemeinschaft

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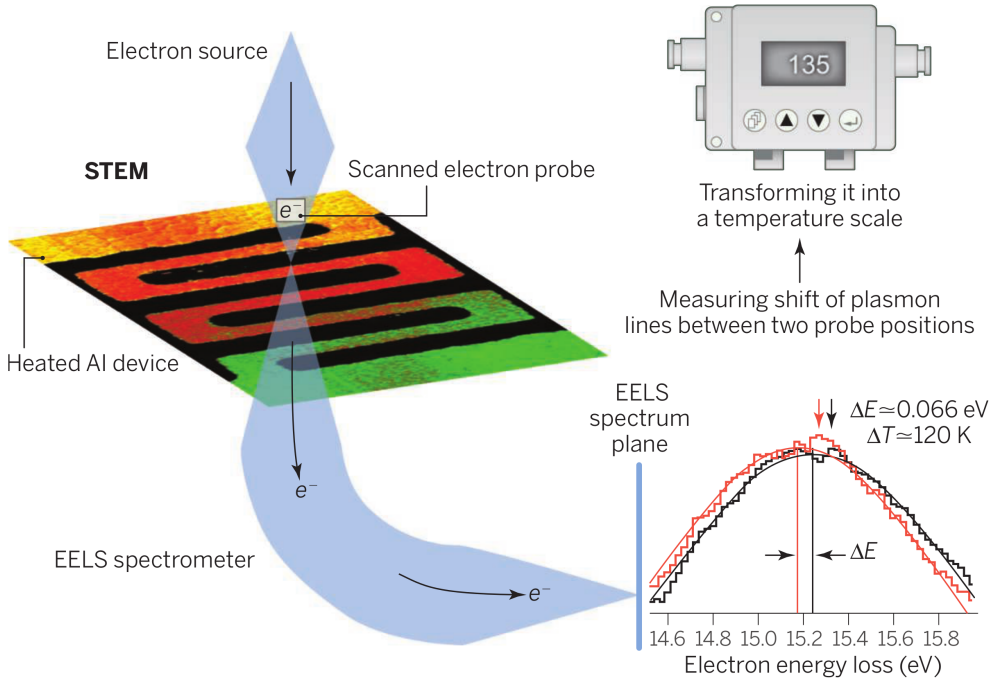
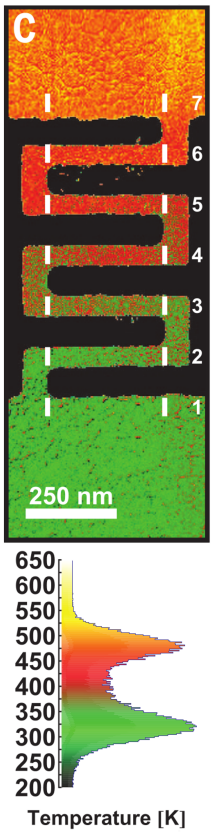
Measuring temperatures at the nanoscale

Nanoscale temperature mapping in operating microelectronic devices

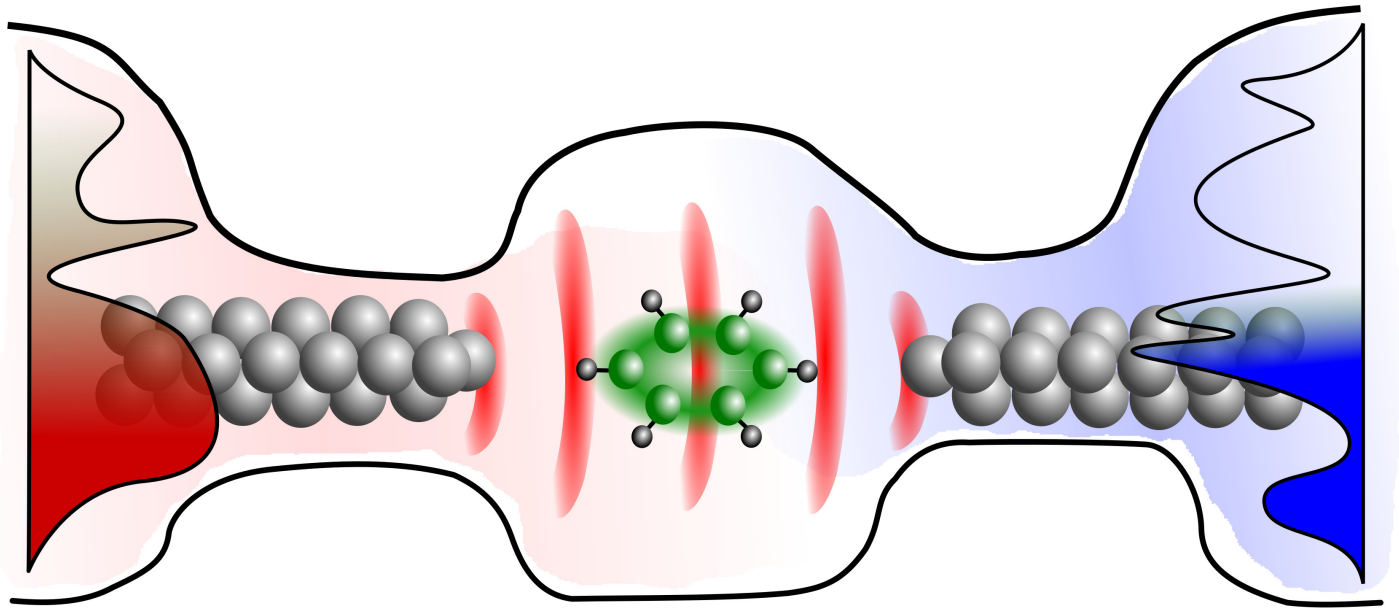
Matthew Mecklenburg,[1*] William A. Hubbard,[2,3] E. R. White,[2,3] Rohan Dhall,[4] Stephen B. Cronin,[4] Shaul Aloni,[5] B. C. Regan[2,3*]





Mecklenburg *et al.*, Science, **347** (6222): 629-632 (2015)

Typical nanoscale transport setup



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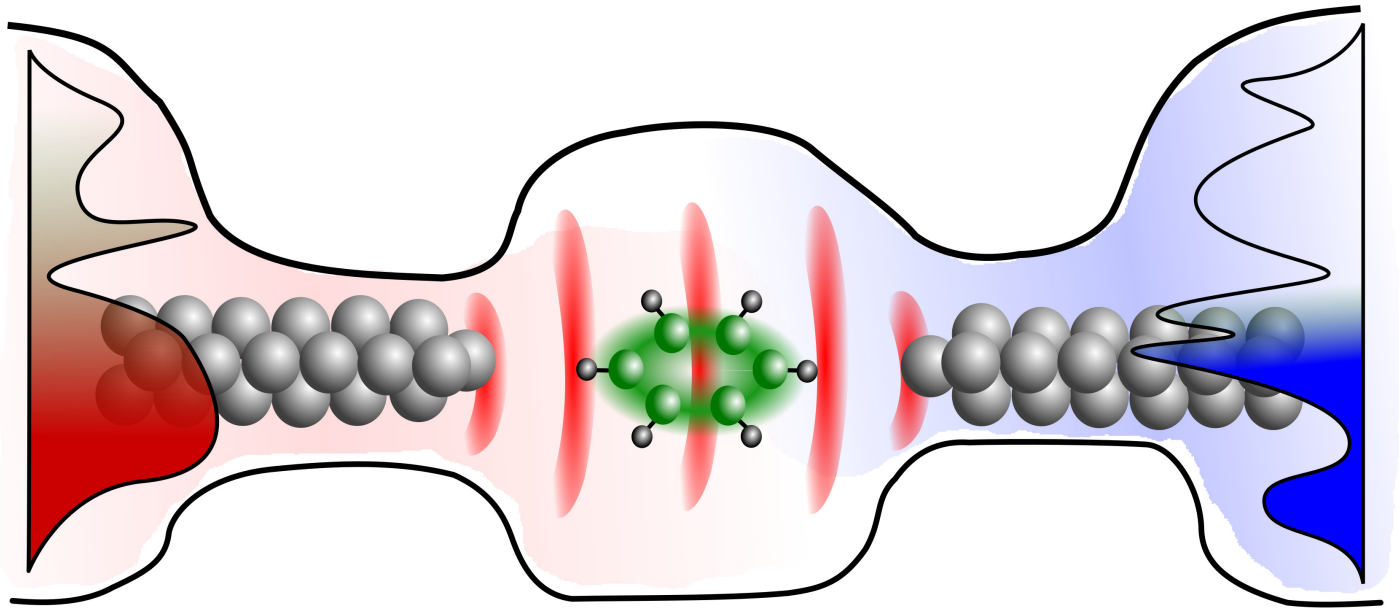

$$\begin{pmatrix} \mathcal{I} \\ \mathcal{J} \end{pmatrix} = -\begin{pmatrix} \boldsymbol{\sigma} & \boldsymbol{\sigma S T_0} \\ \boldsymbol{\sigma \Pi} & \boldsymbol{T_0 \kappa + \sigma \Pi^2} \end{pmatrix} \begin{pmatrix} \delta\mu \\ \frac{\delta T}{T_0} \end{pmatrix}$$

Thermal DFT and Luttinger's ψ-field

Including the effect of electron-electron interactions

(FGE, M. Di Ventra, G. Vignale, Phys. Rev. Lett. **112**, 196401 (2014))

- DFT efficient tool to incorporate interactions
- Standard formulation focuses on charge density
- Our proposal: Inclusion of the energy density as basic variable

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Thermal DFT and Luttinger's ψ-field

Luttinger's ψ - field

(J. M. Luttinger, Phys. Rev. **135**, A1505 (1964))

- Mechanical proxy for local temperature variations
  (B. S. Shastry, Rep. Prog. Phys. **72**, 016501 (2009))
- Low frequencies $\omega \ll 1/\tau_0$, long wave length $k \ll 1/l_0$
- Allows to compute conductivities via Kubo formalism

$$\begin{pmatrix} \mathcal{I} \\ \mathcal{J} \end{pmatrix} = -\begin{pmatrix} \boldsymbol{\sigma} & \boldsymbol{\sigma S T_0} \\ \boldsymbol{\sigma \Pi} & \boldsymbol{T_0 \kappa + \sigma \Pi^2} \end{pmatrix} \begin{pmatrix} \delta\mu + \delta v \\ \frac{\delta T}{T_0} + \delta\psi \end{pmatrix}$$

Thermal DFT and Luttinger's ψ-field

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$$\imath\hbar\partial_t\phi_i(\boldsymbol{r}) = \left[-\frac{\hbar^2}{2}\boldsymbol{\nabla}\cdot\frac{1+\boxed{\psi+\bar{\psi}_{\rm xc}}}{m}\boldsymbol{\nabla}+\boxed{(1+\psi)v+\tilde{v}_{\rm Hxc}+\bar{v}_{\rm xc}}\right]\phi_i(\boldsymbol{r})$$

Peculiarities of the KS construction

$$\bar{v}_{\rm xc}(\boldsymbol{r},t) = -\frac{\delta \bar{S}_{xc}[n,h_s]}{\delta n(\boldsymbol{r},t)}$$

$$\bar{\psi}_{\rm xc}(\boldsymbol{r},t) = -\frac{\delta \bar{S}_{xc}[n,h_s]}{\delta h_s(\boldsymbol{r},t)}$$

How to connect the interacting system and the noninteracting KS system ?

Peculiarities of the KS construction

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How to connect the interacting system and the noninteracting KS system ?

Interacting system		KS system
$n(\boldsymbol{r},t)$	$=$	$n(\boldsymbol{r},t)$
$h(\boldsymbol{r},t) - h^{\rm eq}[n(t)](\boldsymbol{r})$	$=$	$h_{\rm s}(\boldsymbol{r},t) - h_{\rm s}^{\rm eq}[n(t)](\boldsymbol{r})$

The KS system reproduces the excess energy density !!

$$h(\boldsymbol{r},t) = h_{\rm s}(\boldsymbol{r},t) + \mathcal{E}_{\rm Hxc}[n(t)](\boldsymbol{r}) \quad , \quad \mathcal{E}_{\rm Hxc}[n(t)](r) = h^{\rm eq}[n(t)](\boldsymbol{r}) - h_{\rm s}^{\rm eq}[n(t)](\boldsymbol{r})$$

Adiabatic local density approximation

$$\imath\hbar\partial_t\phi_i(\boldsymbol{r}) = \left[-\frac{\hbar^2}{2}\boldsymbol{\nabla}\cdot\frac{1+\psi+\bar{\psi}_{xc}^{\mathrm{ALDA}}}{m}\,\boldsymbol{\nabla} + (1+\psi)\,(v+v_{\mathrm{Hxc}}) + \bar{v}_{xc}^{\mathrm{ALDA}}\right]\phi_i(\boldsymbol{r})$$

$$\bar{v}_{\mathrm{xc}}^{\mathrm{ALDA}}(\boldsymbol{r},t) = -\tfrac{1}{\beta}\frac{\partial\bar{s}_{\mathrm{xc}}(n(\boldsymbol{r},t),h_{\mathrm{s}}(\boldsymbol{r},t))}{\partial n(\boldsymbol{r},t)}$$

$$\bar{\psi}_{\mathrm{xc}}^{\mathrm{ALDA}}(\boldsymbol{r},t) = -\tfrac{1}{\beta}\frac{\partial\bar{s}_{\mathrm{xc}}(n(\boldsymbol{r},t),h_{\mathrm{s}}(\boldsymbol{r},t))}{\partial h_{\mathrm{s}}(\boldsymbol{r},t)}$$

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$$\bar{s}_{\mathrm{xc}}(n(\boldsymbol{r},t),h_{\mathrm{s}}(\boldsymbol{r},t)) = s^{\mathrm{unif}}(n,h_{\mathrm{s}}+\epsilon_{\mathrm{Hxc}}(n)) - s_{\mathrm{s}}^{\mathrm{unif}}(n,h_{\mathrm{s}})$$

... where $s^{\mathrm{unif}}(n,h)$, $\epsilon_{\mathrm{xc}}^{\mathrm{unif}}(n)$ is taken from the uniform electron gas (PRL 110, 146405, 2013, PRL 112, 076403 2014)

Dynamical correction

Inverting the linear response relation

$$\begin{pmatrix} \boldsymbol{\nabla} \bar{v}_{\mathrm{xc}}^{\mathrm{dyn}} \\ \boldsymbol{\nabla} \bar{\psi}_{\mathrm{xc}}^{\mathrm{dyn}} \end{pmatrix} = \left(\underline{\boldsymbol{L}}^{-1} - \underline{\boldsymbol{L}}_{\mathrm{s}}^{-1} \right) \begin{pmatrix} \boldsymbol{\jmath} \\ \boldsymbol{\jmath}_q \end{pmatrix}$$

Dynamical correction

Inverting the linear response relation

$$\begin{pmatrix} \boldsymbol{\nabla}\bar{v}_{\mathrm{xc}}^{\mathrm{dyn}} \\ \boldsymbol{\nabla}\bar{\psi}_{\mathrm{xc}}^{\mathrm{dyn}} \end{pmatrix} = \left[\begin{pmatrix} \rho + \frac{\beta_0\Pi^2}{\kappa} & \frac{\beta_0\Pi}{\kappa} \\ \frac{\beta_0\Pi}{\kappa} & \frac{\beta_0}{\kappa} \end{pmatrix} - \begin{pmatrix} \rho_{\mathrm{s}} + \frac{\beta_0\Pi_{\mathrm{s}}^2}{\kappa_{\mathrm{s}}} & \frac{\beta_0\Pi_{\mathrm{s}}}{\kappa_{\mathrm{s}}} \\ \frac{\beta_0\Pi_{\mathrm{s}}}{\kappa_{\mathrm{s}}} & \frac{\beta_0}{\kappa_{\mathrm{s}}} \end{pmatrix} \right] \begin{pmatrix} \boldsymbol{\jmath} \\ \boldsymbol{\jmath}_q \end{pmatrix}$$

Dynamical correction

Inverting the linear response relation

$$\begin{pmatrix} \boldsymbol{\nabla} \bar{v}_{\mathrm{xc}}^{\mathrm{dyn}} \\ \boldsymbol{\nabla} \bar{\psi}_{\mathrm{xc}}^{\mathrm{dyn}} \end{pmatrix} = \begin{pmatrix} \frac{1}{n} \overleftarrow{\boldsymbol{\nabla}} \eta_{\mathrm{xc}} \overrightarrow{\boldsymbol{\nabla}} \frac{1}{n} + \frac{\beta_0 \Pi^2}{\kappa} & \frac{\beta_0 \Pi}{\kappa} \\ \frac{\beta_0 \Pi}{\kappa} & \frac{\beta_0}{\kappa} \end{pmatrix} \begin{pmatrix} \boldsymbol{\jmath} \\ \boldsymbol{\jmath}_q \end{pmatrix}$$

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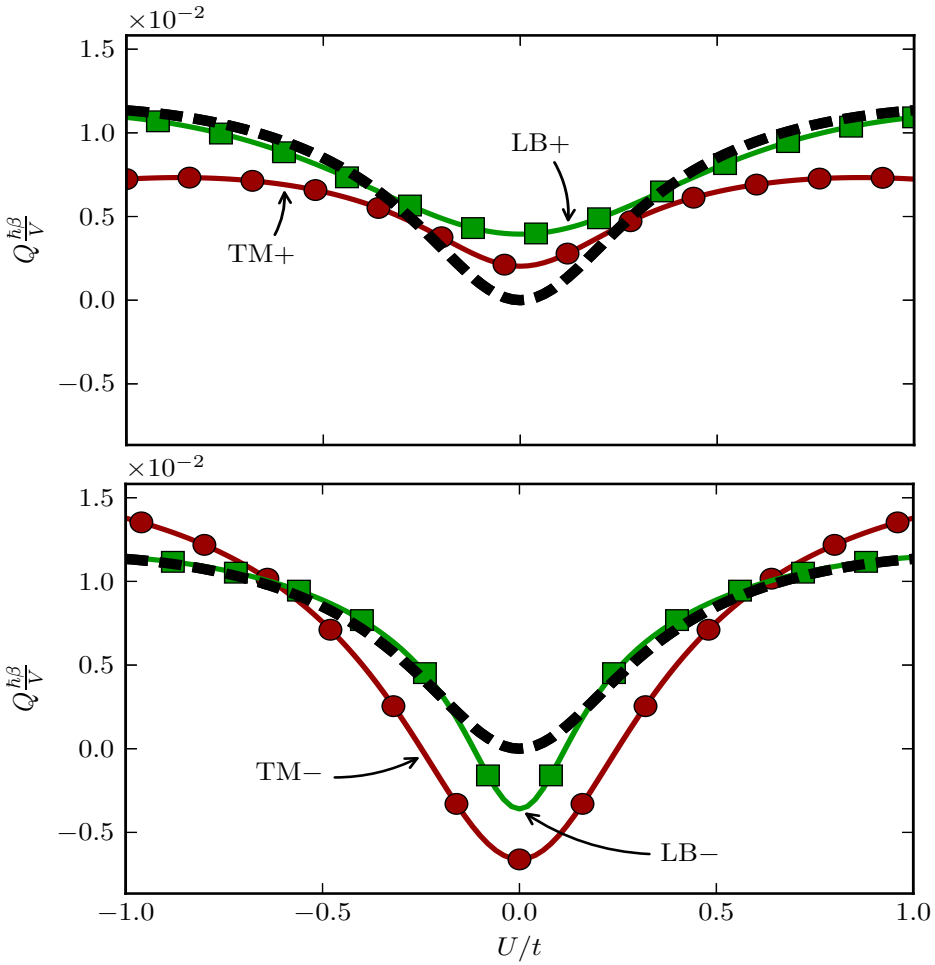
Vignale-Kohn type relaxation

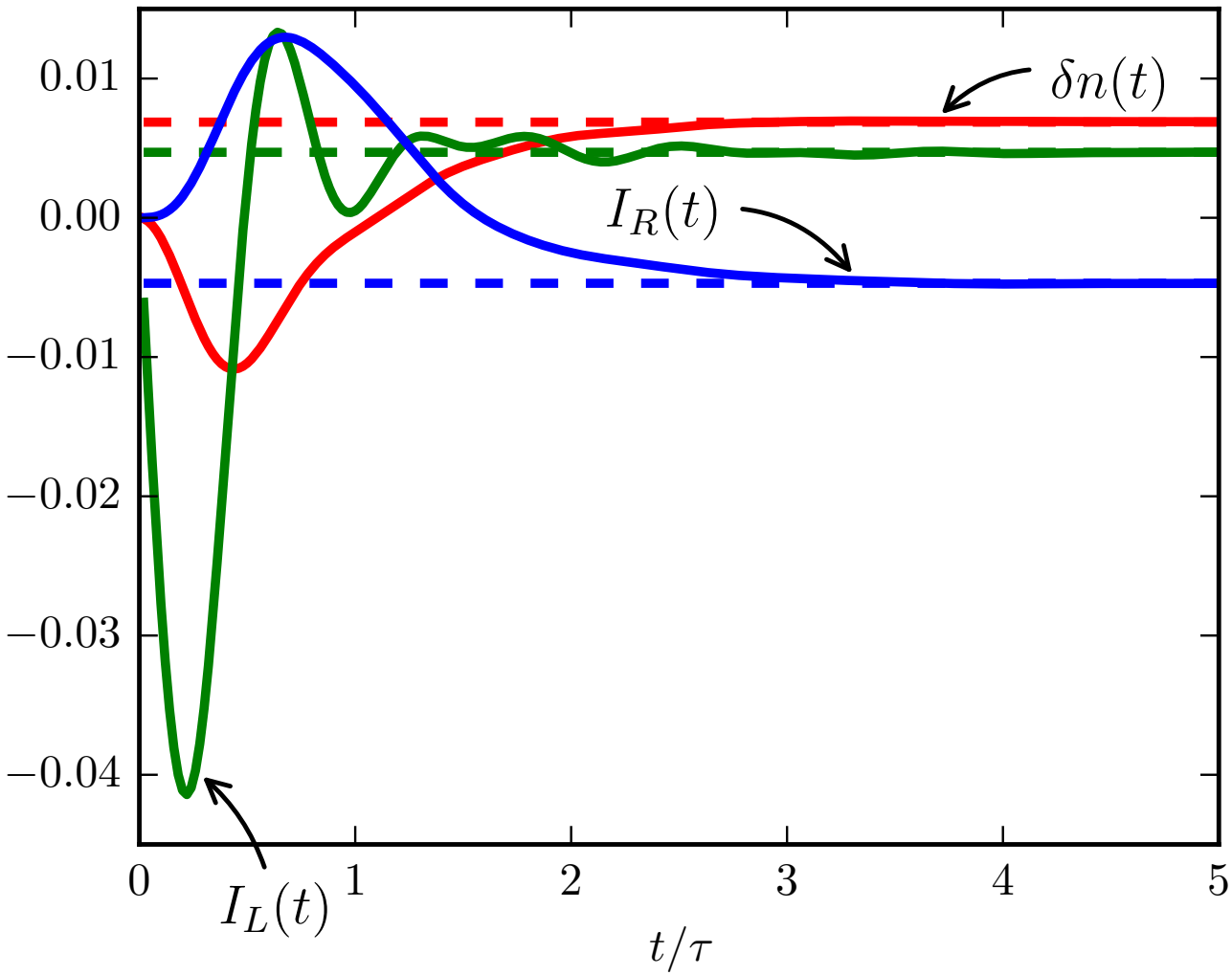
$$W_{\mathrm{xc}} = \int \mathrm{d}^3 r \left\langle \eta_{\mathrm{xc}} \left| \boldsymbol{\nabla} \frac{\boldsymbol{\jmath}}{n} \right|^2 + \frac{\beta_0}{\kappa} \left| \boldsymbol{\jmath}_q + \Pi \boldsymbol{\jmath} \right|^2 \right\rangle$$

Applications

Steady state heat current through single site impurity



Time-dependent density



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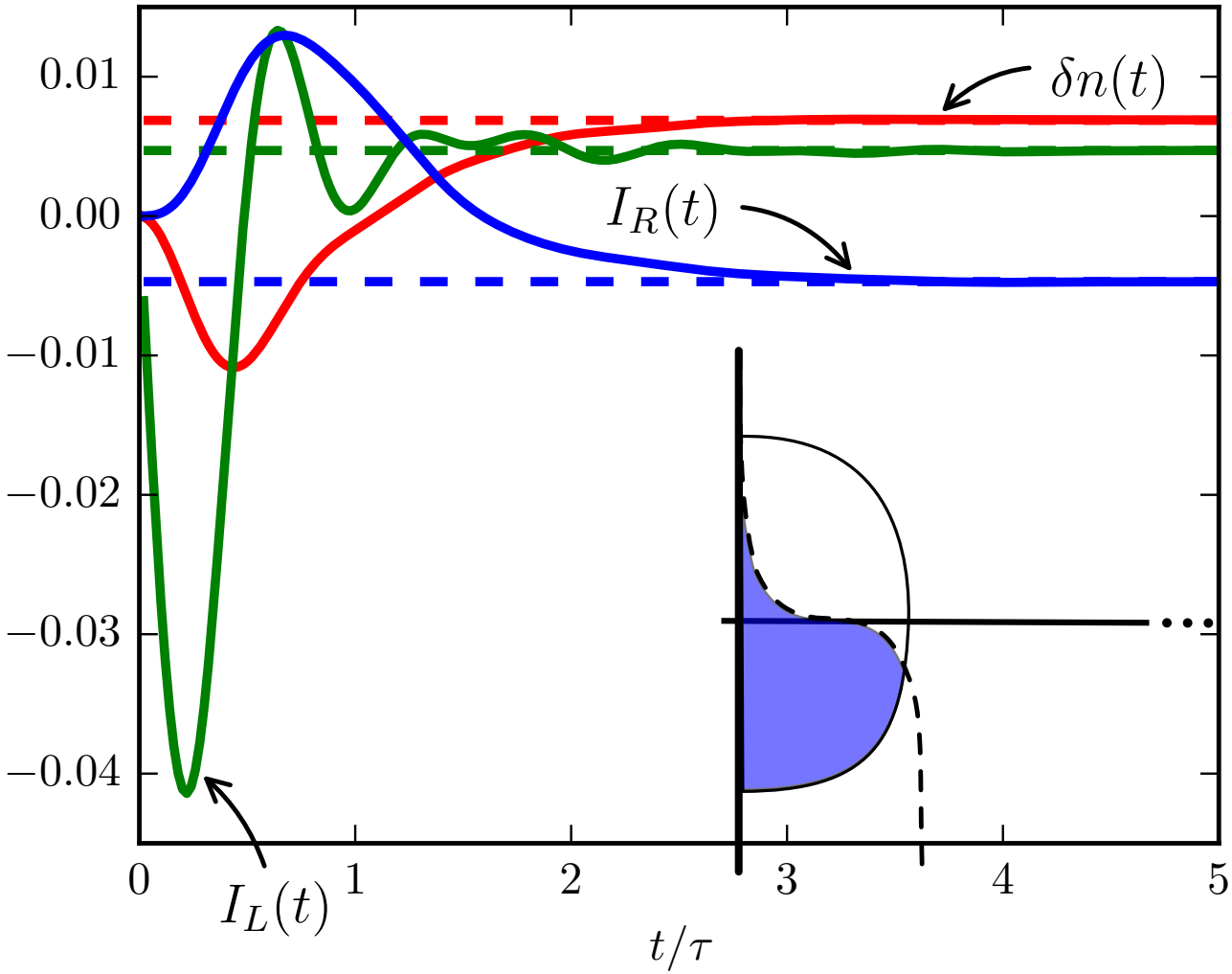

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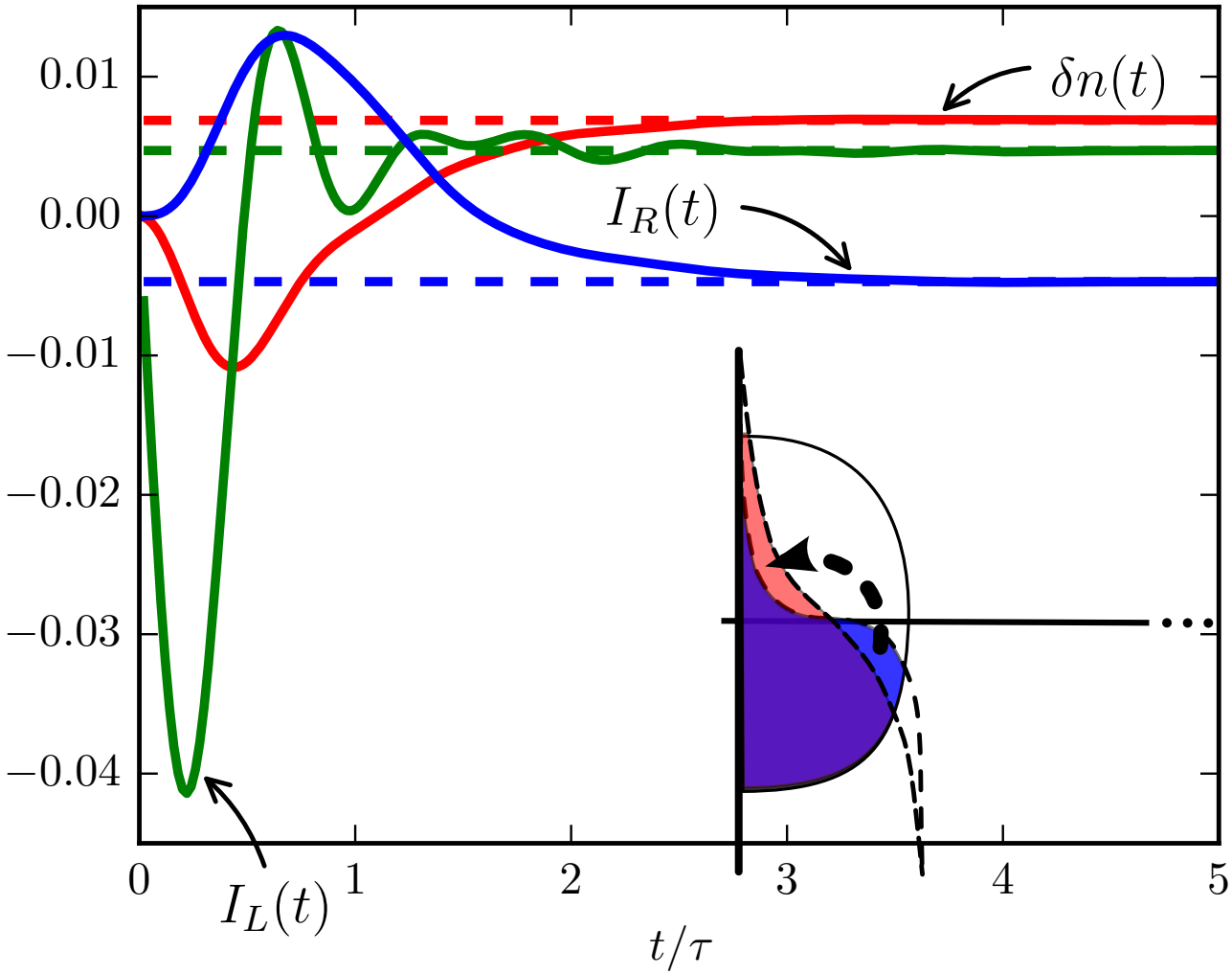

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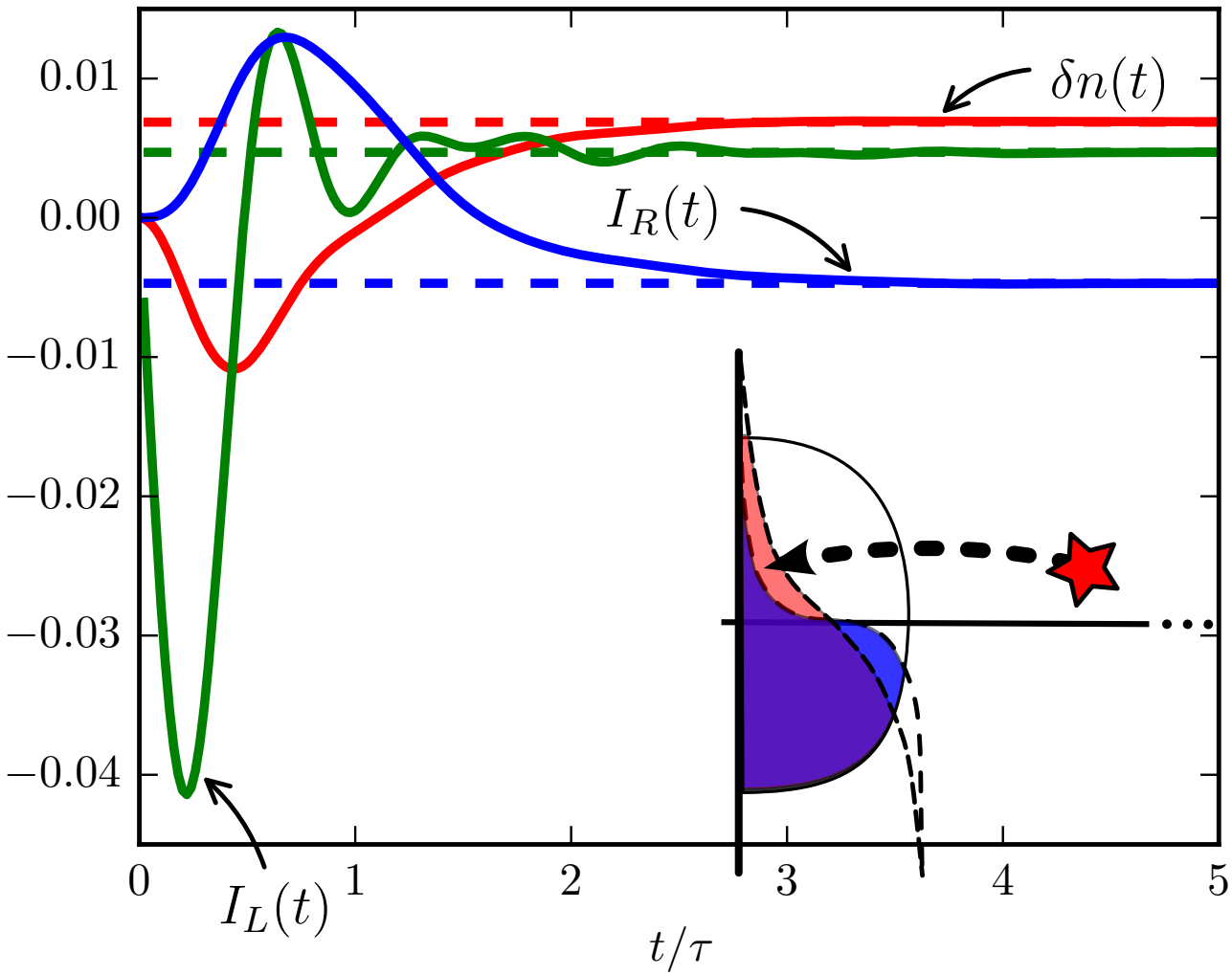
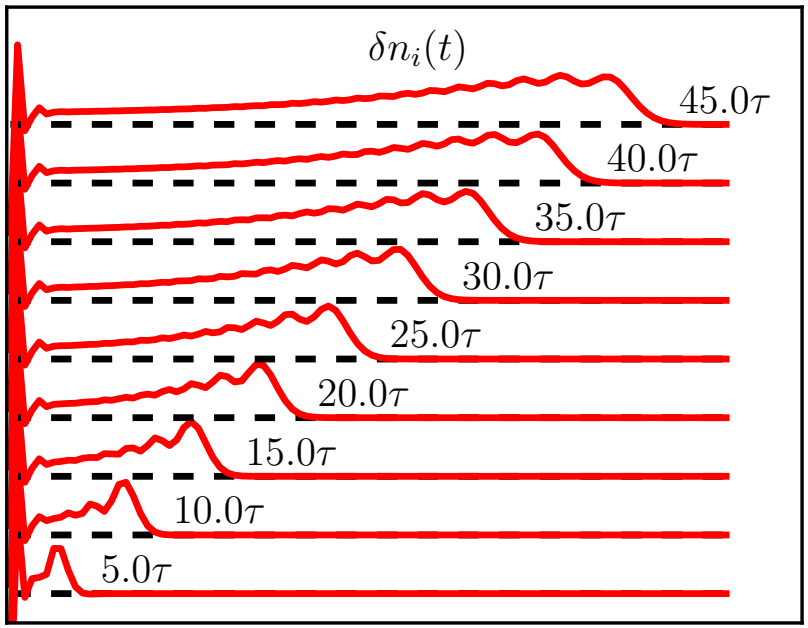
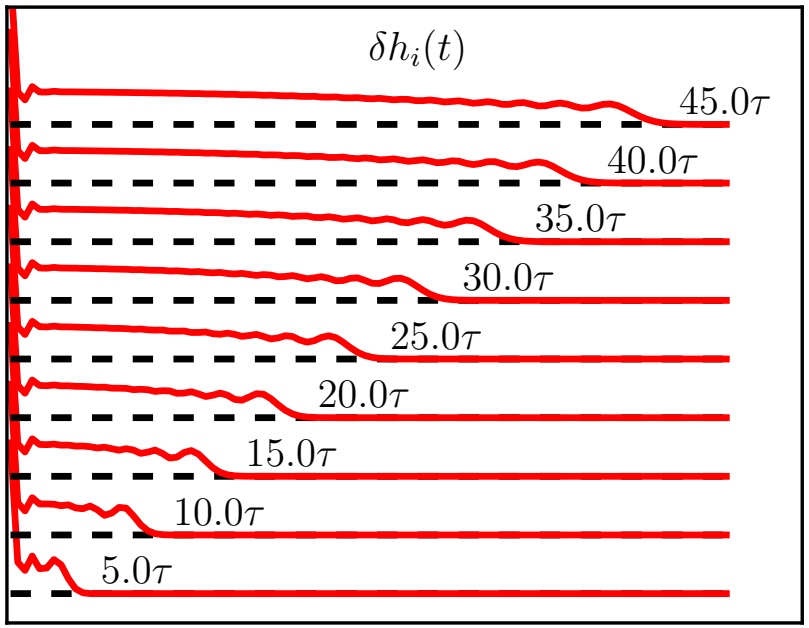


Temperature induced charge and energy wave in atomic chain (100 sites)

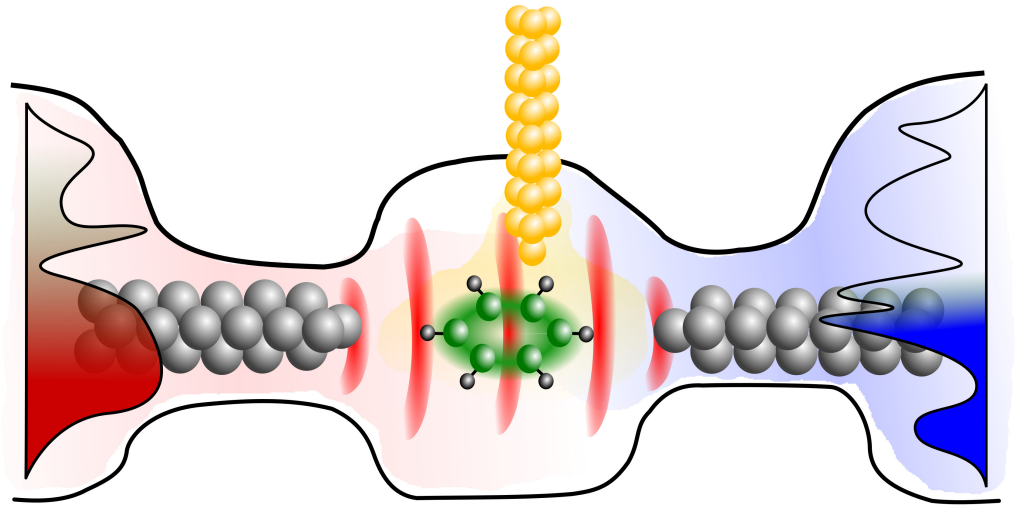




Defining a local temperature via zero-current conditions

$$\mathcal{I}_\alpha = \frac{1}{\hbar}\sum_{\alpha'}\frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\epsilon\, T_{\alpha\alpha'}(\epsilon)\,(f_\alpha - f_{\alpha'})$$

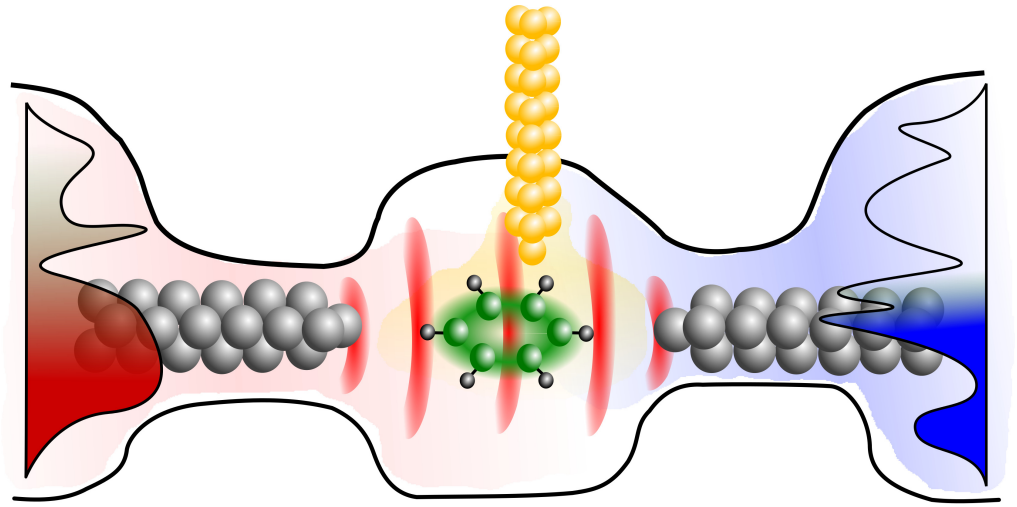
$$\mathcal{J}_\alpha = \frac{1}{\hbar}\sum_{\alpha'}\frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\epsilon\, \epsilon T_{\alpha\alpha'}(\epsilon)\,(f_\alpha - f_{\alpha'})$$



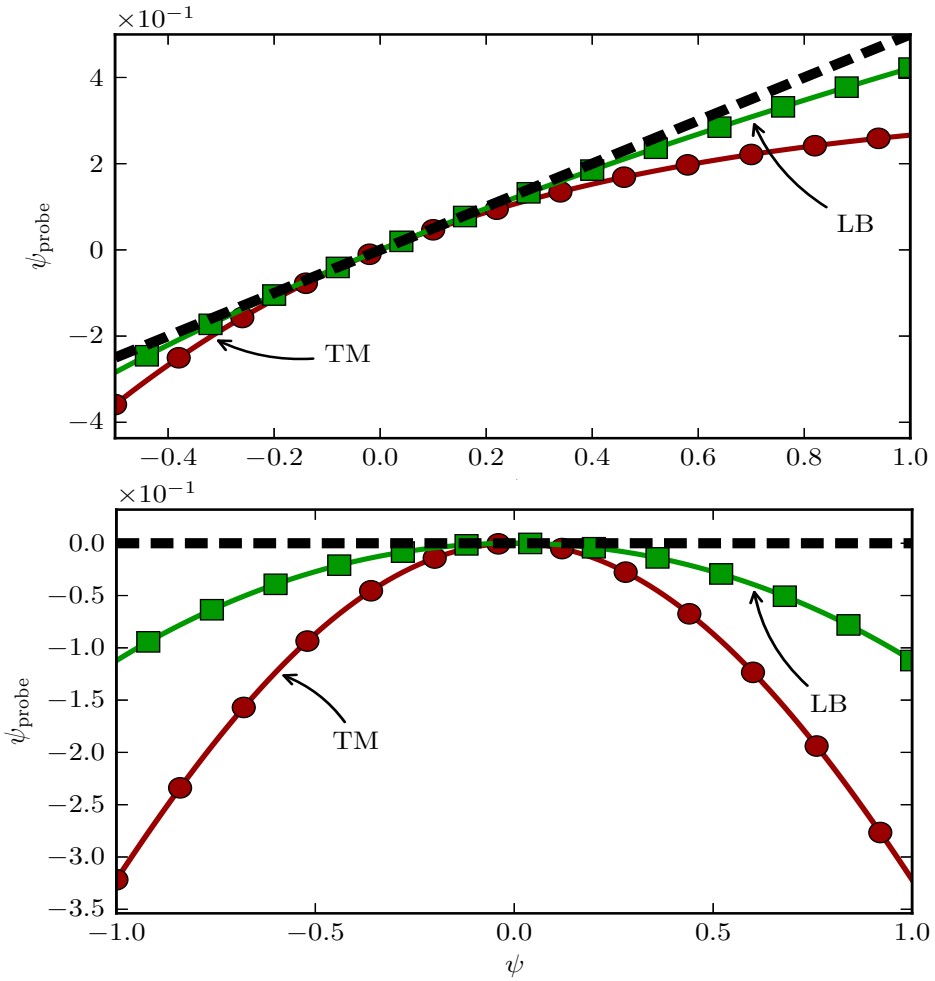
Defining a local temperature via zero-current conditions

$$n_0 = \sum_\alpha \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\epsilon \; D_\alpha(\epsilon) f_\alpha = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\epsilon \; D(\epsilon) f_{\mathrm{probe}}$$

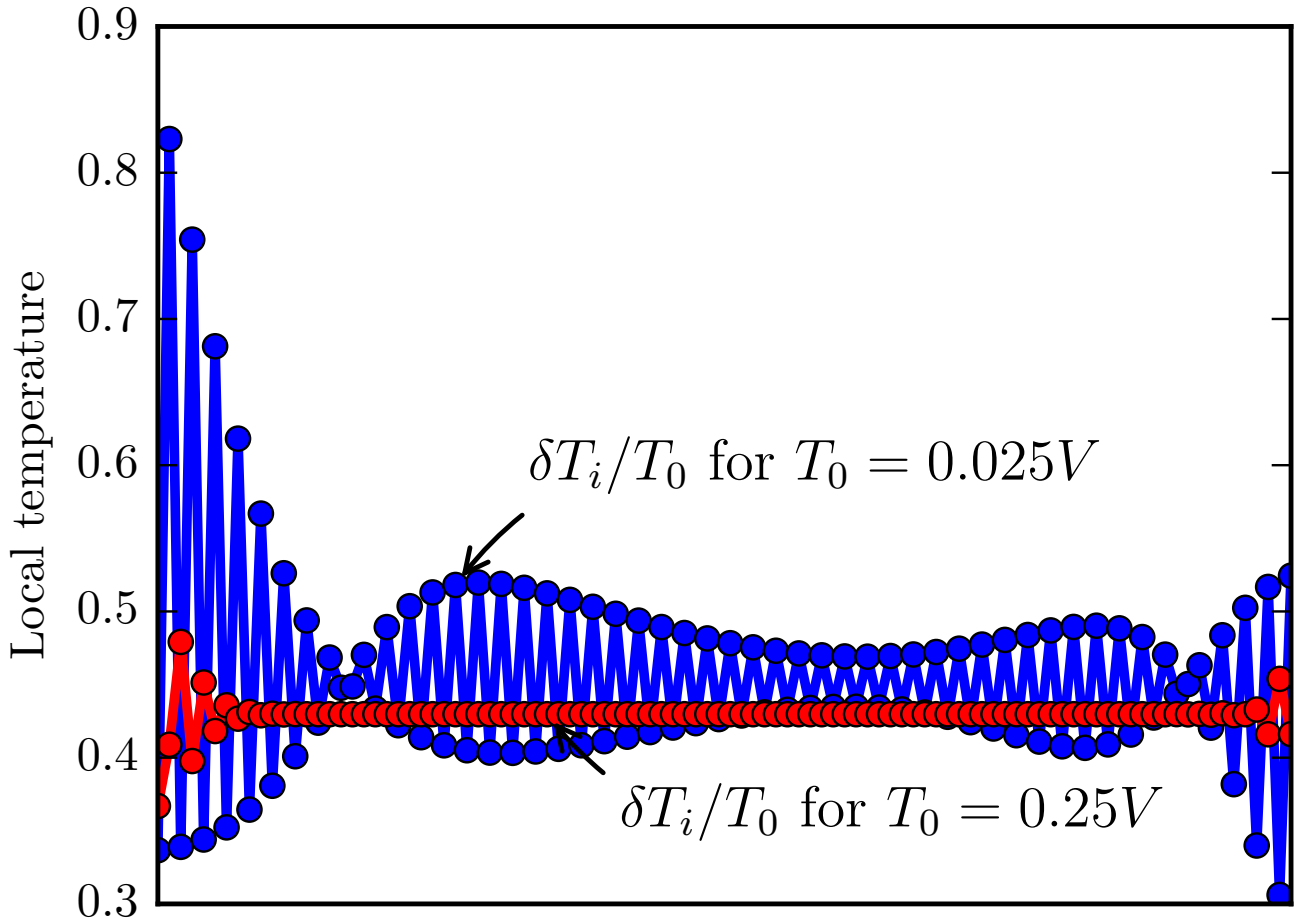
$$h_0 = \sum_\alpha \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\epsilon \; \epsilon D_\alpha(\epsilon) f_\alpha = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathrm{d}\epsilon \; \epsilon D(\epsilon) f_{\mathrm{probe}}$$



Local temperature for single site impurity



Local temperature for atomic chain (100 sites) (I)



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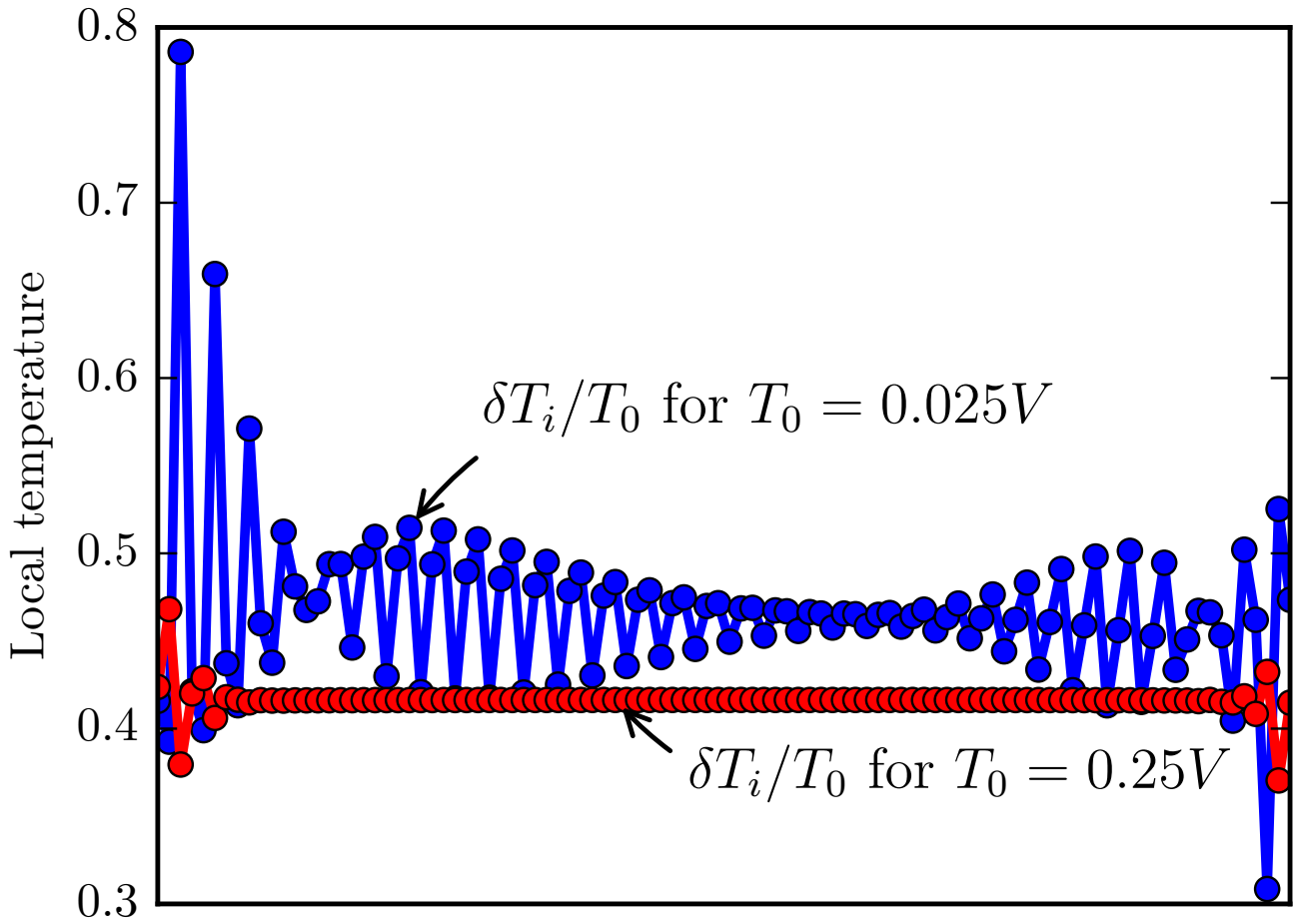

Summary

Conclusion

- Thermal DFT: ab-initio description of thermoelectric transport
- Time-dependent "Thermoelectrics"
- Local temperature at the nano scale

Summary

Conclusion

- Thermal DFT: ab-initio description of thermoelectric transport
- Time-dependent "Thermoelectrics"
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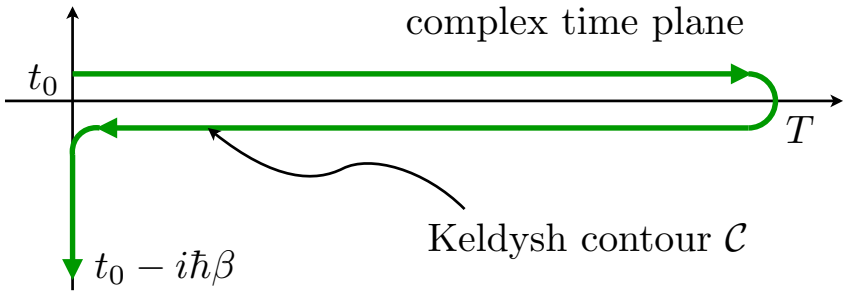
Outlook and open questions

- Development of functionals
- Implementation of "thermal" KS equations
- When does the electron-electron interaction become important for energy transport (Hydrodynamic regime) ?
- What is this ψ potential ? Can it be microscopically justified ?

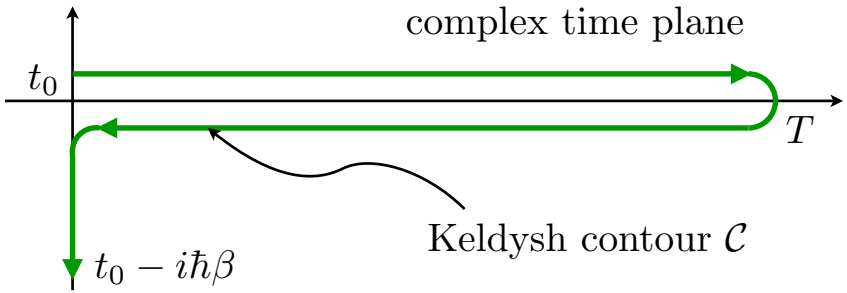
Thank you for you attention!

The action functional



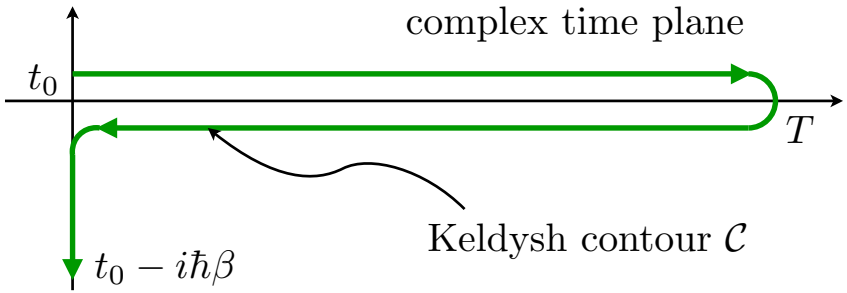
$$A[\tilde{v},\psi] \equiv \imath \ln \mathrm{Tr}\left\{ \mathrm{T}_\tau e^{-\imath \int_{\mathcal{C}} \left[\hat{H} + \int \mathrm{d}^3 r \, \left\{\psi(\boldsymbol{r},\tau)\hat{h}(\boldsymbol{r}) + \tilde{v}(\boldsymbol{r},\tau)\hat{n}(\boldsymbol{r})\right\}\right]} \right\}$$

The action functional



$$A[\tilde{v}^{\mathrm{eq}}, \psi^{\mathrm{eq}}] = \imath \ln \mathrm{Tr}\left\{ e^{-\beta\left[\hat{H} - \mu\hat{N} + \int \mathrm{d}^3 r \, \left\{\psi(\boldsymbol{r})\hat{h}(\boldsymbol{r}) + \tilde{v}(\boldsymbol{r})\hat{n}(\boldsymbol{r})\right\}\right]} \right\}$$

The action functional



$$-\imath\beta\Omega[\tilde{v}^{\mathrm{eq}},\psi^{\mathrm{eq}}] = \imath\,\mathrm{ln}\mathrm{Tr}\Big\{e^{-\beta\left[\hat{H}-\mu\hat{N}+\int \mathrm{d}^3r\,\left\{\psi(\boldsymbol{r})\hat{h}(\boldsymbol{r})+\tilde{v}(\boldsymbol{r})\hat{n}(\boldsymbol{r})\right\}\right]}\Big\}$$

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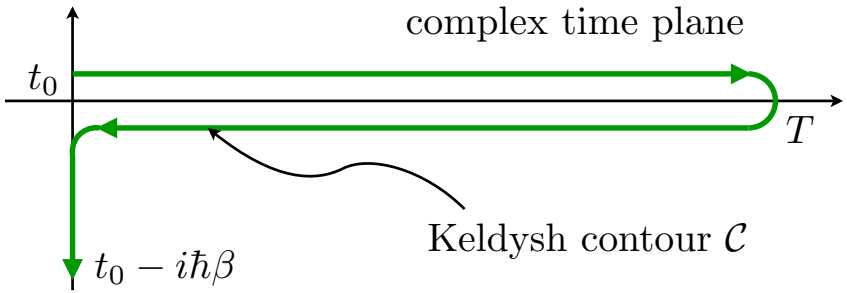

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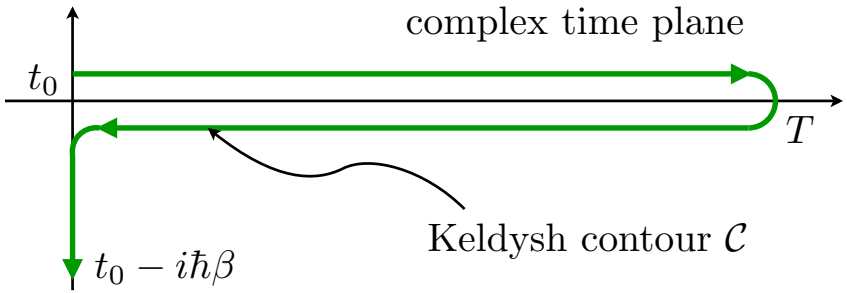
$$h(\boldsymbol{r},\tau) = \frac{\delta A[\tilde{v},\psi]}{\delta \psi(\boldsymbol{r},\tau)}$$

$$n(\boldsymbol{r},\tau) = \frac{\delta A[\tilde{v},\psi]}{\delta \tilde{v}(\boldsymbol{r},\tau)}$$

The action functional



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$$h(\boldsymbol{r},\tau) = \frac{\delta A[\tilde{v},\psi]}{\delta \psi(\boldsymbol{r},\tau)} \qquad n(\boldsymbol{r},\tau) = \frac{\delta A[\tilde{v},\psi]}{\delta \tilde{v}(\boldsymbol{r},\tau)}$$

Legendre transformation yields $A[n,h]$

$$\psi(\boldsymbol{r},\tau) = -\frac{\delta A[n,h]}{\delta h(\boldsymbol{r},\tau)} \qquad \tilde{v}(\boldsymbol{r},\tau) = -\frac{\delta A[n,h]}{\delta n(\boldsymbol{r},\tau)}$$

Decomposition of the action functional

Adiabatic approximation

$$A^{\mathrm{adia}}[n,h] = \int_{\mathcal{C}} F[n(\tau),h(\tau)] = \int_{\mathcal{C}} \int \mathrm{d}^3 r\; h(\boldsymbol{r},\tau) - \frac{1}{\beta}\int_{\mathcal{C}} S[n(\tau),h(\tau)]$$
$$= \int_{\mathcal{C}} \int \mathrm{d}^3 r\; h(\boldsymbol{r},\tau) - \frac{1}{\beta}\bar{S}^{\mathrm{adia}}[n,h]$$

Decomposition of the action functional

Adiabatic approximation

$$A^{\mathrm{adia}}[n,h] = \int_{\mathcal{C}} F[n(\tau),h(\tau)] = \int_{\mathcal{C}} \int \mathrm{d}^3 r\; h(\boldsymbol{r},\tau) - \frac{1}{\beta}\int_{\mathcal{C}} S[n(\tau),h(\tau)]$$
$$= \int_{\mathcal{C}} \int \mathrm{d}^3 r\; h(\boldsymbol{r},\tau) - \frac{1}{\beta}\bar{S}^{\mathrm{adia}}[n,h]$$

Dynamical contribution (everything else!)

$$A[n,h] = \int_{\mathcal{C}} \int \mathrm{d}^3 r\; h(\boldsymbol{r},\tau) - \frac{1}{\beta}\left(\bar{S}^{\mathrm{adia}}[n,h] + \bar{S}^{\mathrm{dyn}}[n,h]\right)$$

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Exchange-correlation contribution

$$\bar{S}_{\mathrm{xc}}[n,h_{\mathrm{s}}] = \bar{S}\left[n, h_{\mathrm{s}} + \epsilon_{\mathrm{Hxc}}[n]\right] - \bar{S}_{\mathrm{s}}\left[n,h_{\mathrm{s}}\right]$$