ADIABATIC PREPARATION OF A CORRELATED SYMMETRY-BROKEN INITIAL STATE

WITH THE GENERALIZED KADANOFF–BAYM ANSATZ

Riku Tuovinen[1], Denis Golež[2], Michael Schüler[2], Philipp Werner[2], Martin Eckstein[3], and Michael Sentef[1]

[1] Max Planck Institute for the Structure and Dynamics of Matter, Germany

[2] University of Fribourg, Switzerland

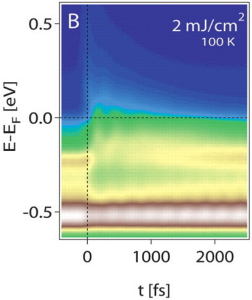
[3] Friedrich-Alexander University Erlangen-Nürnberg, Germany

mpsd
Max-Planck-Institut für
Struktur und Dynamik der Materie

Solving the Two-time Kadanoff–Baym Equations, Kiel, March 11th 2019

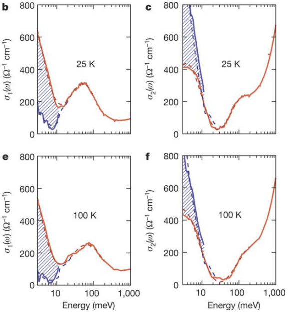
TRANSIENT SPECTROSCOPY OF ORDERED PHASES

Charge-density wave



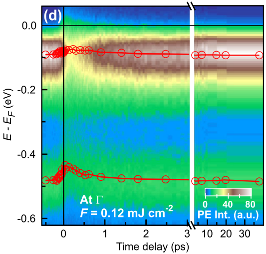
F. Schmitt *et al.*, Science **321**, 1649 (2008)

Superconductivity



M. Mitrano *et al.*, Nature **530**, 461 (2016)

Excitonic insulator



S. Mor *et al.*, Phys. Rev. Lett. **119**, 086401 (2017)

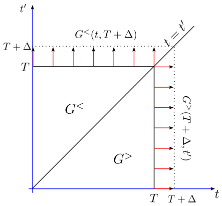
NONEQUILIBRIUM GREEN'S FUNCTION THEORY*†‡

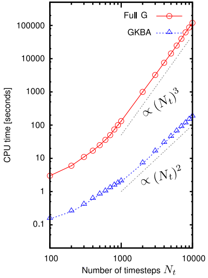
- Two-time Green's functions
  $G(t,t') = -\mathrm{i}\langle T[\hat{\psi}(t)\hat{\psi}^{\dagger}(t')]\rangle$
  (expensive for both CPU and RAM)

$$[\mathrm{i}\partial_t - h\,]G = \delta + \int \mathrm{d}t\ \Sigma\ G$$

System (h)   Many-body effects (Σ)

- Generalized Kadanoff–Baym Ansatz (GKBA) as cheaper alternative
  $G^{\lessgtr}(t,t') \approx$
  $\mathrm{i}\left[G^{\mathrm{R}}(t,t')G^{\lessgtr}(t',t') - G^{\lessgtr}(t,t)G^{\mathrm{A}}(t,t')\right]$





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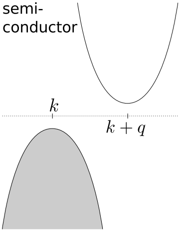
*A. Stan, N. E. Dahlen, and R. van Leeuwen, J. Chem. Phys. **130**, 224101 (2009)

†S. Hermanns, K. Balzer, and M. Bonitz, Phys. Scr. **T151**, 014036 (2012)

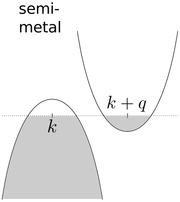
‡RT, D. Golež, M. Schüler, P. Werner, M. Eckstein, and M. A. Sentef, Phys. Status Solidi B (2018) (`arXiv:1808.00712`)

EXCITONIC INSULATOR (EI) PHASE*

Indirect semiconductor (small gap) or -metal (small overlap)



Reduce the gap below exciton binding energy $\Rightarrow$ EI phase



Reduce the overlap $\Rightarrow$ reduce the number of free carriers $\Rightarrow$ less screening $\Rightarrow$ EI phase

$\sim$ BCS superconductivity: electrons form Cooper pairs

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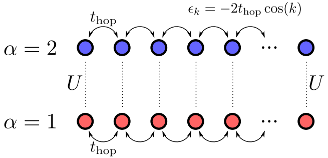
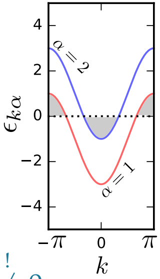
*N. F. Mott, Phil. Mag. **6**, 287 (1961); L. V. Keldysh and Yu. V. Kopaev, Sov. Phys. Solid State **6**, 2219 (1965); D. Jérome, T. M. Rice, and W. Kohn, Phys. Rev. **158**, 462 (1967)

MODEL FOR THE EXCITONIC INSULATOR* †

One-dimensional two-band system with interband Hubbard interaction



$$
\begin{aligned}
\hat{H}(t) &= \hat{H}_{\rm eq} + \hat{H}_{\rm ext}(t), \\
\hat{H}_{\rm eq} &= \sum_{k\alpha} (\epsilon_{k\alpha} + \Delta_\alpha) \hat{c}^\dagger_{k\alpha} \hat{c}_{k\alpha} + \sum_i U \hat{c}^\dagger_{i,1} \hat{c}_{i,1} \hat{c}^\dagger_{i,2} \hat{c}_{i,2}, \\
\hat{H}_{\rm ext}(t) &= \sum_k (E(t) \hat{c}^\dagger_{k,2} \hat{c}_{k,1} + \text{h.c.})
\end{aligned}
$$



Excitonic order parameter $\phi \equiv N_k^{-1} \sum_k{}' \langle \hat{c}^\dagger_{(k+\pi)1} \hat{c}_{k2} \rangle \overset{!}{\neq} 0$

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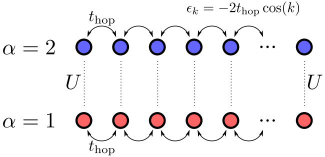
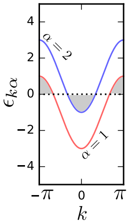
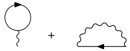
*D. Golež, P. Werner, and M. Eckstein, Phys. Rev. B **94**, 035121 (2016)

†RT, D. Golež, M. Schüler, P. Werner, M. Eckstein, and M. A. Sentef, Phys. Status Solidi B (2018) (`arXiv:1808.00712`)

STARTING POINT: HARTREE–FOCK STATE



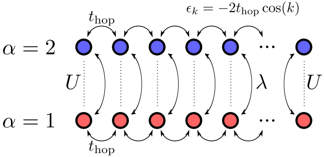


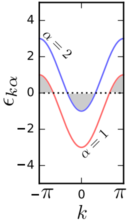
1. $h_0|\varphi_0\rangle = \epsilon_0|\varphi_0\rangle$
2. $\rho_0 = \sum_j f(\epsilon_0^j)|\varphi_0^j\rangle\langle\varphi_0^j|$
3. $\Sigma_{\rm HF}[\rho] =$ 
4. $(h_0 + \Sigma_{\rm HF})|\psi\rangle = \epsilon|\psi\rangle$
5. $\rho = \sum_j f(\epsilon^j)|\psi^j\rangle\langle\psi^j|$

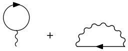
[6. $\rho = a\rho_{\rm new} + (1-a)\rho_{\rm old}$]

7. $\sum_j |\epsilon^j_{\rm new} - \epsilon^j_{\rm old}|/(\epsilon^j_{\rm new} + \epsilon^j_{\rm old}) <$ tolerance

STARTING POINT: HARTREE–FOCK STATE + SEEDING





1. $(h_0 + \lambda)|\varphi_0\rangle = \epsilon_0|\varphi_0\rangle$
2. $\rho_0 = \sum_j f(\epsilon_0^j)|\varphi_0^j\rangle\langle\varphi_0^j|$
3. $\Sigma_{\text{HF}}[\rho] =$ 
4. $(h_0 + \lambda + \Sigma_{\text{HF}})|\psi\rangle = \epsilon|\psi\rangle$
5. $\rho = \sum_j f(\epsilon^j)|\psi^j\rangle\langle\psi^j|$

[6. $\rho = a\rho_{\text{new}} + (1-a)\rho_{\text{old}}$]

7. $\sum_j |\epsilon_{\text{new}}^j - \epsilon_{\text{old}}^j| / (\epsilon_{\text{new}}^j + \epsilon_{\text{old}}^j) <$ tolerance

seeding $\lambda \to 0$

EQUILIBRIUM BY MATSUBARA GREEN FUNCTION

$$
\begin{aligned}
k^{\mathrm{M}}(\tau-\tau') &\equiv -\mathrm{i}k(-\mathrm{i}\tau,-\mathrm{i}\tau') \qquad (k=G,\Sigma) \\
(-\partial_\tau - h_{\mathrm{eq}})G^{\mathrm{M}}(\tau-\tau') &= \delta(\tau-\tau') + \int_0^\beta \mathrm{d}\bar{\tau}\,\Sigma^{\mathrm{M}}(\tau-\bar{\tau})G^{\mathrm{M}}(\bar{\tau}-\tau')
\end{aligned}
$$

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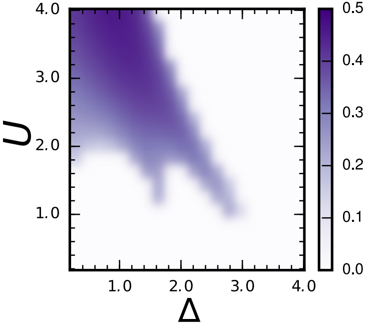
"Phase diagrams" using different self-energy approximations

HF

2B



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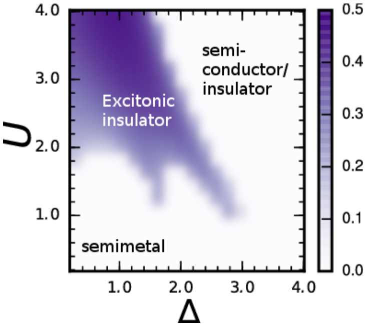
HF 2B



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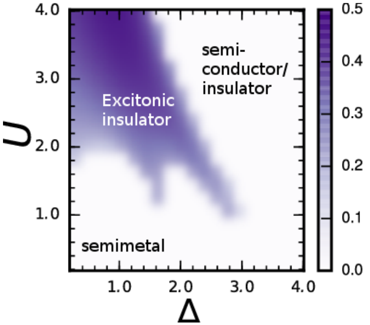
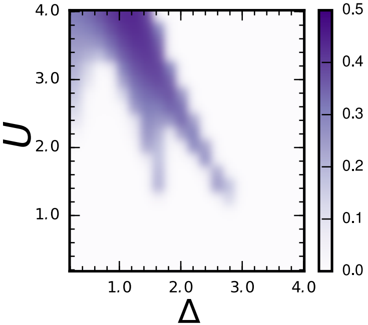


2B



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"Phase diagrams" using different self-energy approximations

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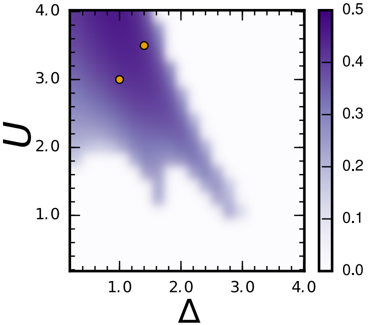

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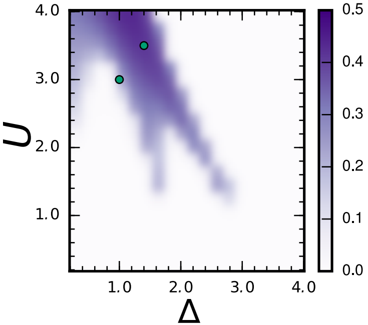
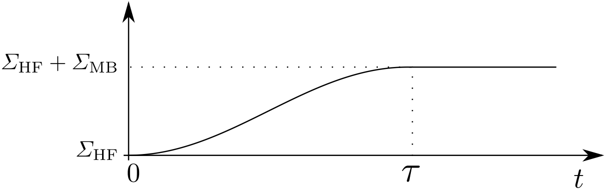


EQUILIBRIUM BY GKBA: ADIABATIC SWITCHING*



*RT, D. Golež, M. Schüler, P. Werner, M. Eckstein, and M. A. Sentef, Phys. Status Solidi B (2018) (arXiv:1808.00712)

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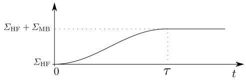

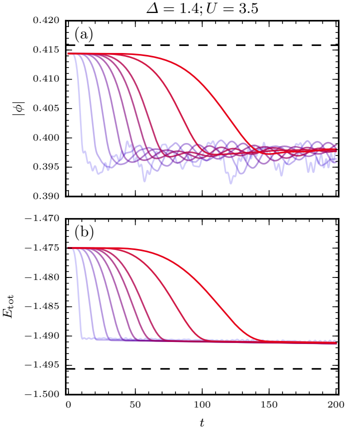

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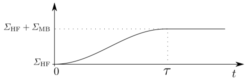

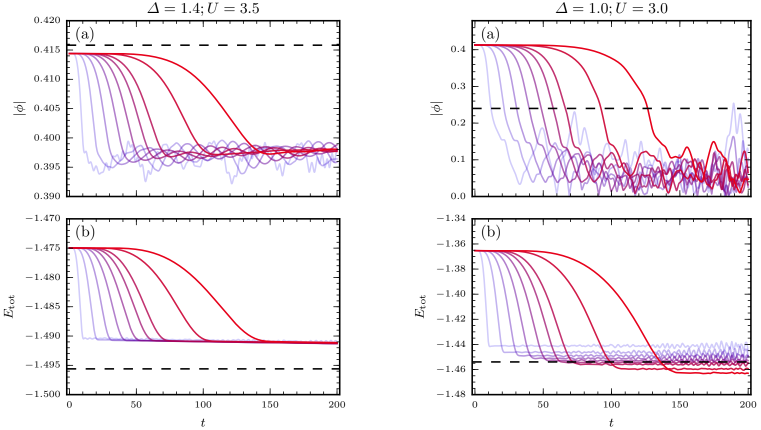

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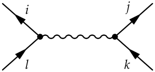
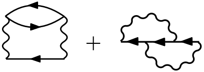
NUMERICAL INTERMEZZO

- Here for simplicity symmetric interaction $v_{ijkl} = v_{ij}\delta_{il}\delta_{jk}$



- 2B self-energy

$\Sigma_{2B} =$



$$= \xi \sum_{kl} v_{ik}(t) v_{jl}(t') G_{ij}(t,t') G_{lk}(t',t) G_{kl}(t,t') \qquad (\xi \in \{1,2\})$$

$$- \sum_{kl} v_{ik}(t) v_{jl}(t') G_{il}(t,t') G_{lk}(t',t) G_{kj}(t,t')$$

- Contract indices to manipulate into entrywise- or normal matrix products (`python: opt_einsum`)
- Use external `linalg` libraries for products (vs. looping)
- Combine with the dissection algorithm*

*E. Perfetto and G. Stefanucci, Phys. Status Solidi B (2019) (`arXiv:1810.03412`)

REMARK: GKBA + INITIAL CORRELATIONS*

In principle, the collision integral should include the vertical track of the time contour

$$\begin{aligned} I(t) &= \int_{t_0}^{t} \mathrm{d}\bar{t}[\Sigma^{>}(t,\bar{t})G^{<}(\bar{t},t) - \Sigma^{<}(t,\bar{t})G^{>}(\bar{t},t)] \\ &- \mathrm{i}\int_{t_0}^{\beta} \mathrm{d}\tau \Sigma^{\rceil}(t,\tau)G^{\lceil}(\tau,t) \end{aligned}$$

*D. Karlsson, R. van Leeuwen, E. Perfetto, and G. Stefanucci, Phys. Rev. B **98**, 115148 (2018)

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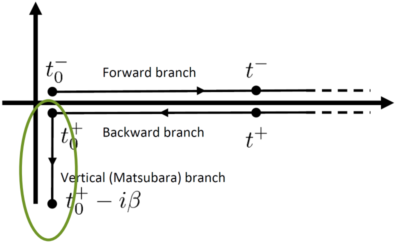
&- \mathrm{i} \int_{t_0}^{\beta} \mathrm{d}\tau \Sigma^{\rceil}(t,\tau)G^{\lceil}(\tau,t)
\end{aligned}
$$

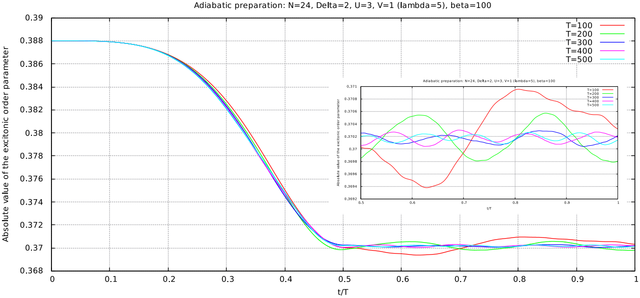


Generalized fluctuation–dissipation theorem $\rightsquigarrow$

$$
I^{\mathrm{ic}}(t) \equiv \int_{-\infty}^{t_0} \mathrm{d}\bar{t} \, [\Sigma^{>}(t,\bar{t})G^{<}(\bar{t},t) - \Sigma^{<}(t,\bar{t})G^{>}(\bar{t},t)]
$$

*D. Karlsson, R. van Leeuwen, E. Perfetto, and G. Stefanucci, Phys. Rev. B **98**, 115148 (2018)

GKBA + INITIAL CORRELATIONS (ORDERED PHASE)



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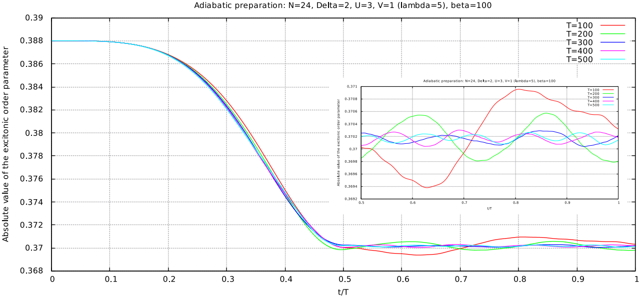

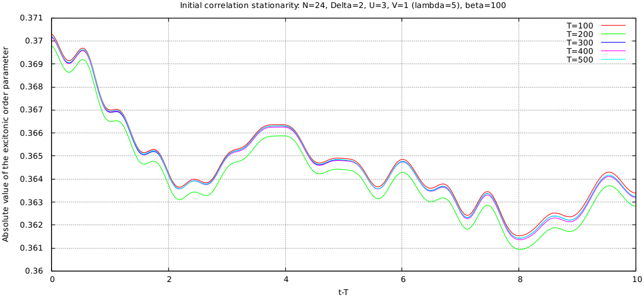

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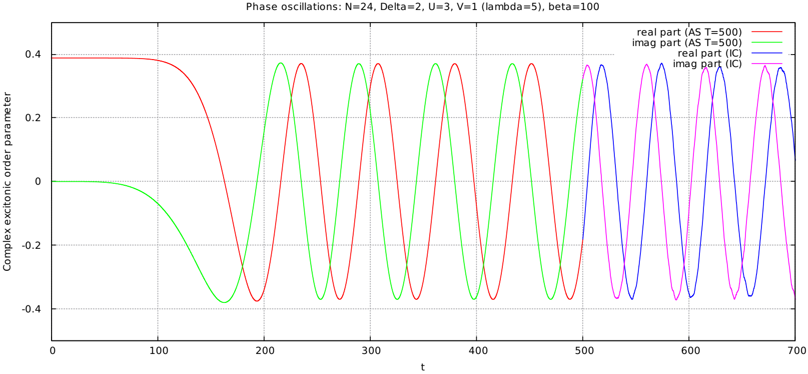

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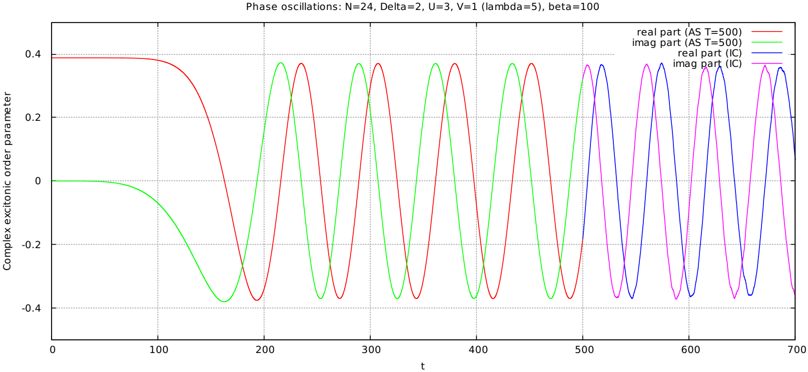
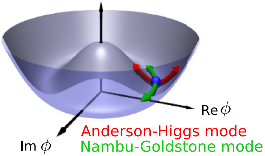
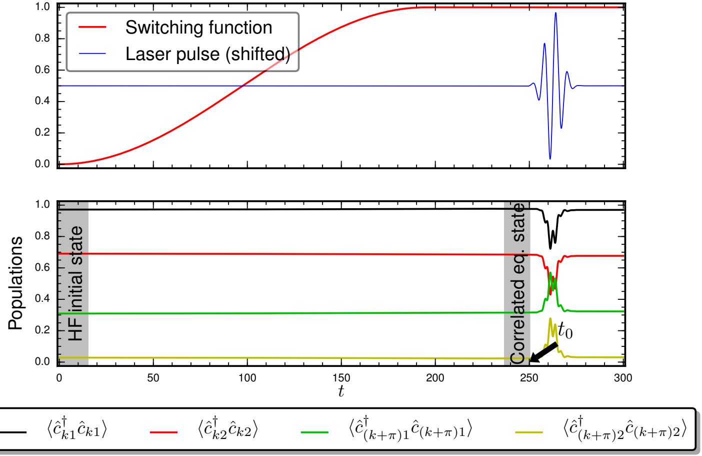
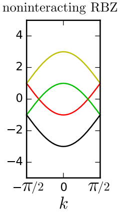




OUT-OF-EQUILIBRIUM: POPULATIONS AND ORDER





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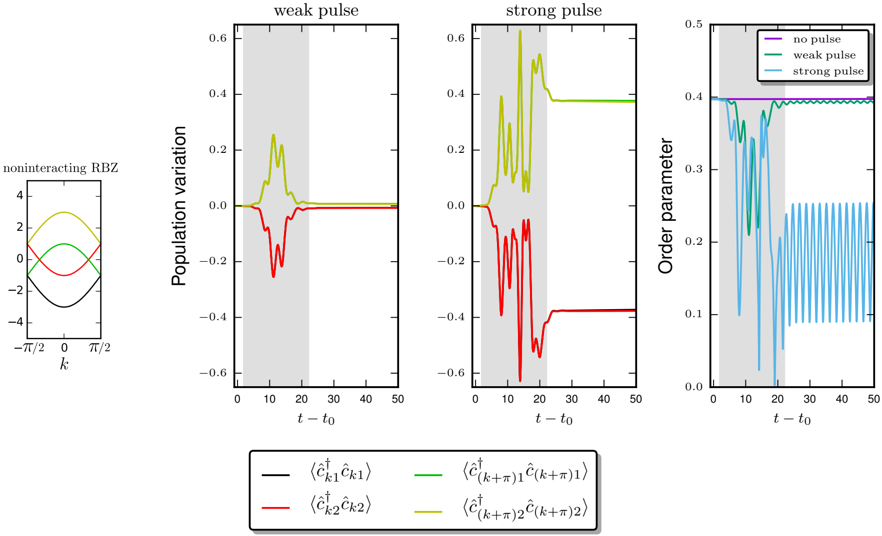

SUMMARY

- Ultrafast experiments available in, e.g., transition-metal dichalcogenide materials exhibiting the EI phase
- Theoretical description is a challenge (electronic correlations, transient regime, . . .)
- Generalized Kadanoff–Baym Ansatz computationally tractable (assess validity vs. full KBE)
- Equilibrium: symmetry-broken correlated initial state with nonzero excitonic order parameter (using the GKBA)
- Out-of-equilibrium: light-induced population inversion and melting of the excitonic condensate

RT, D. Golež, M. Schüler, P. Werner, M. Eckstein, and M. A. Sentef, Phys. Status Solidi B (2018) (arXiv:1808.00712)

mpsd
Emmy Noether-Programm
DFG Deutsche Forschungsgemeinschaft
FNSNF Swiss National Science Foundation
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