Correlated electrons out of equilibrium: Short-time dynamics to quasi-steady states

Martin Eckstein

”Quantum Matter: Computation Meets Experiments”

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# Controlling materials out of equilibrium

*image from Basov et al., Nature Materials 2017*
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Field-induced phenomena

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Distributional engineering

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# Controlling materials out of equilibrium

Field-induced phenomena
Distributional engineering
Non-equilibrium steady states

*(image from Basov et al., Nature Materials 2017)*

*(dc) current-induced switching*
This talk: Overview of attempts to compute electronic structure of correlated electrons under non-equilibrium conditions (following ultra-short excitations or in non-thermal steady states.)

Field-induced phenomena
Distributional engineering
Non-equilibrium steady states

Image from Basov et al., Nature Materials 2017
# Real-time dynamics of correlated electrons

**Hubbard model**

\[
H = -t \sum_{\langle ij\rangle, \sigma=\uparrow,\downarrow} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}
\]

*plus electric fields (Peierls substitution)*

\[
t_{ij} \rightarrow t_{ij} e^{i\phi_{ij}(t)}
\]

\[
\phi_{ji}(t) = e^{\vec{A}(t) \cdot (\vec{r}_j - \vec{r}_i)}
\]

(plus possibly dissipation, phonons)

**Aim:** Unitary time-evolution or dissipative steady states
Real-time dynamics of correlated electrons

Example: single-band Hubbard model after/during laser excitation; $U=3$, bandwidth=4

Intertwined evolution of spectrum and occupation

Spectrum

Occupation $N(\omega, t)$

Charge transfer, field-induced band narrowing

Relaxation

From Eckstein & Werner, PRB 2012
# Non-equilibrium Green’s functions

Keldysh formalism:

\[
A(\omega, t) = -\frac{1}{\pi} \text{Im} \int ds e^{i\omega s} G^{\text{ret}}(t + \frac{s}{2}, t - \frac{s}{2})
\]

\[
N(\omega, t) = -\frac{1}{\pi} \text{Im} \int ds e^{i\omega s} G^{<}(t + \frac{s}{2}, t - \frac{s}{2})
\]

\[\iff \quad G(t, t') = -i \langle Tcc(t)c^\dagger(t') \rangle\]

\(t > 0\): time-dependent H

Initial state: equilibrium
Non-equilibrium Green’s functions

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\[
N(\omega, t) = -\frac{1}{\pi} \text{Im} \int ds \ e^{i\omega s} G^{\text{<}}(t + \frac{s}{2}, t - \frac{s}{2})
\]

\[\iff G(t, t') = -i \langle T c(t) c(\pi + t') \rangle\]

Initial state: equilibrium

\[t > 0: \text{time-dependent } H\]

Time-dep. mean-field solution, DMFT, ...
Non-equilibrium Green’s functions

Computational challenges (I): Long-time-memory in Quantum kinetic equations

\[
G = G_0 + \sum_{\text{previous times}}
\]

\[
\int dt_1 dt_2 G_0(t, t_1) \Sigma(t_1, t_2) G(t_2, t)
\]

open source code: nessi.tuxfamily.org
Non-equilibrium Green’s functions

**Computational challenges (I):** Long-time-memory in Quantum kinetic equations

\[ G(t, t') = G_0(t, t') + \int_{\text{previous times}} dt_1 dt_2 G_0(t, t_1) \Sigma(t_1, t_2) G(t_2, t) \]

"causal" time-propagation:

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Non-equilibrium Green’s functions

Computational challenges (I): Long-time-memory in Quantum kinetic equations

$$ G(t, t') = G_0(t, t') + \sum \int dt_1 dt_2 G_0(t, t_1) \Sigma(t_1, t_2) G(t_2, t) $$

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# Non-equilibrium Green’s functions

## Computational challenges (I): Long-time-memory in Quantum kinetic equations

\[
G = G_0 + \int_{\text{previous times}} dt_1 dt_2 G_0(t, t_1) \Sigma(t_1, t_2) G(t_2, t)
\]

\[G(t, t')\]

„causal“ time-propagation:

open source code: nessi.tuxfamily.org
# Non-equilibrium Green’s functions

Computational challenges (I): Long-time-memory in Quantum kinetic equations

\[ G(t, t') = G_0(t, t) + \Sigma \]

\[
\int dt_1 dt_2 G_0(t, t_1) \Sigma(t_1, t_2) G(t_2, t)
\]

previous times

\[ G(t, t') \]

„causal“ time-propagation:

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„causal“ time-propagation:

Memory: eg. GW simulation

6 orbitals, 400 k-points, W=10eV

\( \Delta t = 0.01/W = 0.06\text{fs} \Rightarrow >1\text{TB for 100fs} \)

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Sometimes overcome by Truncation of memory integrals: Possible if there is time-scale separation

\[ \Sigma(t, t') \approx 0 \text{ for } |t - t'| > t_c \]

Schueler et al, PRB 2018, Stahl et al., in preparation
# Non-equilibrium Green’s functions

Computational challenges (II):

**DMFT:** Quantum impurity problem = impurity in time-dependent bath

\[ G_{imp} = G_{loc} \]
\[ \Sigma_{imp} = \Sigma_{loc} \]

(numerically unbiased) impurity solvers:

- **MPS**  

- **CTQMC**  
  Gull et al. (2012, ...), Cohen et al. (2018)

- **Keldysh QMC**  
  Parcollet e al.

Here: Still NCA
Real-time dynamics of correlated electrons

Example: single-band Hubbard model after/during laser excitation; $U=3$, bandwidth=4

Intertwined evolution of spectrum and occupation

charge transfer, field-induced band narrowing

Relaxation

from Eckstein & Werner, PRB 2012
Real-time dynamics of correlated electrons

Example: single-band Hubbard model after / during laser excitation; \( U=3 \), bandwidth=4

Intertwined evolution of spectrum and occupation

Here: Thermalization at \( T>T_{\text{in}} \)

\[ N(\omega) = f(\omega)A(\omega) \]

U, bandwidth, T of same order: thermalization within few inverse hoppings

(Non-equilibrium DMFT: Quantum kinetic theory for strongly correlated electrons without quasiparticle or relaxation time approximations)
# Lifetime of photo-doped states

\[ \text{time} = \frac{1}{\text{hopping}} \]

Thermalization of the double occupancy after (pretty much any kind of) excitation:

\[ d(t) = d(T_{\text{eff}}) + A e^{-t/\tau} \]

\( T_{\text{eff}} \) determined by total energy
# Lifetime of photo-doped states

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\[ d(t) = d(T_{\text{eff}}) + A e^{-t/\tau} \]

\( T_{\text{eff}} \) determined by total energy

Long-lived non-thermal population in the upper Hubbard band

\( \text{Mott transition} \)

\[ \text{[time]} = \frac{1}{\text{hopping}} \]
# Internal relaxation

- Phonons (quasi external heat bath)
- Non-local charge and spin fluctuations (beyond DMFT)
- Short range (Cluster DMFT)  
- Long range (extended DMFT, DMFT+GW)

Golez, Eckstein, Werner PRB 2015, PRL 2017, PRB 2019
Golez, Boenke Eckstein, Werner PRB 2019
Bittner, Golez, Eckstein, Werner PRB 2019
(Short-range) spin correlations

Hole motion:
- defects in AFM order
- transfer of energy to spin

Active also in paramagnetic phase?
(Short-range) spin correlations

Hole motion:
- defects in AFM order
- transfer of energy to spin

Active also in paramagnetic phase?

Single band 2d Hubbard model $U/t=12$; 2x2 cluster DCA, NCA solver

NN Spin correlations (in the cluster)

$$\Sigma_{t, t'} = \sum_{K} \Sigma_{K, K}(t, t') \theta_{K}(k)$$
(Short-range) spin correlations


single band 2d Hubbard model \( U/t=12 \); 2x2 cluster DCA, NCA solver

NN Spin correlations

Occupied DOS \( N_k(\omega, t) \)

strong spin correlations: inter Hubbard band relaxation within few 1/hopping
(Short-range) spin correlations

single band 2d Hubbard model $U/t=12$; 2x2 cluster DCA, NCA solver

NN Spin correlations

Occupied DOS $N_k(\omega, t)$

strong spin correlations: inter Hubbard band relaxation within few 1/hopping
Long-lived steady states

Spins, phonons, etc …
# Long-lived steady states

Pre-thermal states  Quasi-steady states due to relaxation bottlenecks and constraints
Long-lived steady states

Spins, phonons, etc …

New orders, non-thermal symmetry broken states?

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# Long-lived steady states

Spins, phonons, etc …

New orders, non-thermal symmetry broken states?

Pre-thermal states

Quasi-steady states due to relaxation bottlenecks and constraints

\[ \Rightarrow \text{Controlled by few macroscopic variables?} \]
# Long-lived steady states

Spins, phonons, etc …

New orders, non-thermal symmetry broken states?

**Pre-thermal states**

Quasi-steady states due to relaxation bottlenecks and constraints

⇒ Controlled by few macroscopic variables?

**Non-equilibrium steady states**

External driving and dissipation

Stabilize non-thermal pre-thermal state through weak driving?

see also talk by Z.Lenarzic
# Long-lived steady states
Directly targeting a non-equilibrium steady state
dissipation + continuous driving (bias) \(\Rightarrow\) Non-equilibrium steady state

\[
G(t, t') \rightarrow G(t - t', t_{av} = (t + t')/2) \rightarrow G(\omega) = A(\omega)F(\omega)
\]

Keldysh formalism:

\[
\hat{G}(\omega) = \hat{G}_0(\omega) + \hat{G}_0(\omega)\hat{\Sigma}(\omega)\hat{G}(\omega)
\]

Interaction & dissipation
# Photo-doping by bath

Non-equilibrium steady state:

Two bath coupled to every site of the lattice:
Photo-doping by bath

Non-equilibrium steady state: Two bath coupled to every site of the lattice:

Steady state with occupation in upper band largely independent of bath details if state is controlled by few bottlenecks.
### Photo-doping by bath

**Non-equilibrium steady state:** Two bath coupled to every site of the lattice:

\[ \omega \]

\[ A(\omega) \]

\[ A_{in}(\omega) \]

\[ A_{out}(\omega) \]

\[ \mu_{in} \]

\[ \mu_{out} \]
Photo-doping by bath

Non-equilibrium steady state: Two bath coupled to every site of the lattice:

\[ \omega \]

\[ A(\omega) \]

\[ \mu_{in} \]

\[ A_{in}(\omega) \]

\[ \mu_{out} \]

\[ A_{out}(\omega) \]

\[ \beta_{eff} = 7.79 \]

(a)
Photo-doping by bath

Non-equilibrium steady state:

Two bath coupled to every site of the lattice:

State characterized by two (self-consistently determined) state variables:
Temperature, double occupancy (photo-doping)
Phase-diagram of photodoped system

Scan for different $\mu_{\text{bath}}$:

Photo-doping (doublon and hole filling)
# Phase-diagram of photodoped system

Scan for different $\mu_{\text{bath}}$:

$$1/T_{\text{eff}}$$

Photo-doping (doublon and hole filling)

Instabilities? AFM, CDW, SC, …

$$H = H_{\text{System+Bath}} + \hbar \eta \sum_j \eta^x_j$$

$$\chi_{\eta} = \left. \frac{\langle \eta^x_j \rangle_{\text{ness}}}{\hbar \eta} \right|_{\hbar \to 0}$$
# $\eta$-pairing superconductivity

Singly-occupied sites: AFM

Photo-doping (doublon and hole filling)
# \( \eta \)-pairing superconductivity

![Graph showing inverse temperature vs. temperature with a question mark in the middle.]

- Only doublons & holes: \( \eta \)
- Singly-occupied sites: AFM

Large U limit: only doubly occupied/empty sites

**SU(2)\(_C\) symmetry**

\[
\begin{align*}
\eta^+_j &= (-1)^j c_{j\uparrow}^\dagger c_{j\downarrow}^\dagger \\
\eta^-_j &= (-1)^j c_{j\downarrow} c_{j\uparrow} \\
\eta^z_j &= \frac{1}{2}(n_j - 1)
\end{align*}
\]

\[H_{\text{eff}} = \sum_{\langle ij \rangle} J_{\text{ex}} S_i \cdot S_j\]

Long-range \( \eta \)-pairing: \( \langle \eta^+_j \rangle \equiv \Delta \)

\( \Rightarrow \) Staggered phase Superconductivity (exact excited state on bipartite lattice, Yang, PRL 63, 2144 (1989))
# $\eta$-pairing superconductivity

Singly-occupied sites: AFM

Only doublons & holes: $\eta$

$\eta$-pairing superconductivity

$\eta$-correlations

Peronacci, Schiro, Parcollet 2019

Singly-occupied sites: AFM

Only doublons & holes: $\eta$

$\eta$-pairing superconductivity

Photo-doping (doublon and hole filling)

$\Rightarrow$ photo-excitation to populate doublons?

finite size (ED): Enhancing $\eta$-correlations Kaneko et al, 2018,2019

Pumping doublons $d>1/4$ by driving Peronacci, Schiro, Parcollet 2019
# $\eta$-pairing superconductivity

Extended region of non-thermal superconductivity in the Hubbard model
a real steady state
CDW in attractive Hubbard model

Attractive interaction \( U < 0 \) at half filling:
s-wave superconductivity and charge density wave order degenerate

\[
\begin{align*}
H &= -t \sum_{\langle ij \rangle, \sigma = \uparrow, \downarrow} c_i^{\dagger} c_j \sigma + U \sum_i n_{i \uparrow} n_{i \downarrow} \\
\psi_{CDW} &= \langle n_A - n_B \rangle \\
\psi_{SC} &= 2 \langle c_i \uparrow c_i \downarrow \rangle
\end{align*}
\]

Can one stabilize superconductivity under non-equilibrium conditions?
# Model Hamiltonians

Non-equilibrium steady state: Balance of power input $JV$ and dissipation to bath.

Environment at given temperature

Current $J$

Voltage bias $V$
Attractive HM at half filling: $U = -2.5; \text{ bandwidth } = 4\sqrt{2}$

Coexisting steady state solutions $\Rightarrow$ first-order transition

1) Melting of CDW through charge excitations
2) Metal unstable against infinitesimal CDW fluctuations: $\Rightarrow$ close to thermal mechanism?
Attractive HM at half filling: $U = -2.5$; bandwidth $= 4\sqrt{2}$

Distribution functions:

Only slight deviations from Fermi function (due to el.-el. scattering)

⇒ in the following analysis, first use $T_{\text{eff}}$ to characterize steady state
Attractive HM at half filling: $U = -2.5$; bandwidth $= 4\sqrt{2}$

CDW in steady state as function of effective temperature

- Order parameter in steady state lowered with respect to equilibrium
- CDW susceptibility reduced in steady-state metal
- Effective temperature at transition reduced with respect to equilibrium
# Non-equilibrium phase diagram

Attractive HM at half filling:

- CDW suppressed by current beyond the Joule heating effect
- Robust “supercooled metallic phase”
# Summary & Outlook

Long-lived non-thermal orders through population engineering

Probing non-equilibrium phase-diagram as auxiliary steady state

- Multi-orbital systems
- Hidden states is systems with spin-orbital order
  (Include feedback on lattice?)
- Ideal for unbiased impurity solvers:
  - MPS, CTQMC, Keldysh QMC

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