DMFT (and GW+DMFT) for Systems out of Equilibrium

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Deutsche
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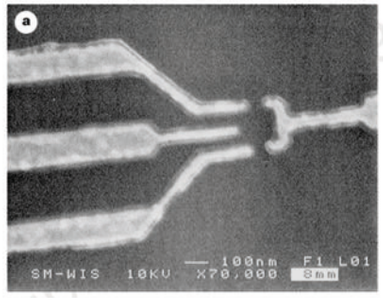
Motivation for out of equilibrium studies

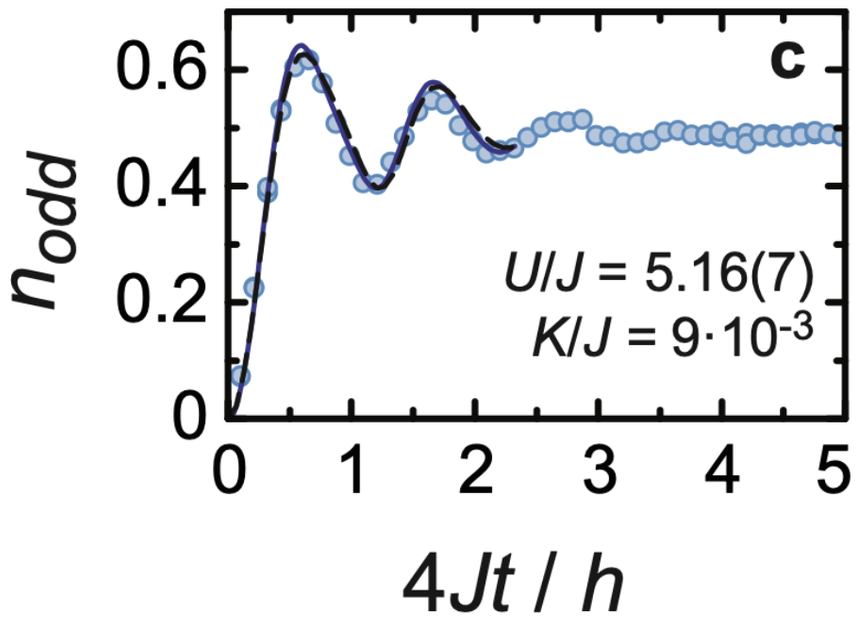
⇨ Real time formulation of many body physics avoids analytical continuation

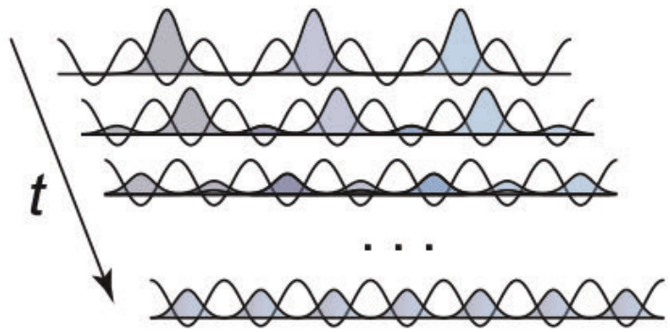
⇨ Transport beyond linear response



from Goldhaber Gordon et al. Nature (1998)

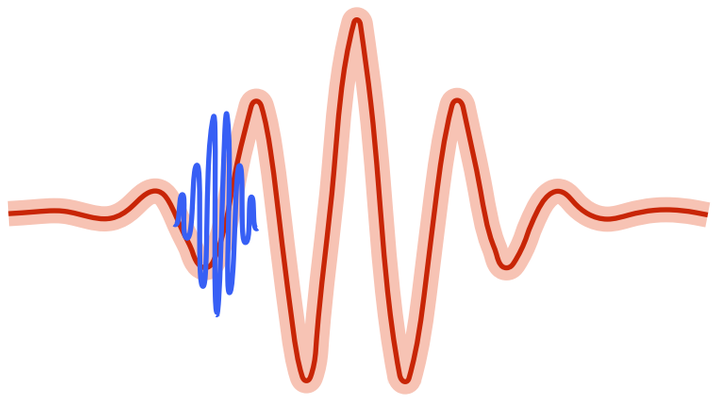
⇨ Controlled quantum many body dynamics in quantum simulators





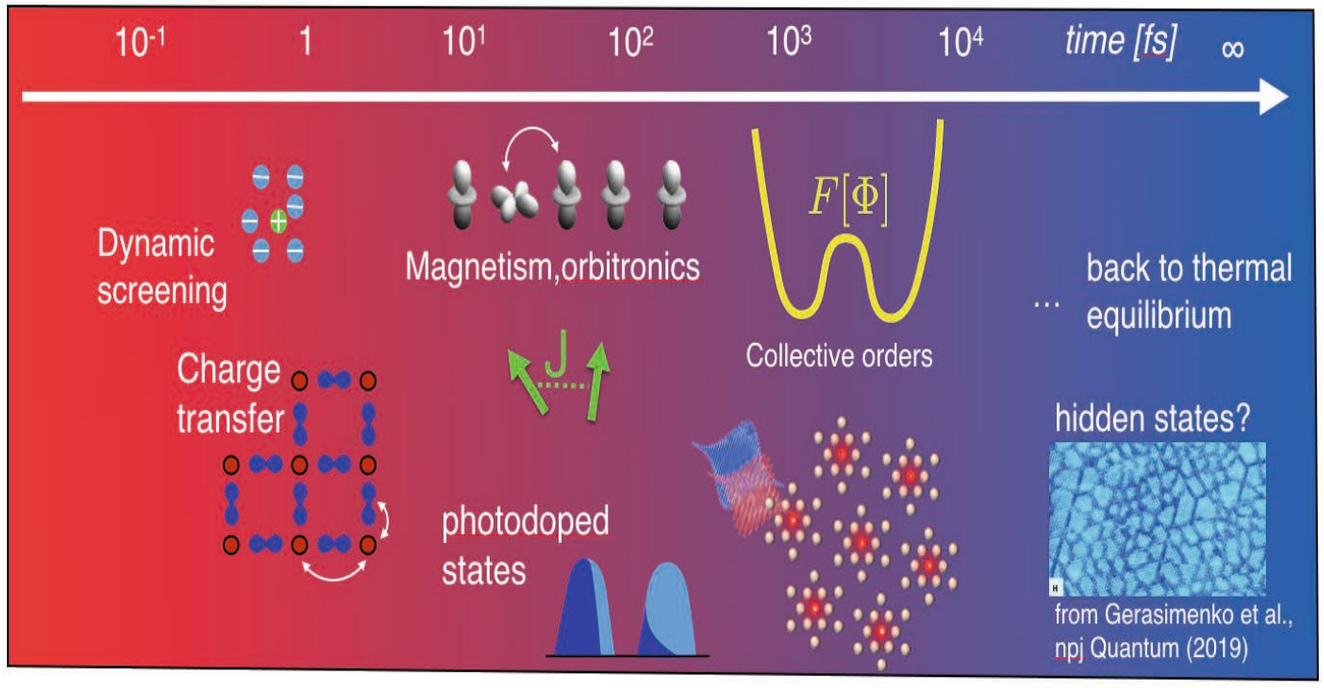
Trotzky et al, Nature Physics (2012)

⇨ Ultrafast dynamics in solids



Ultrafast dynamics in solids

Ultrafast pump-probe experiments: Selective probe of the dynamics of various degrees of freedom on different timescales: Xray, tr-ARPES, XAS



⇨ Reach novel states out of equilibrium?

⇨ What can we learn from the dynamics about the relevant degrees of freedom and their interactions?

some reviews:

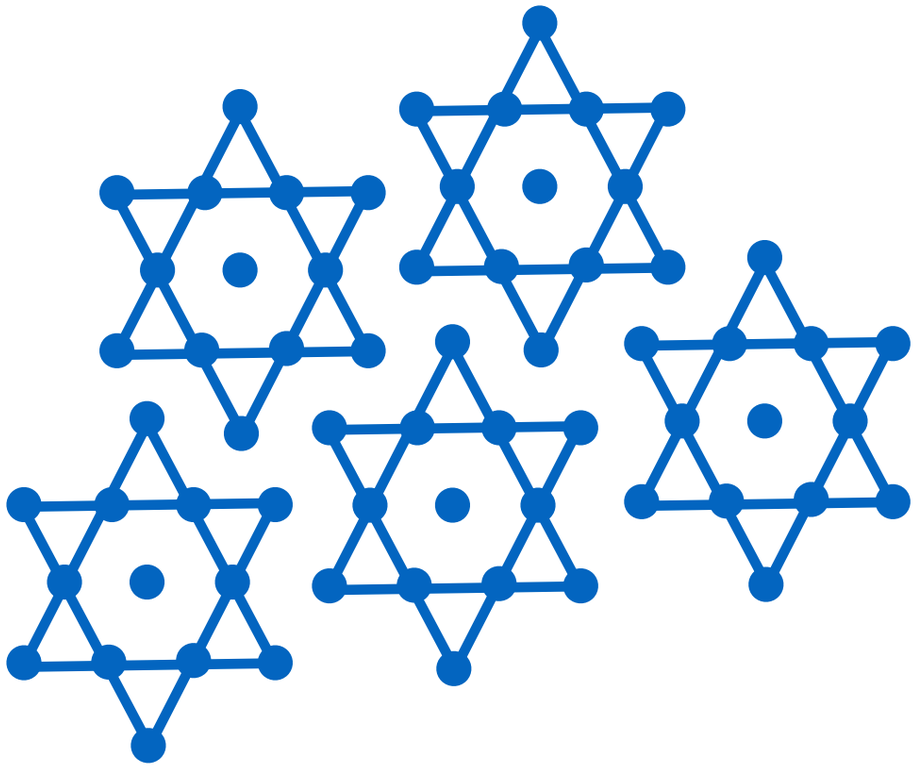
- Aoki et al. Rev. Mod. Phys. 86, 779 (2014)
- Giannetti et al. Advances in Physics, **65**, 58 (2016)
- Basov, Hsieh, Averitt, Nature Materials **16**, 1077 (2017)
- de la Torre et al., Rev. Mod. Phys. **93**, 041002 (2021)

Early motivation for non-equilibrium DMFT:

Perfetti et al. PRL (2006), NJP (2008)

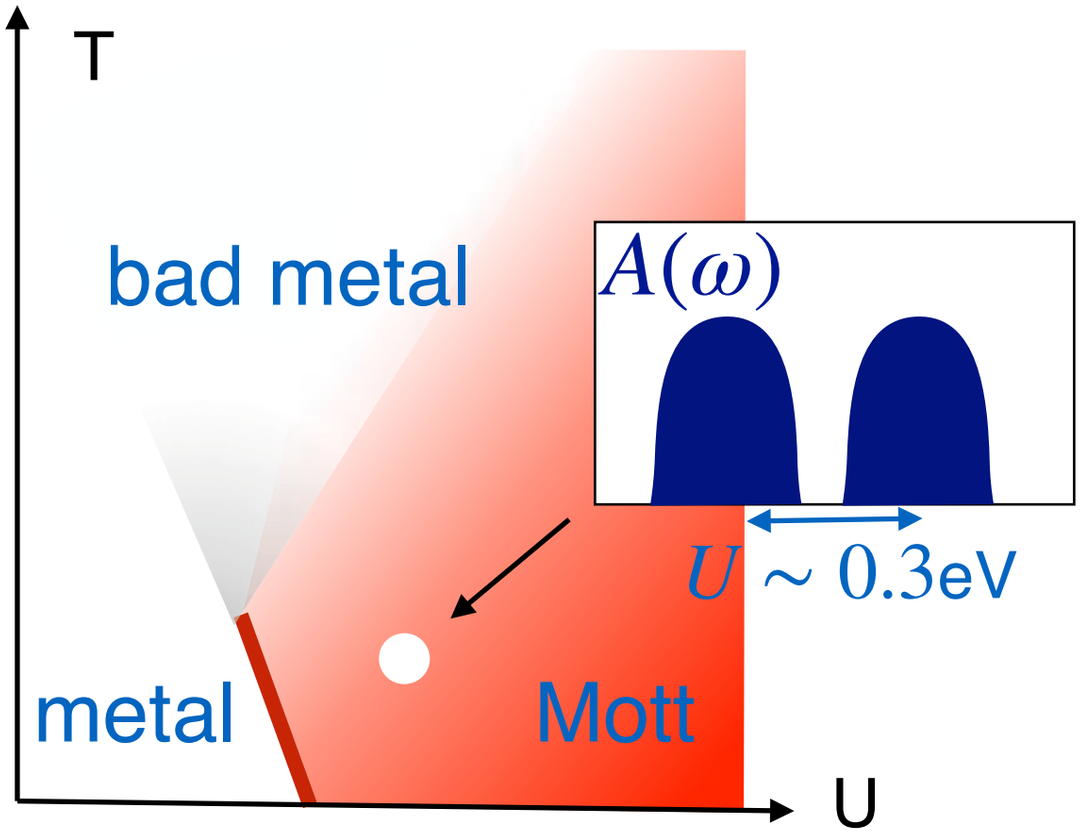
Low temperature state of TaS_2:

13 atom CDW reconstruction



Leaves half-filled valence orbital per cluster

Mott state described by single band Hubbard Model (?)



(AFM order suppressed on triangular lattice)

Early motivation for non-equilibrium DMFT:

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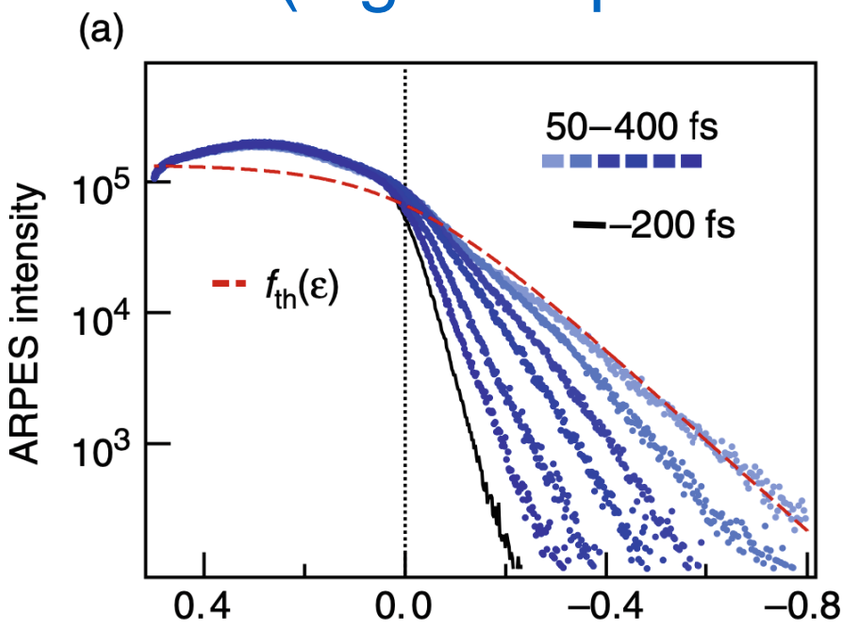
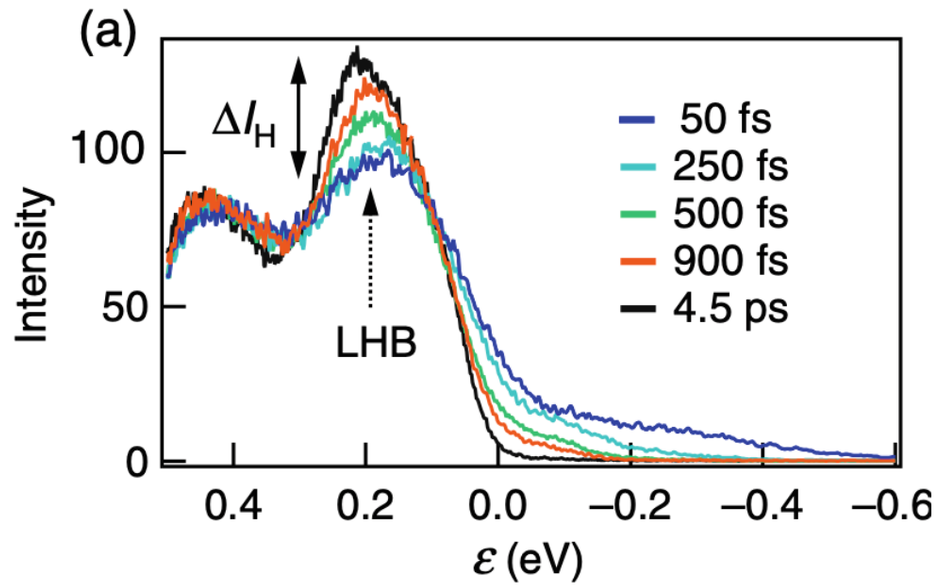
Metal (high temperature):



"hot electrons"

Insulating phase (low temperature):



⇨ Almost instantaneous response of the lower Hubbard band

⇨ Filling in of gap

Thermalization of the electrons? $|\Psi(t)\rangle \longrightarrow \rho \sim e^{-H/T_{eff}}$

(more generally: emergence of universal behavior, governed by few almost conserved quantities)

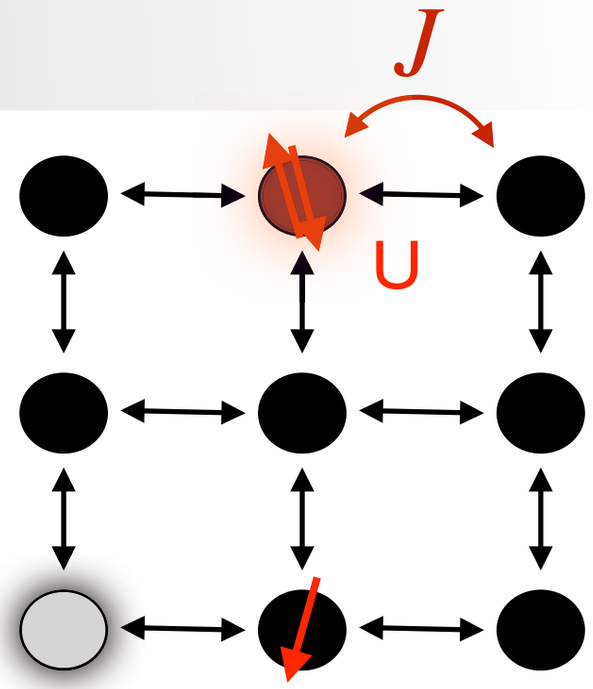
Non-thermal electronic structure?

Photo-induced phase transitions? Hidden phases?

Models

Standard lattice models … such as Hubbard model:

$$H = \sum_{i,j} \sum_{\sigma} J(R_i - R_j)\, c^{\dagger}_{i,\sigma} c_{j,\sigma} + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$



+ Coupling to time-dependent electromagnetic fields: $\vec{E} = -\partial_t \vec{A}(t)$

Peierls Phase $J_{ab} \rightarrow J_{ab} \exp\left\{ -e \int_{\vec{R}_a}^{\vec{R}_b} d\vec{r} \cdot \vec{A}(\vec{r}, t) \right\}$

i.e., for homogeneous fields: $\epsilon(\vec{k}) \rightarrow \epsilon(\vec{k} - e\vec{A}(t))$

Derivation (projecting time-dependent continuum model on Wannier orbitals: Luttinger, Phys. Rev. (1951), Li et. al, PRB 101, 205140 (2020)

(gives also dipolar matrix elements)

(Simple) theoretical approaches

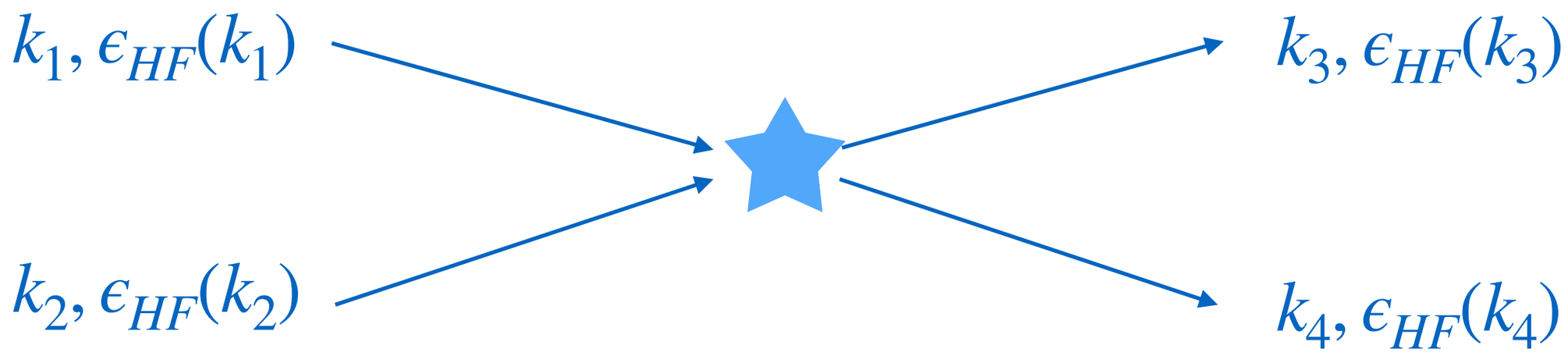
Time-dependent mean field theory (Hartree Fock):

$$U\sum_j n_{j,\uparrow}n_{j,\downarrow} \longrightarrow \sum_{j,\sigma} n_{j,\sigma}V_{j,\sigma}(t) + \text{const.}, \qquad V_{j,\sigma} = U\langle n_{j,\bar{\sigma}}(t)\rangle$$

- Change of band structure due to population transfer (in particular for multi-orbital systems)
- Effective time-dependent potential similar to time-dependent density functional theory (tdDFT) in the adiabatic approximation
- Does not describe thermalization (single particle momentum is conserved)

(Simple) theoretical approaches

Kinetic equations:

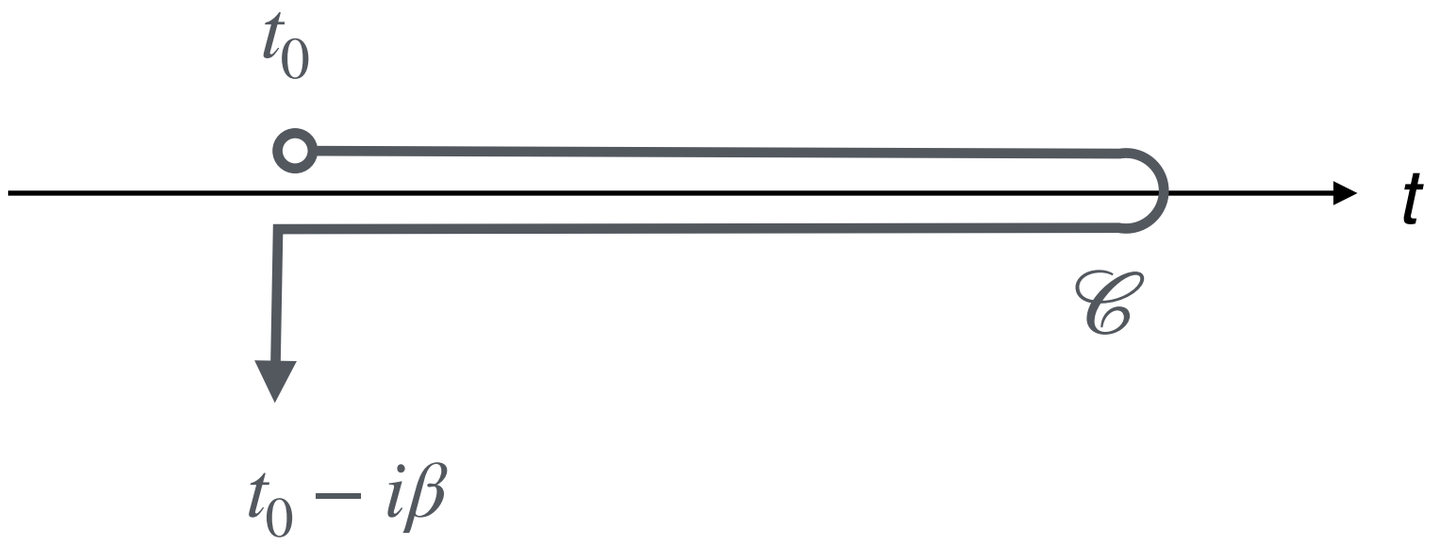


- Quasiparticles well defined in non-equilibrium state?
- Response of electronic structure beyond mean field, beyond rigid bands?

⇨ Non-equilibrium Green's functions techniques:

≡ Non-perturbative quantum kinetic equations without quasiparticle approximation

Keldysh formalism



Formulation of many-body theory in real time

Coupled equations for time-dependent spectrum and occupation

References

- Aoki et al. Rev. Mod. Phys. 86, 779 (2014)
- A. Kamenev, Field theory of non-equilibrium systems

General setting:

- initial state $|\Psi_i\rangle$ or density matrix $\rho = \sum_i w_i |\Psi_i\rangle\langle\Psi_i| = \frac{1}{Z} e^{-\beta H(0)}$
- time evolution $|\Psi_i(t)\rangle = \underbrace{\mathcal{U}(t,t_0)}_{\#} |\Psi_i\rangle$ $\quad \# = T_t \exp\left(-i \int_{t_0}^{t} d\bar{t}\, H(\bar{t}) \right)$

⇨ time-dependent expectation values?

$$\langle O(t)\rangle = \sum_i w_i \langle \Psi_i(t)|O|\Psi_i(t)\rangle = \mathrm{tr}\left[\rho\, \mathcal{U}(t_0,t)\, O\, \mathcal{U}(t,t_0)\right]$$

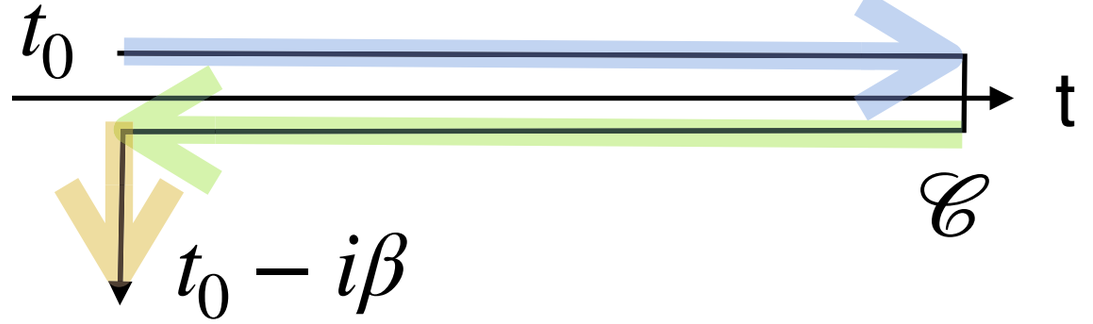
Keldysh contour

$$\langle O(t)\rangle = \mathrm{tr}\left[\rho\, \mathcal{U}(t_0,t)\, O\, \mathcal{U}(t,t_0)\right]$$

⇨ Representation as contour-ordered expectation value:

$$= \frac{1}{Z}\mathrm{tr}\left[\left(T_\tau e^{-\int_0^\beta d\tau\, H(t_0)}\right)\left(\bar{T}_t e^{-i\int_t^{t_0} d\bar{t}\, H(\bar{t})}\right) O \left(T_t e^{-i\int_{t_0}^{t} d\bar{t}\, H(\bar{t})}\right)\right]$$

$$= \frac{1}{Z}\mathrm{tr}\left[T_{\mathscr{C}}\, e^{-i\int_{\mathscr{C}} d\bar{t}\, H(\bar{t})}\, O(t)\right]$$



Contour ordering $T_{\mathscr{C}} A(t)B(t') = \begin{cases} A(t)B(t') & t \text{ later on } \mathscr{C} \\ \pm B(t')A(t) & t \text{ earlier on } \mathscr{C} \end{cases}$

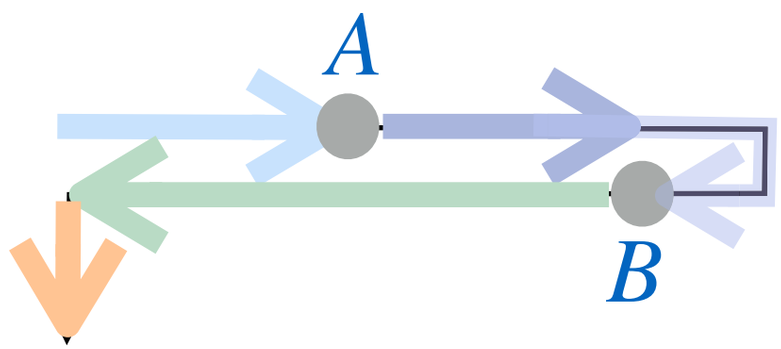
Contour-ordered correlation functions

Analogous: Two- and N-point correlation functions:

$$\langle T_{\mathscr{C}} A(t)B(t')\ \cdots\rangle \equiv \frac{1}{Z}\mathrm{tr}\left(T_{\mathscr{C}}\, e^{-i\int_{\mathscr{C}} d\bar{t}\, H(\bar{t})}\ A(t)B(t')\ \cdots\right)$$

e.g., for $t'_- >_{\mathscr{C}} t_+$:

$\langle T_{\mathscr{C}} A(t_+)B(t'_-)\rangle =$



$$= \pm\frac{1}{Z}\mathrm{tr}\left[\rho\, U(t_0,t')B\,U(t',t)A\,U(t,t_0)\right]$$

real-time-correlation function

Contour ordering: convenient bookkeeping of different operator orderings

(… which all have different physical significance, see below)

Keldysh path integral

- Contour-ordered ordered evolution operator on “closed contour” has path integral representation of analogous to imaginary-time contour:

$$\mathrm{tr}\left(T_{\mathscr{C}}\, e^{-i\int_{\mathscr{C}} d\bar{t}\, H(\bar{t})} \ \ldots \right) = \int \mathscr{D}[\bar{c}, c]\, e^{iS_{\mathscr{C}}} \ \ldots \quad S_{\mathscr{C}} = \int_{\mathscr{C}} dt \left[\bar{c}(t) i\partial_t c(t) - H(t) \right]$$

integrate over all (anti)-periodic path $c(0_+) = \pm\, c(-i\beta)$

- Check: Restriction to imag. time contour: $t = -i\tau, \quad \tau \in [0,\beta]$:

$$i\int_{\mathscr{C}} dt \to \int_0^\beta d\tau, \quad \partial_t \to i\partial_\tau \quad \Rightarrow \quad e^{iS_{\mathscr{C}}} \to e^{-\int_0^\beta d\tau \left[\bar{c}\partial_\tau c + H(t) \right]}$$

usual imaginary time action

⇨ Concepts like Wick’s theorem, effective action, diagrammatic perturbation theory, field theoretical tricks like Hubbard Stratonovich transformation … carry over 1:1 to Keldysh formalism

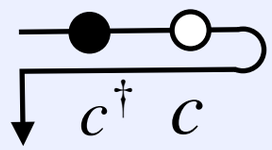
Contour-ordered Green's functions

- Contour-ordered Green's functions

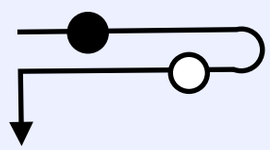
$$G(t,x,t',x') = -i\langle T_{\mathscr{C}}\, c_x(t) c^\dagger_{x'}(t')\rangle:$$

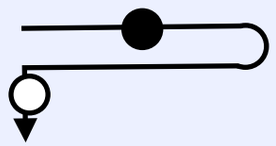
x, x': spin/orbital/momentum indices, omitted in the following ⇨
$G(t,t')$ is a matrix in orbital indices

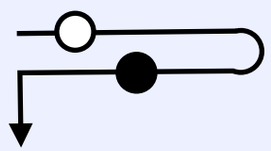
- $\mathscr{C}$-ordering ≡ bookkeeping of operator orderings … here there are 9:

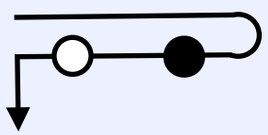
$G(t_+, t'_+) =$ 

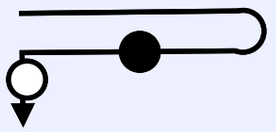
$\equiv G^t(t,t')$

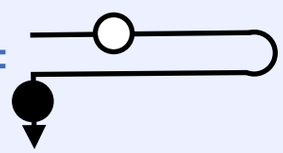
$G(t_-, t'_+) =$ 
$-i\langle c(t)c^\dagger(t')\rangle \equiv G^>(t,t')$

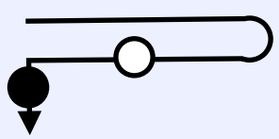
$G(-i\tau, t_+) =$ 
$\equiv G^{vt}(\tau,t')$

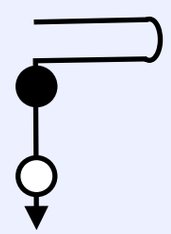
$G(t_+, t'_-) =$ 
$i\langle c^\dagger(t')c(t)\rangle \equiv G^<(t,t')$

$G(t_-, t'_-) =$ 
$\equiv G^{\bar{t}}(t,t')$

$G(-i\tau, t_-) =$ 
$= G^{vt}(\tau,t')$

$G(t_+, -i\tau) =$ 
$\equiv G^{tv}(t,\tau)$

$G(t_-, -i\tau) =$ 
$= G^{tv}(t,\tau)$

$G(-i\tau, -i\tau') =$
$= -i\langle T_\tau c(\tau)c^\dagger(\tau')\rangle$
$= iG^M(\tau-\tau')$


Contour-ordered Green's functions

Equilibrium:

- Translational invariance in time
- Green's functions related to spectrum $A(\omega)$ and universal distribution function $f(\omega) = (e^{\beta\omega} \pm 1)^{-1}$

$$A(\omega) = -\frac{1}{\pi}\mathrm{Im}\, G^R(\omega + i0) \quad G^R(t-t') = -i\theta(t-t')\langle [c(t), c^\dagger(t')]\rangle$$

⇨ "fluctuation dissipation relations":

$G^<(\omega) = 2\pi i\, A(\omega) f(\omega)$ "occupied DOS", photoemission

$G^>(\omega) = -2\pi i\, A(\omega)[1 - f(\omega)]$ "unoccupied density of states"

⇨ Relation to imag time: $G^M(\tau) = -\int d\omega\, A(\omega) e^{-\omega\tau} f(-\omega)$

(analytical continuation)

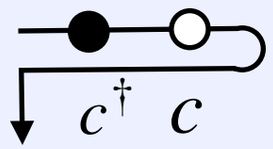
Non-equilibrium Green's functions

Out of equilibrium:

- breaking of time-translational invariance $X(t,t')$ or $X(\omega,t)$
- real time $G(t,t')$ parametrized by two independent functions:

$G^R(t,t') \leftrightarrow$ spectrum $A(\omega,t)$

$G^<(t,t') \leftrightarrow$ non-universal distribution $G^<(\omega,t) = 2\pi i A(\omega,t)F(\omega,t)$

$G(t_+,t'_+) =$ 

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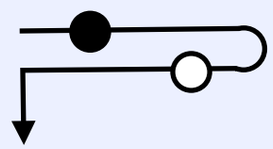
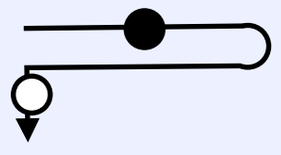
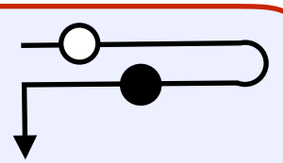
$G(t_-,t'_+) =$ 

$-i\langle c(t)c^\dagger(t')\rangle \equiv G^>(t,t')$

$G(-i\tau,t_+) =$ 

$\equiv G^{vt}(\tau,t')$

$G(t_+,t'_-) =$ 

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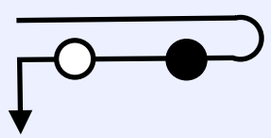
$G(t_-,t'_-) =$ 

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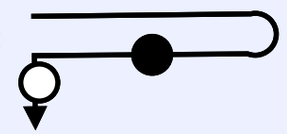
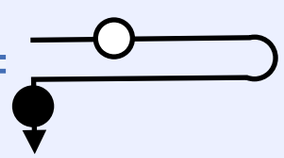
$G(-i\tau,t_-) =$ 

$= G^{vt}(\tau,t')$

$G(t_+,-i\tau) =$ 

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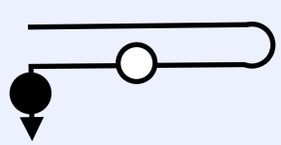
$G(t_-,-i\tau) =$ 

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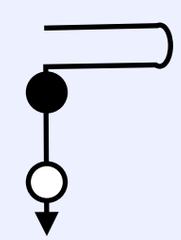
$= iG^M(\tau-\tau')$ 

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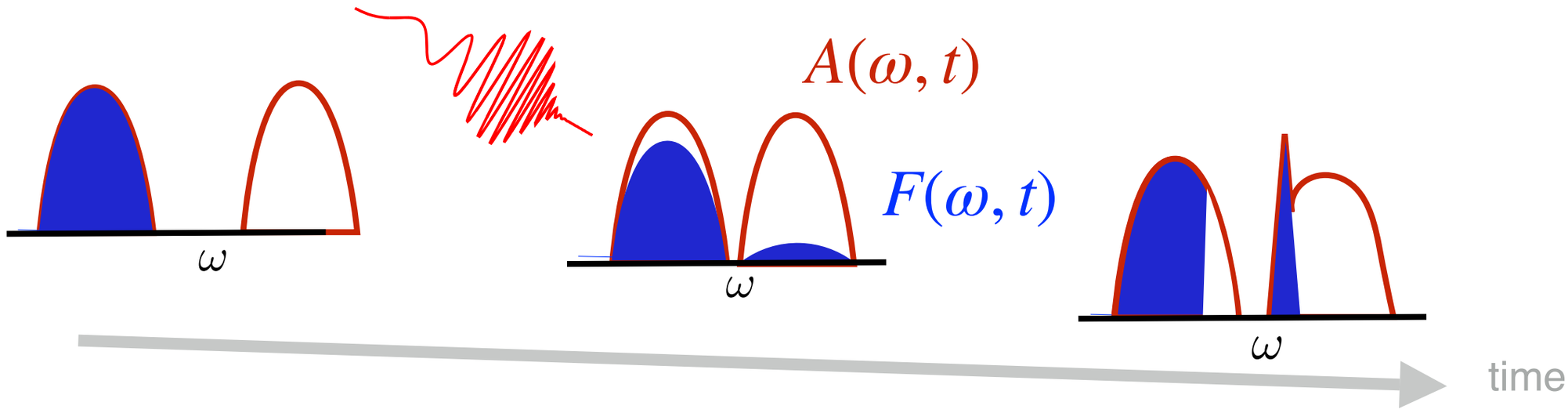
- real time $G(t,t')$ parametrized by two independent functions:

$G^R(t,t') \leftrightarrow$ spectrum $A(\omega,t)$

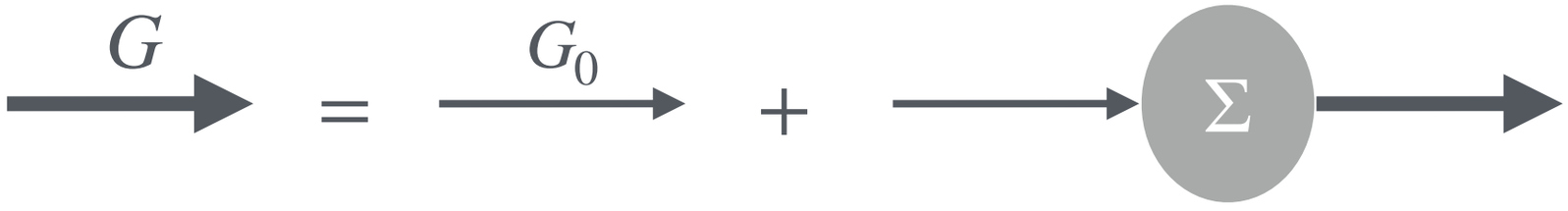
$G^<(t,t') \leftrightarrow$ non-universal distribution $G^<(\omega,t) = 2\pi i A(\omega,t) F(\omega,t)$

⇨ Keldysh formalism: Equations for contour-ordered $G(t,t')$
≡ coupled equations for time-dependent spectrum and occupation



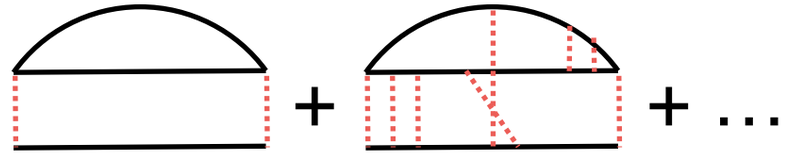
Perturbation theory

Derivation of perturbation theory for $\mathscr{C}$-ordered Green's functions analogous to imag time ordered Green's functions



$$G(t,t') = \underbrace{G_0(t,t')} + \int_{\mathscr{C}} dt_1 dt_2 G_0(t,t_1)\underbrace{\Sigma(t_1,t_2)}G(t_2,t')$$

noninteracting GF (includes dynamics due to external fields) and time-dependent mean-field potentials



Same rules in diagrammatic perturbation theory:

Diagrams on imag part on $\mathscr{C} \rightarrow$ diagrams for Matsubara Green's functions

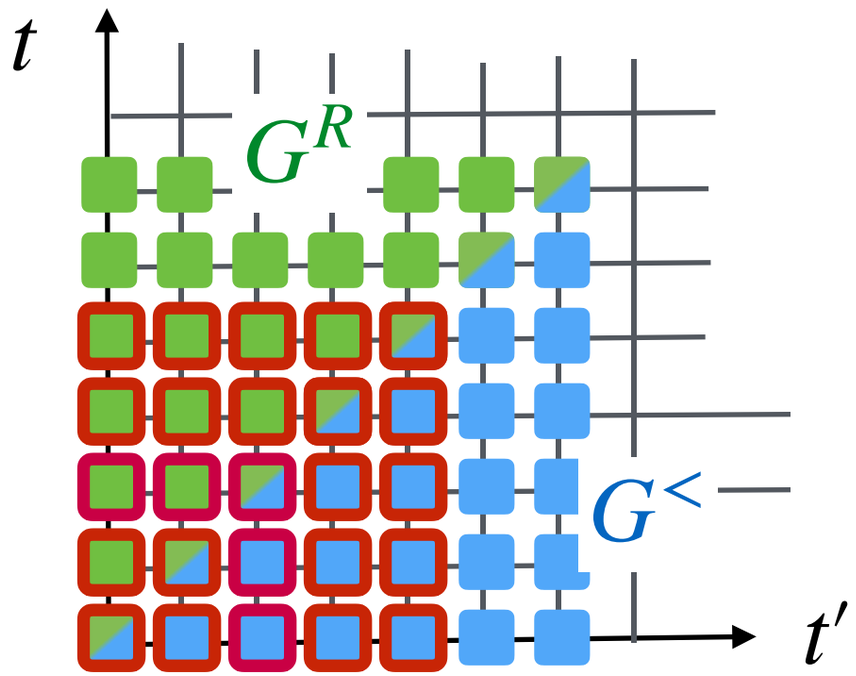
when $t \rightarrow -i\tau,\ i\int_{\mathscr{C}} dt \rightarrow \int_0^\beta d\tau,\ G(-i\tau,-i\tau') \rightarrow iG^M(\tau-\tau')$

Solution of real-time Dyson equation

Give $G_0(t,t')$, determine $G(t,t')$ from

$$G(t,t') = G_0(t,t') + \int_{\mathscr{C}} dt_1 dt_2\, G_0(t,t_1)\,\Sigma[G,G_0](t_1,t_2)\,G(t_2,t') \quad ?$$

Causality: Solution possible via “timestepping”



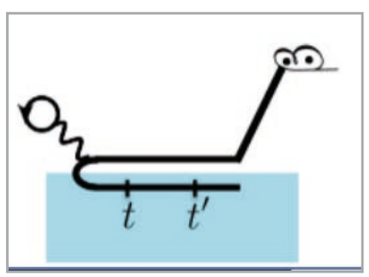
mixed components $G(-i\tau,t)$
not shown for simplicity

Update on time-slice based on previous times (and on initial state Matsubara Green’s function)

Dyson equation mapped to causal integral equastions:
“Kadanoff Baym equations”

Implementation:

Schüler et al. Computer Phys. Comm. **257**, 107484 (2020)



NESSI v1.1.2

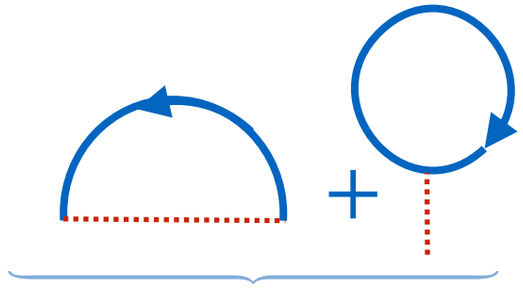
The NonEquilibrium Systems SImulation Library

http://www.nessi.tuxfamily.org/

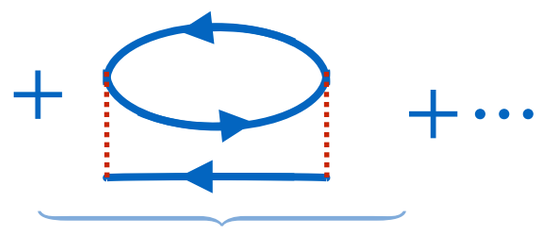
Illustration: Melting of excitonic insulator

- Electron-electron scattering

$$\Sigma(t,t') =$$



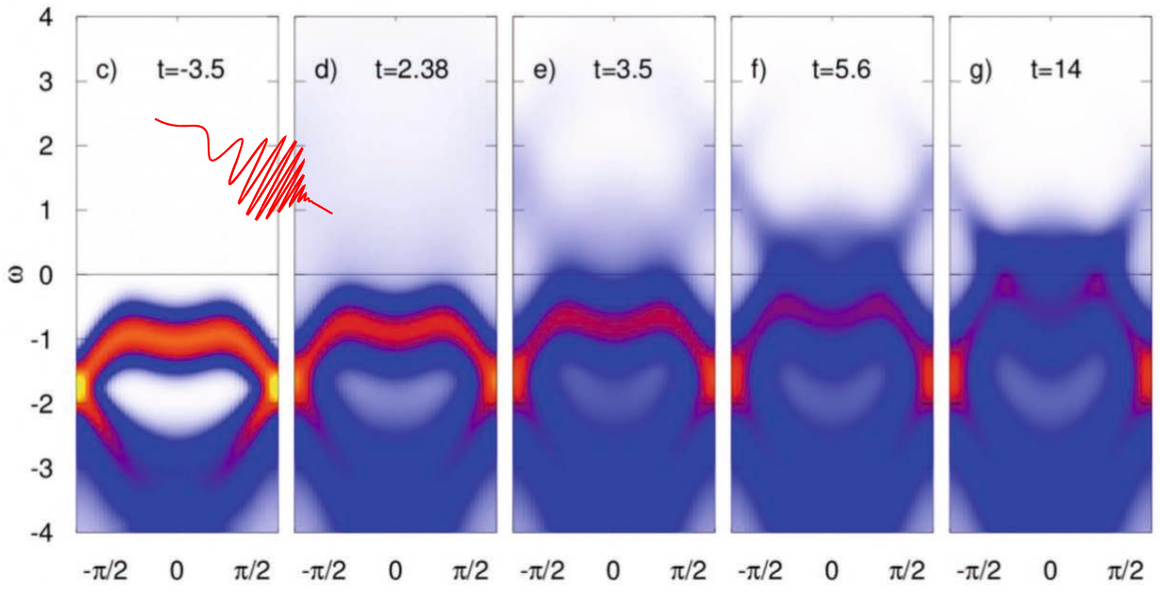
t-d. mean-field:



el-el. scattering

E.g. Melting of exciton insulator

(photoemission $\sim G_k^<(\omega,t)$)



Golež et al. PRB **94**, 035121 (2016)

Equations for contour-ordered Green's function in (t, t')

≡ coupled equations for time-dependent spectrum and occupation

Comment on self-consistency

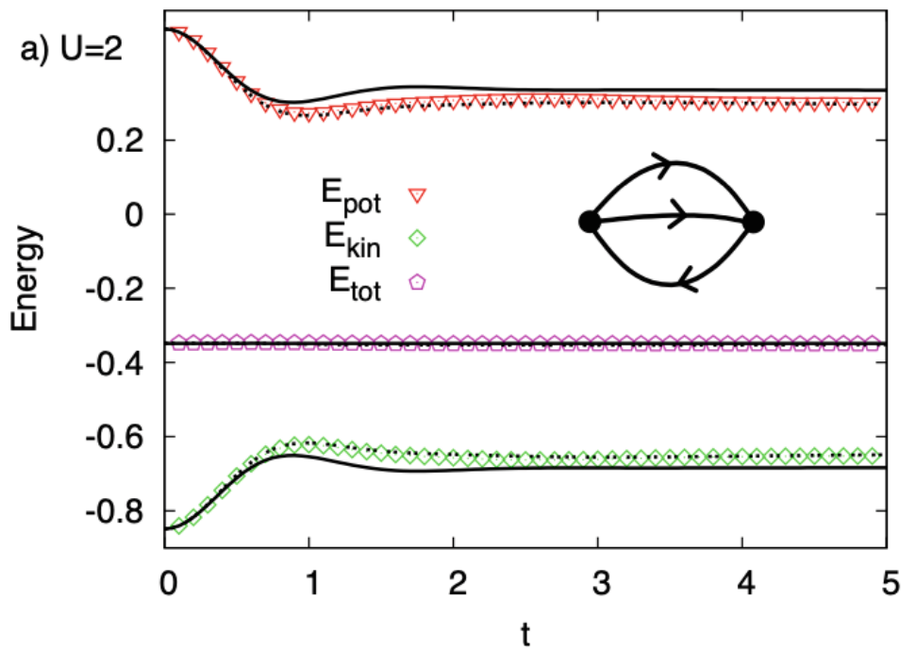
$$H = J \sum_{\langle i,j\rangle,\sigma} c^{\dagger}_{i,\sigma} c_{j,\sigma} + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$

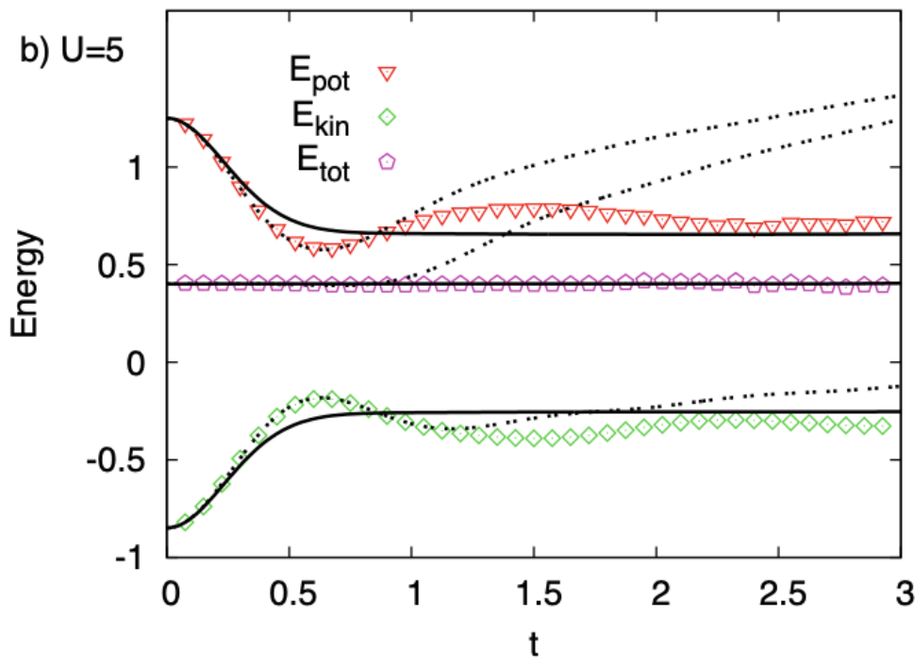
Interaction quench (sudden switch-on of interaction) starting from Fermi gas (U=0)

Second order perturbation theory ($Z \to \infty$ Bethe lattice)

$$\Sigma(t,t') = U(t)U(t')G(t,t')G(t,t')G(t't)$$

Check of energy conservation:





Solid lines: Self-consistent perturbation theory ("conserving")

"IPT" $G \to \mathscr{G}_0$

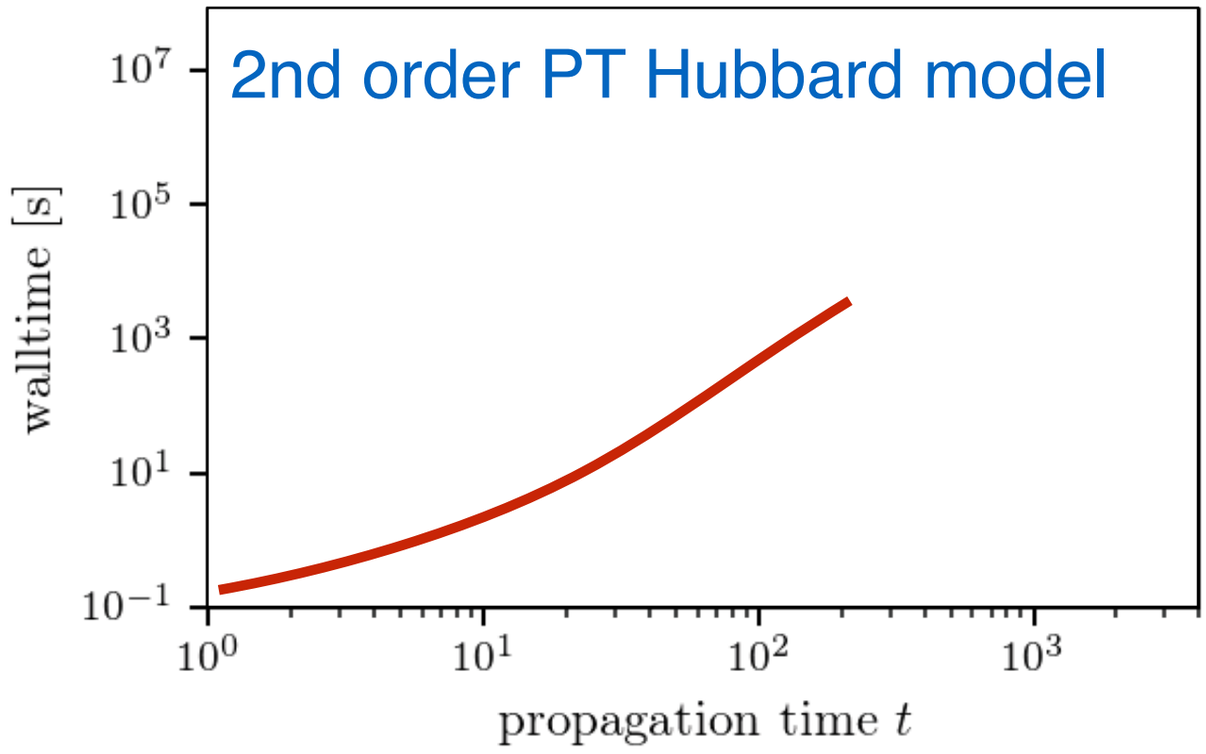
Computational cost / Memory bottleneck

Scaling of numerical cost:

- CPU $\mathcal{O}(N_t^3)$
- memory $\mathcal{O}(N_t^2)$

(OMP parallelisation possible)

more severe …



E.g.: First-principle **multi**-orbital simulations based on standard perturbative approaches?

$L = 10$ orbitals
Energy window 10eV
⇨ $\Delta t \ll 1/\text{eV} = 0.1\text{fs}$)
Simulation time 1000fs

⇨

$n_t = 10^4$
Memory $G^<$ and G^R
$n_t^2 \times L^2 = (10^4)^2 \times (10^2) = 10^{10}$
complex numbers

not entirely out of range, but definitely some improvement needed!

Memory bottleneck

Overcoming the memory constraint? … needed for

⇨ simulations with vastly different timescales: electrons & lattice; electrons & collective modes; prethermalization & thermalisation

⇨ first-principle **multi**-orbital simulations based on standard perturbative approaches

- Generalized Kadanoff Baym Ansatz

  Schlünzen, Joost, Bonitz, Phys. Rev. Lett. 124, 076601 (2020)

- Quantum Boltzmann equations

  Picano, Li, Eckstein, Phys. Rev. B **104**, 085108 (2021)

} additional approximations "physical insight"

- Systematic truncation of memory integrals

  Stahl, Dasari Picano, Li, Werner, Eckstein, PRB **105**, 115146 (2022)

- Hierarchical storage of two-time functions

  Kaye and Golez,arXiv:2010.06511

} Reformulation of numerical solutiuoin

DMFT

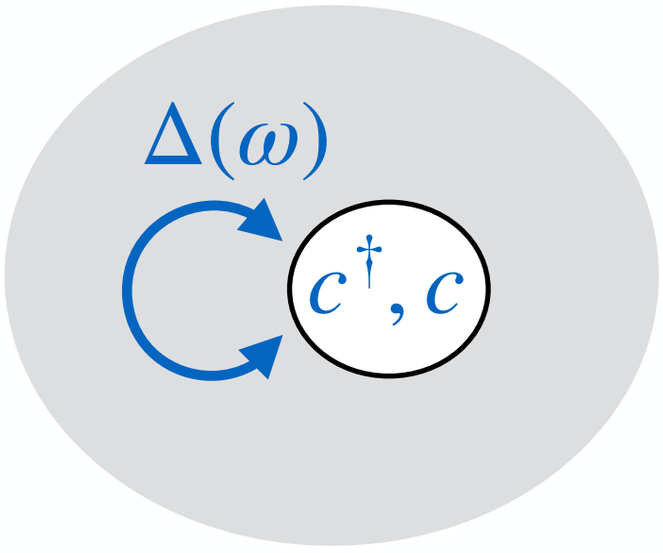
Dynamical mean-field theory

- Best approximation for local self-energy, exact in $d = \infty$

$$G_k = G_{0,k} + G_{0,k} * \Sigma * G_k$$

Metzner & Vollhardt 1989;
Georges & Kotliar 1992;
Georges et al. RMP 1996

- Local (momentum-independent) $\Sigma[G]$ from auxiliary impurity model:



Δ: bath propagator of continuous environment

$$S_{imp} = S_{loc} - \int_0^\beta d\tau d\tau'\, c^*(\tau)\Delta(\tau - \tau')c(\tau)$$

$$\Rightarrow\ G_{imp} = \mathcal{G}_{imp}[\Delta] + \mathcal{G}_{imp}[\Delta] * \Sigma * G_{imp} \overset{!}{=} \sum_k G_k$$

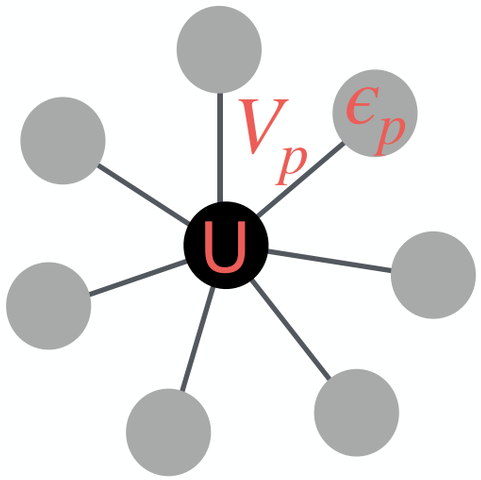
- Non-equilibrium formulation of DMFT

Monien et al 2002; Freericks et al, 2006
Aoki et al., Rev.Mod.Phys. 2014

Non-equilibrium formulation of DMFT

Biggest issue: Solution of the non-equilibrium Quantum impurity model

Equivalent to a time-dependent Anderson model



$$H_{imp} = H_{loc} + \sum_{p,\sigma} \epsilon_p a^\dagger_{p\sigma} a_{p\sigma} + \sum_{p,\sigma} \left(V_p(t) c^\dagger_\sigma a_{p,\sigma} + h.c. \right)$$

Solution: (no multi-purpose approach like in equilibrium yet)

- Quantum Monte Carlo (dynamical sign problem)
- matrix product states (MPS) Wolf, Schollwöck, et al. 2014
  (efficient bath representation, entanglement)
- strong-coupling expansion (hybridizaion expansion)
- weak coupling expansions (“IPT”)

Solution of the Anderson model: Hybridization expansion

$$\langle O(t)\rangle = \pm\frac{1}{Z}\mathrm{tr}\left[\rho\, U(t_0,t) O U(t,t_0)\right]$$

- Unfolding the contour:

$$\mathrm{tr}\Big[\quad t_{max}^{-} \longrightarrow 0 \longrightarrow -i\beta \longrightarrow t_{max}^{+}\quad\Big]$$

- Expansion of action in $\int_{\mathscr{C}} dtdt' c^*(t)\Delta(t,t')c(t')$ (talk by Ph. Werner)

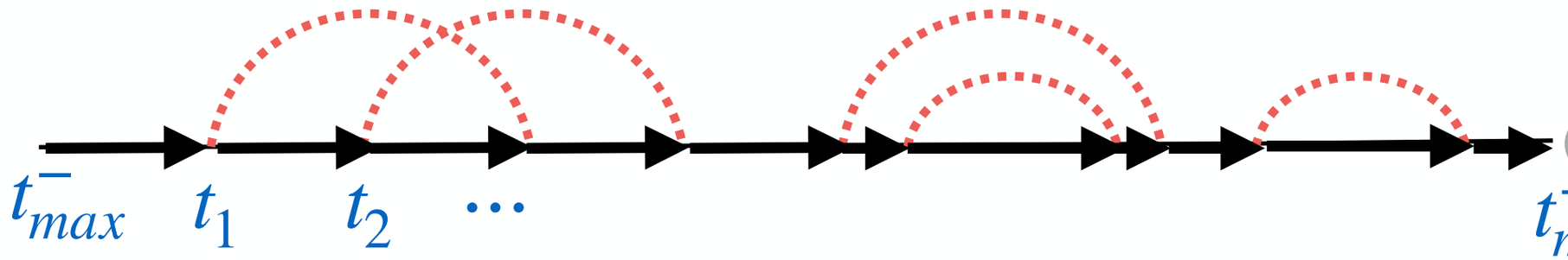
$$= \frac{1}{Z}\int d\{t_j\}\,\mathrm{tr}_{loc}\Big[\quad t_{max}^{-} \; t_1 \; t_2 \; \cdots \; t_{max}^{+}\quad\Big]$$

$$= \frac{1}{Z}\int d\{t_j\}\left(\prod \Delta(t_i,t_j)\right)\mathrm{tr}\left[\mathscr{G}(t_{max},t_1)c\mathscr{G}(t_1,t_2)\cdots O\right]$$

$$\longrightarrow \quad = \mathscr{G}_{n,m}(t,t') = \langle n|\, T_{\mathscr{C}} e^{-i\int_t^{t'} ds H_{loc}(s)}\,|m\rangle$$

Solution of the Anderson model: Hybridization expansion

$$\frac{1}{Z}\int d\{t_j\}\ \mathrm{tr}\Big[$$



$$\Big]$$

Monte Carlo summation of all configurations (CTQMC-Hyb on $\mathscr{C}$):

Werner et al. (2006), Werner, Oka, Millis (2009)

$$\langle O\rangle = \sum_c w_c O_c = \frac{\sum_c |w_c| s_c O_c}{\sum_c |w_c| s_c}$$

dynamical sign problem
($\mathscr{G}, \Delta, \ldots w_c \in \mathbb{C}$)

Note: $Z = \sum_c w_c$ can depend only on Matsubara branch

Quench in the Hubbard model

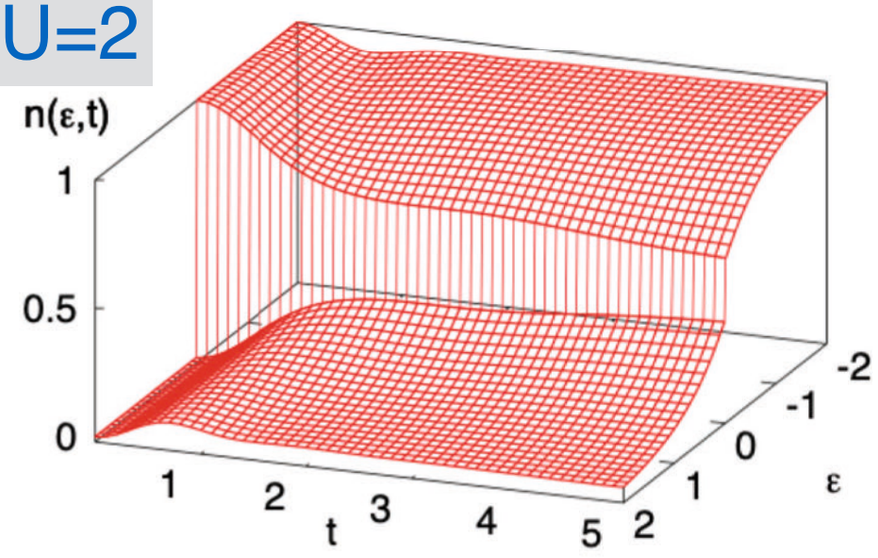
ME, Kollar, Werner (2009)

$$H = J \sum_{\langle i,j \rangle,\sigma} c^{\dagger}_{i,\sigma} c_{j,\sigma} + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$

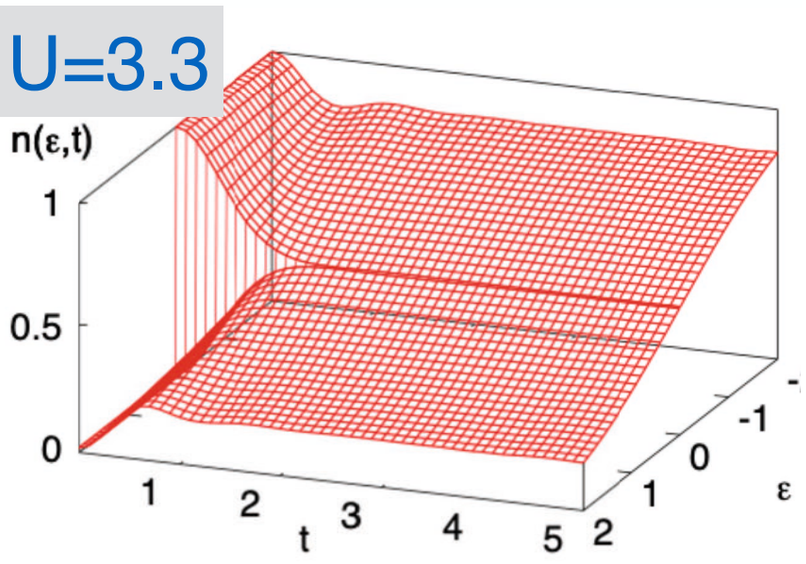
Interaction quench (sudden switch-on of interaction) starting from Fermi gas (U=0)

Relaxation of momentum distribution $n_k(t) = \langle c^{\dagger}_k c_k \rangle$ (bandwidth=4)

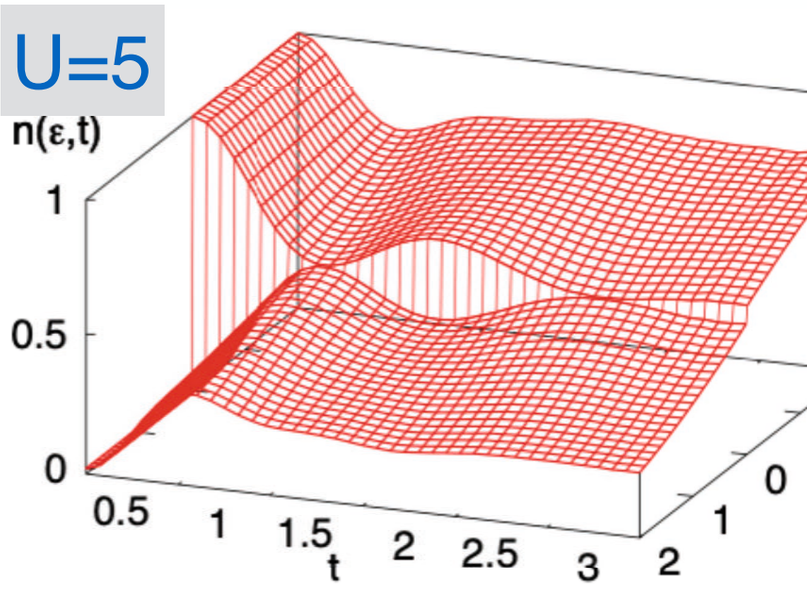
U=2



U=3.3



U=5



"prethermalization"

Moeckel & Kehrein (2008)

Thermalization at DPT

Collapse and revival

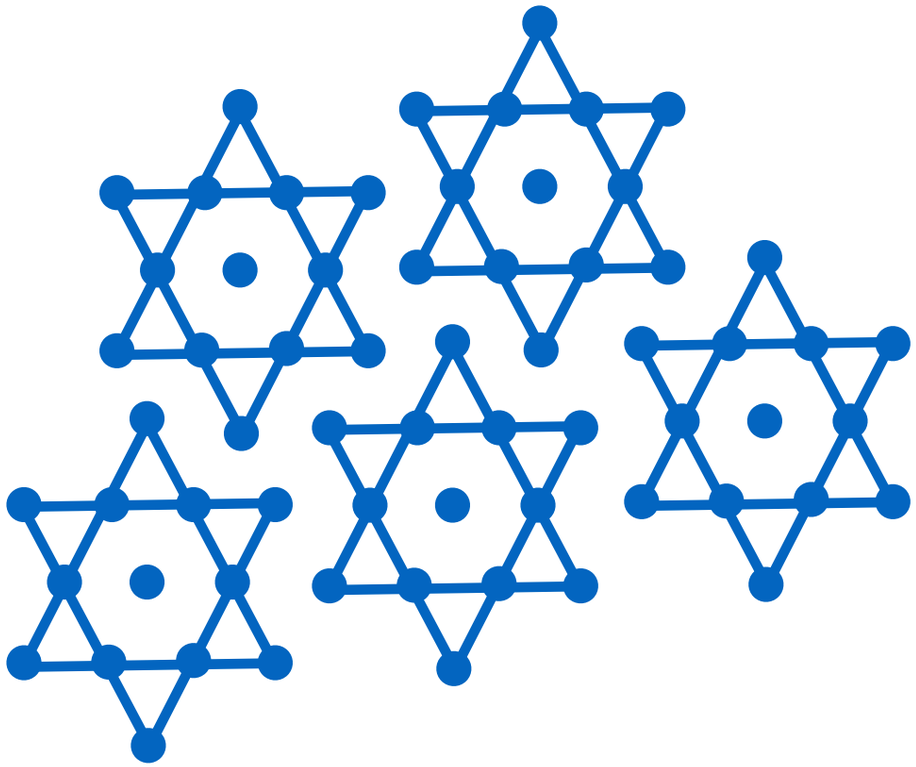
Interesting physics, but only few hopping times accessible

Photo-doping a Mott insulator

Photo-doping: Ultrafast dynamics in TaS_2

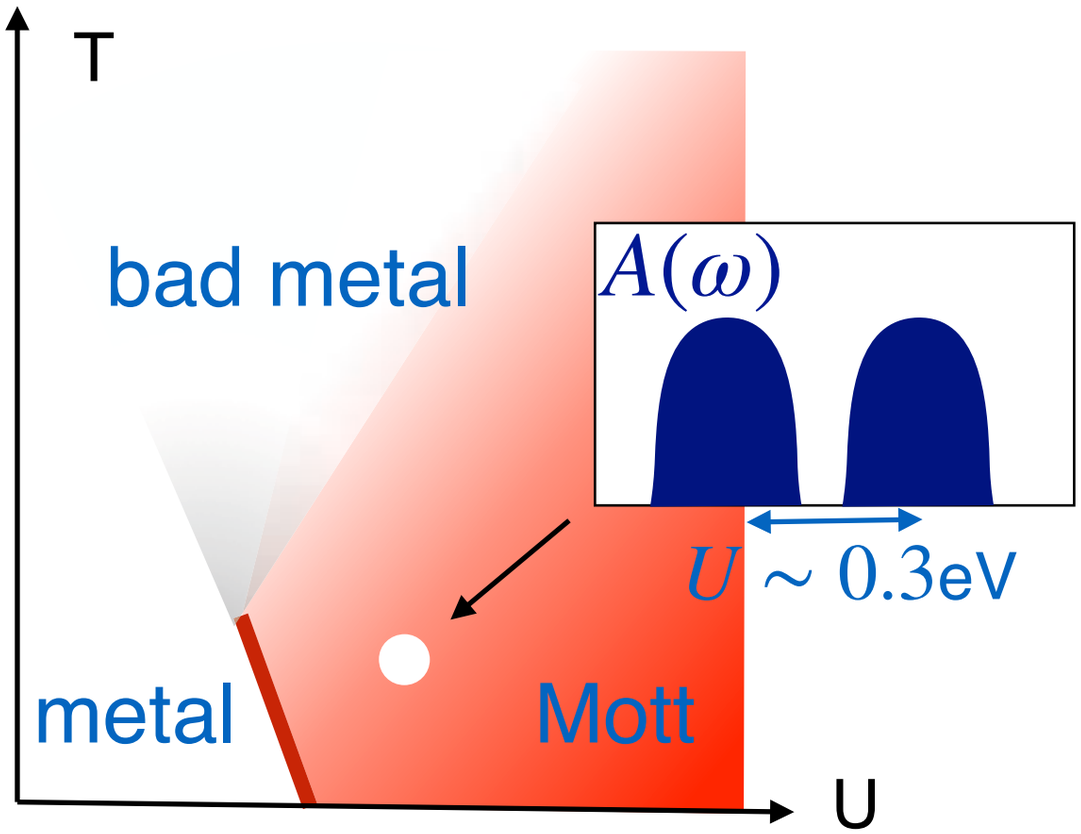
Low temperature state of TaS_2:

13 atom CDW reconstruction



Leaves half-filled valence orbital per cluster

Mott state described by single band Hubbard Model (?)

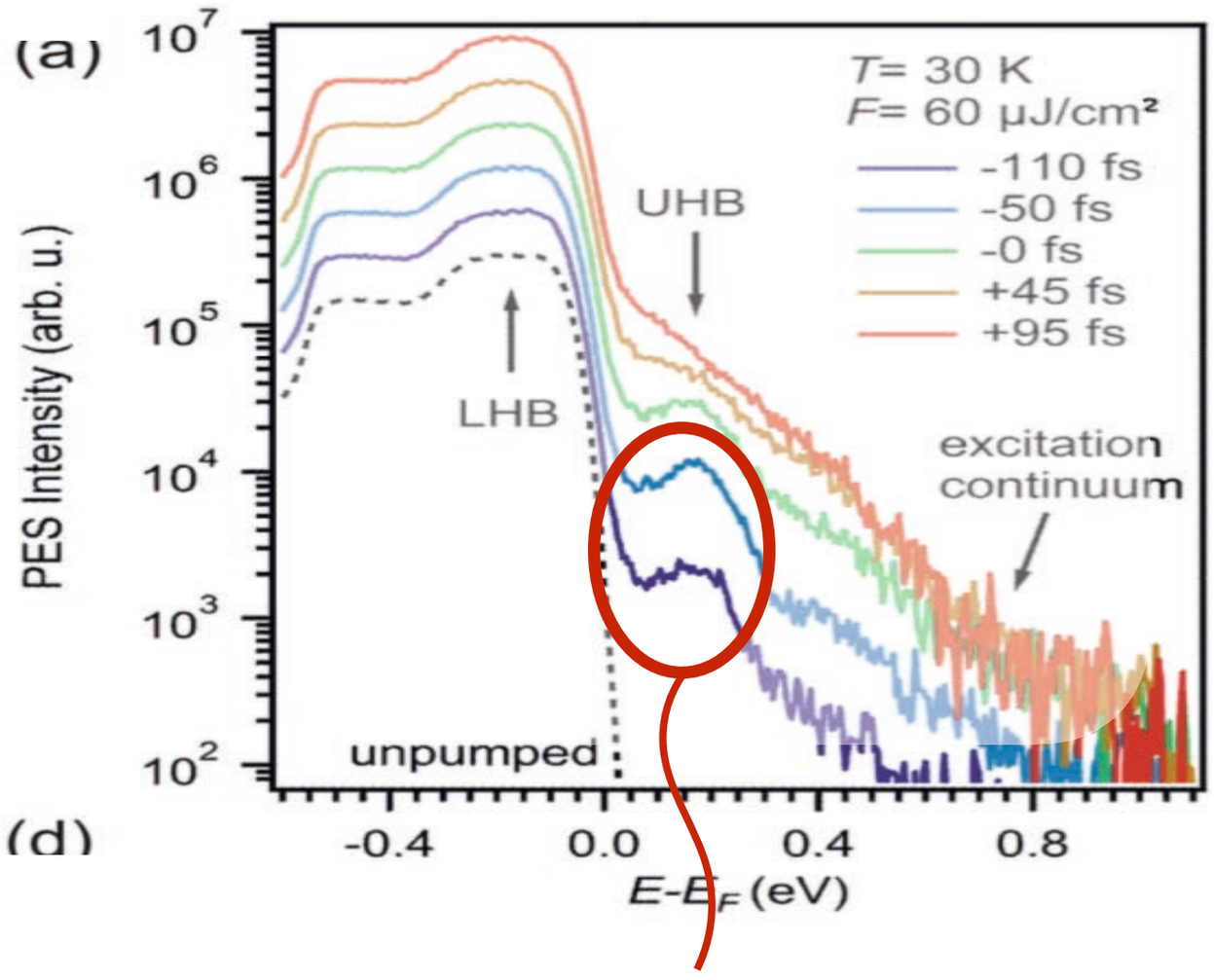


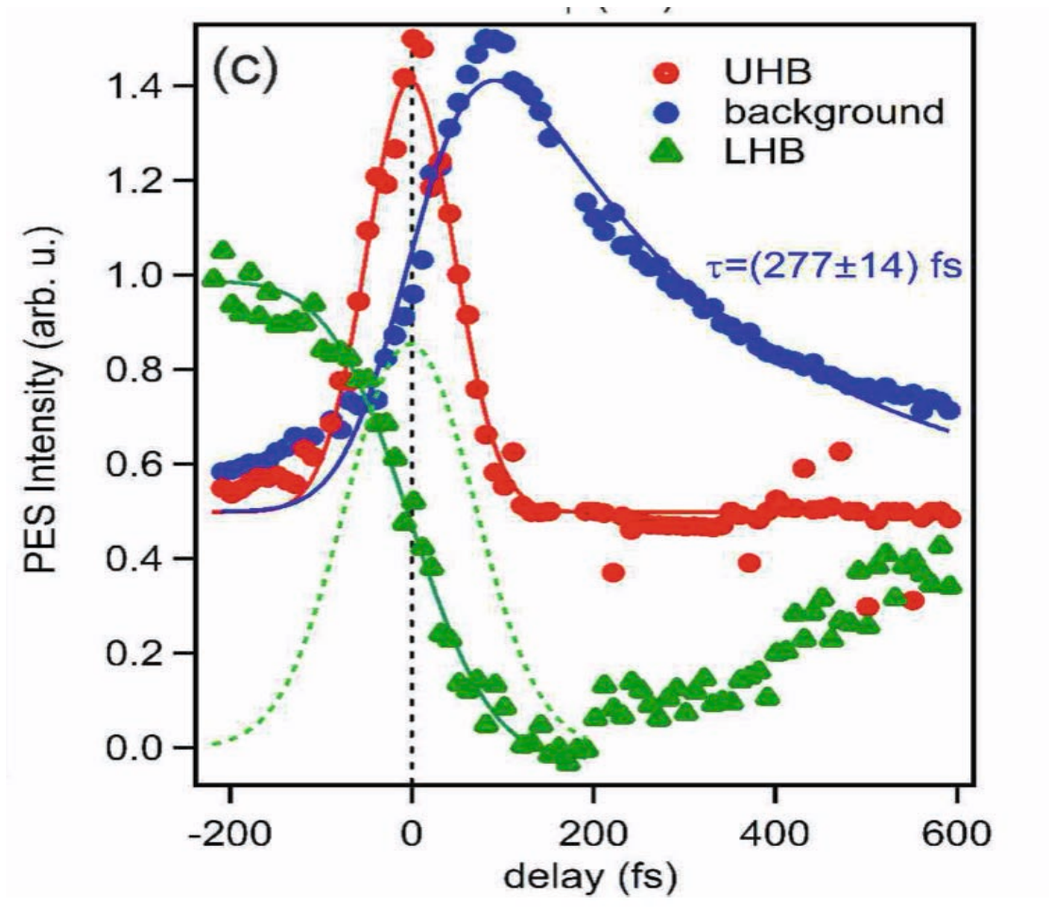
(AFM order suppressed on triangular lattice)

Photo-doping: Ultrafast dynamics in TaS_2

Photo-excitation: Time resolved ARPES (at Γ) after 1.5eV excitation

Ligges et al., Phys. Rev. Lett. **120**, 166401 (2018)



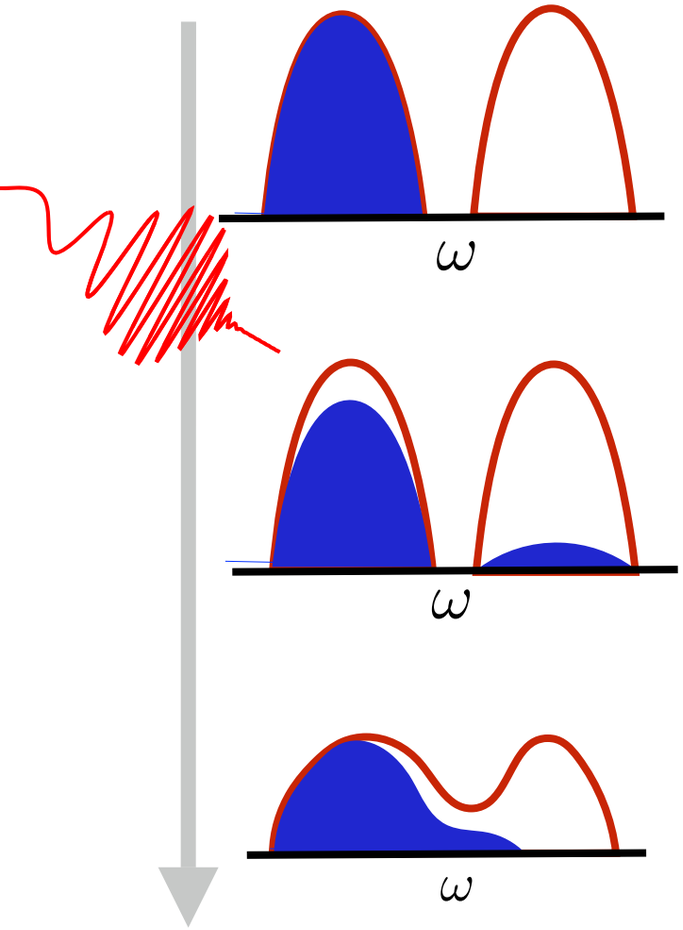


Transient appearance of UHB … disappears within $\lesssim 100$ fs

Earlier Experiments: Perfetti et al., Phys. Rev. Lett. **97**, 067402 (2006)

Photo-doping: Ultrafast dynamics in TaS_2

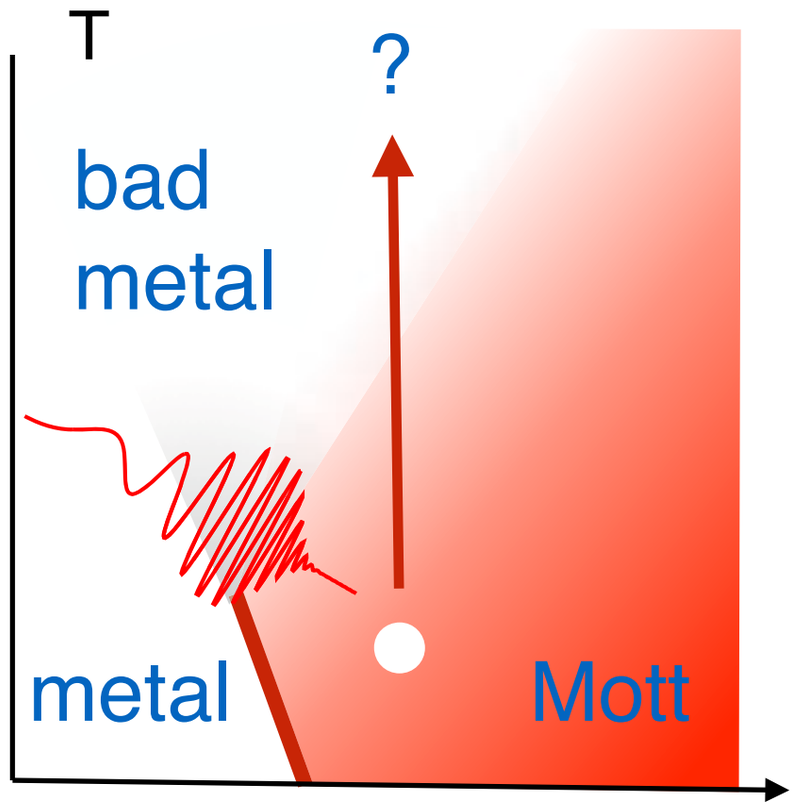
Scenario:



Small gap Mott insulator

Non-thermal state with occupation in Hubbard band

Thermalization to bad metal



⇨ Thermalization time in small gap Mott insulator and bad metal?

⇨ Spectral signature of transient and final photo-excited state?

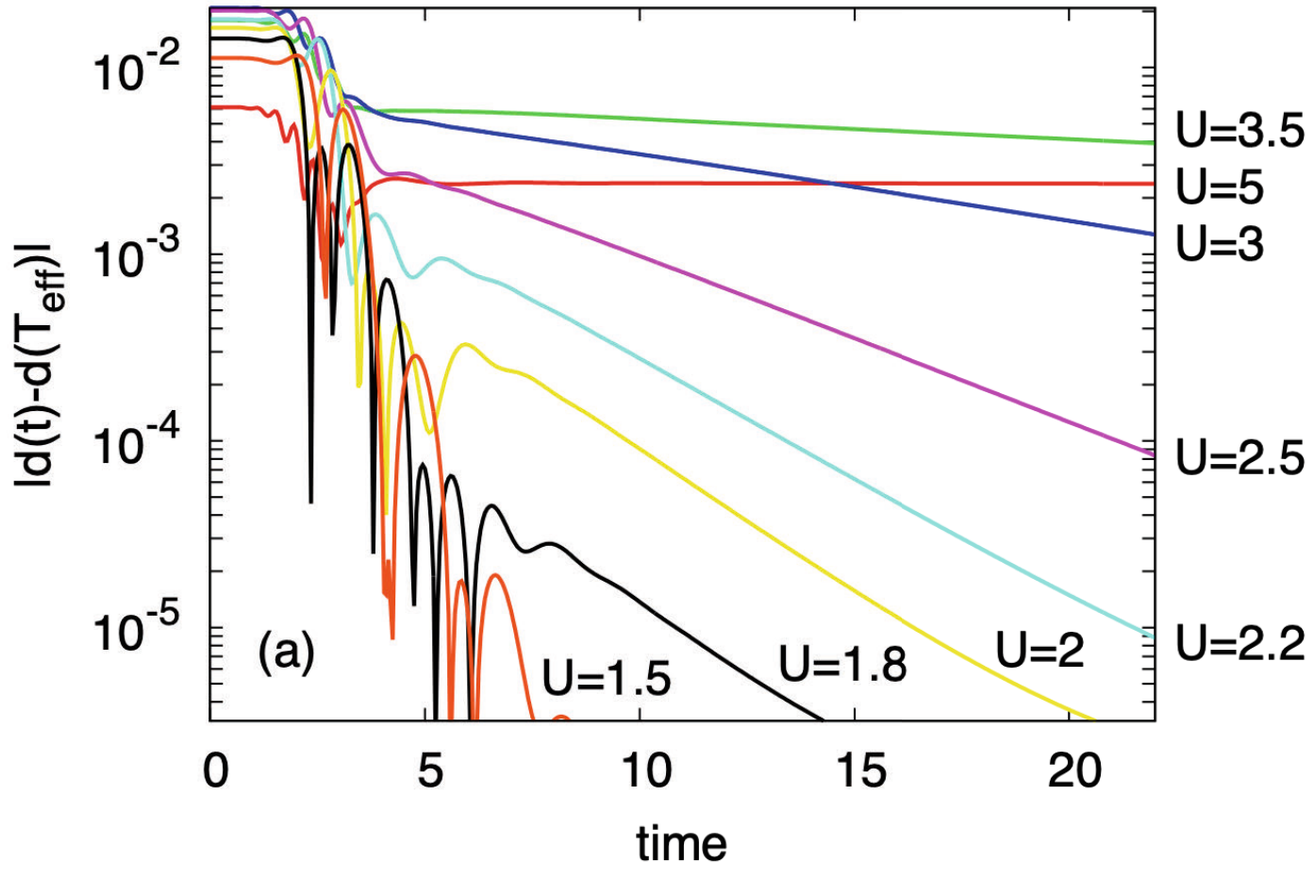
Thermalization of a photo-excited Mott insulator

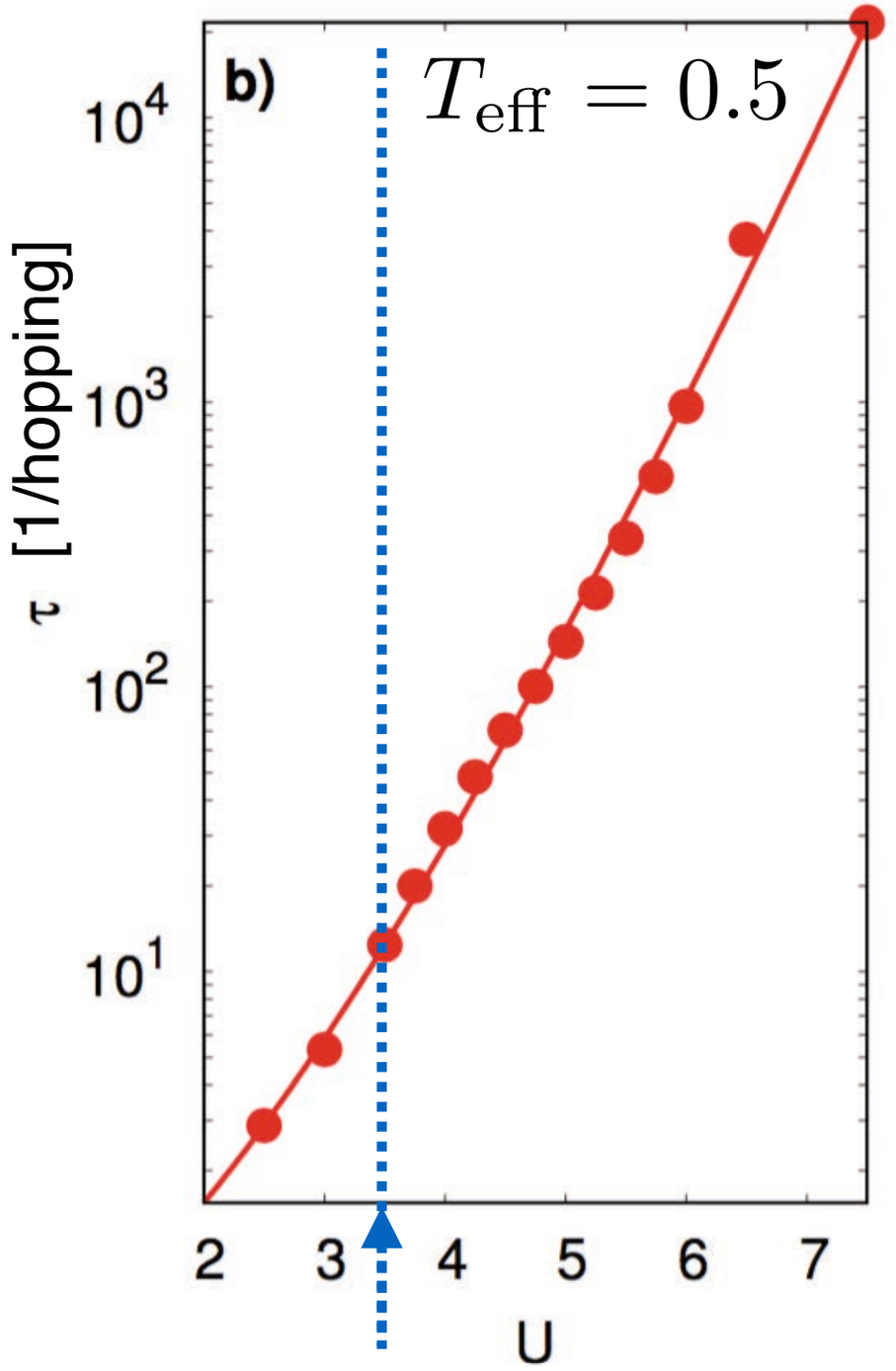
Simulation for a hypercubic lattice
(Metal insulator transition at $U_{MIT} \approx 3.1$)

double occupancy $\langle n_\uparrow n_\downarrow \rangle$ after excitation:

$$d(t) \sim d_{eq}(T_{eff}) + Ae^{-t/\tau}$$

determined by total energy





Mott transition at T=0

Eckstein & Werner, PRB **84**, 035122 (2010)

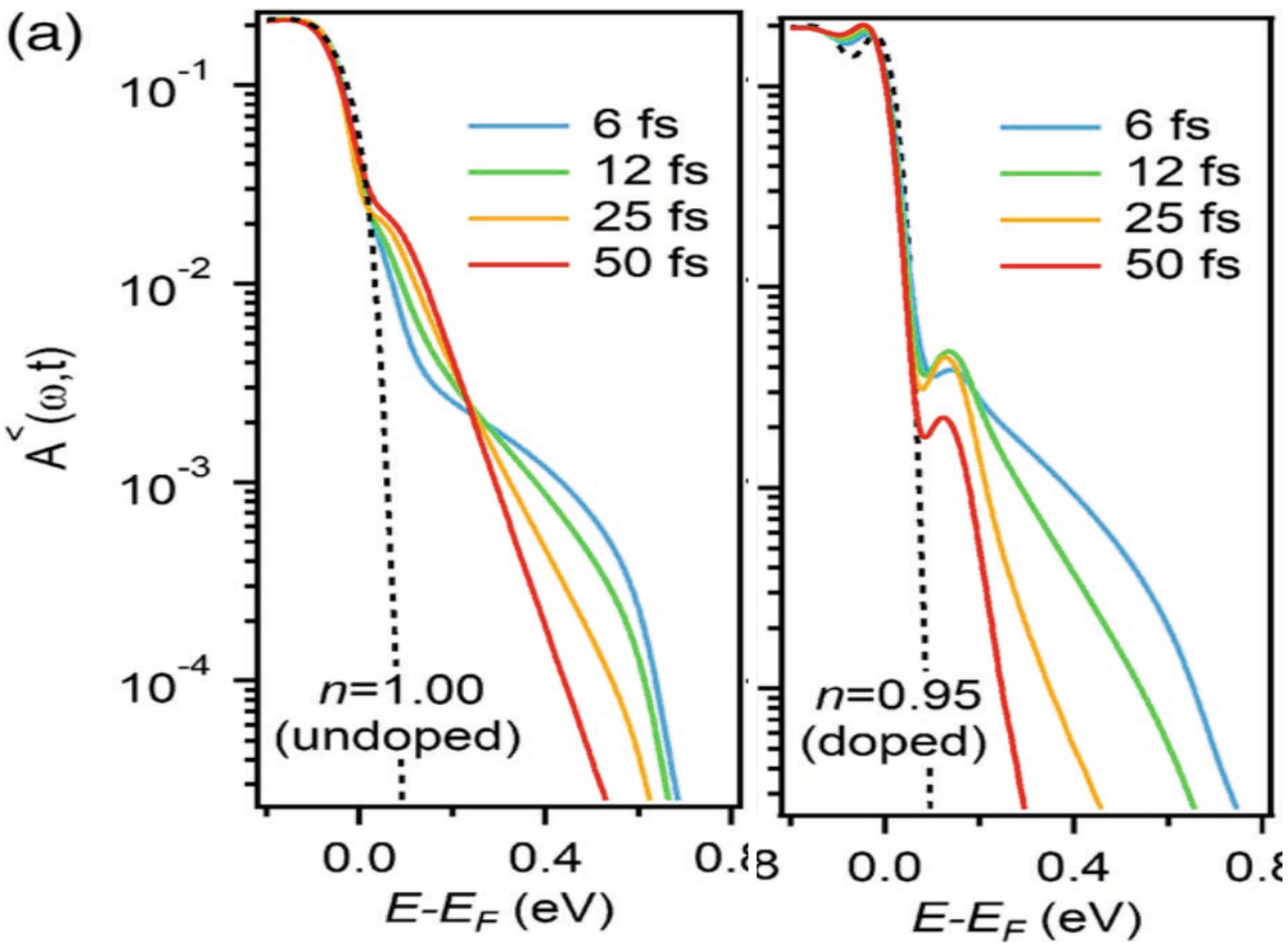
Exponentially slow thermalization for larger Mott gap

Ultra-fast dynamics in TaS_2

Ligges et al., Phys. Rev. Lett. **120**, 166401 (2018)

Simulation for triangular Hubbard model

$$A^<(\omega,t) = \frac{1}{2\pi i}\int ds\, G^<(t+s/2, t-s/2)e^{i\omega s}$$



Thermalization in half-filled model too slow to explain disappearance of UHB

consistent with electronic thermalization in slightly doped regime

Good benchmark for DMFT: Qualitative interpretation of features, reasonable timescales … but too simple model to describe TaS_2

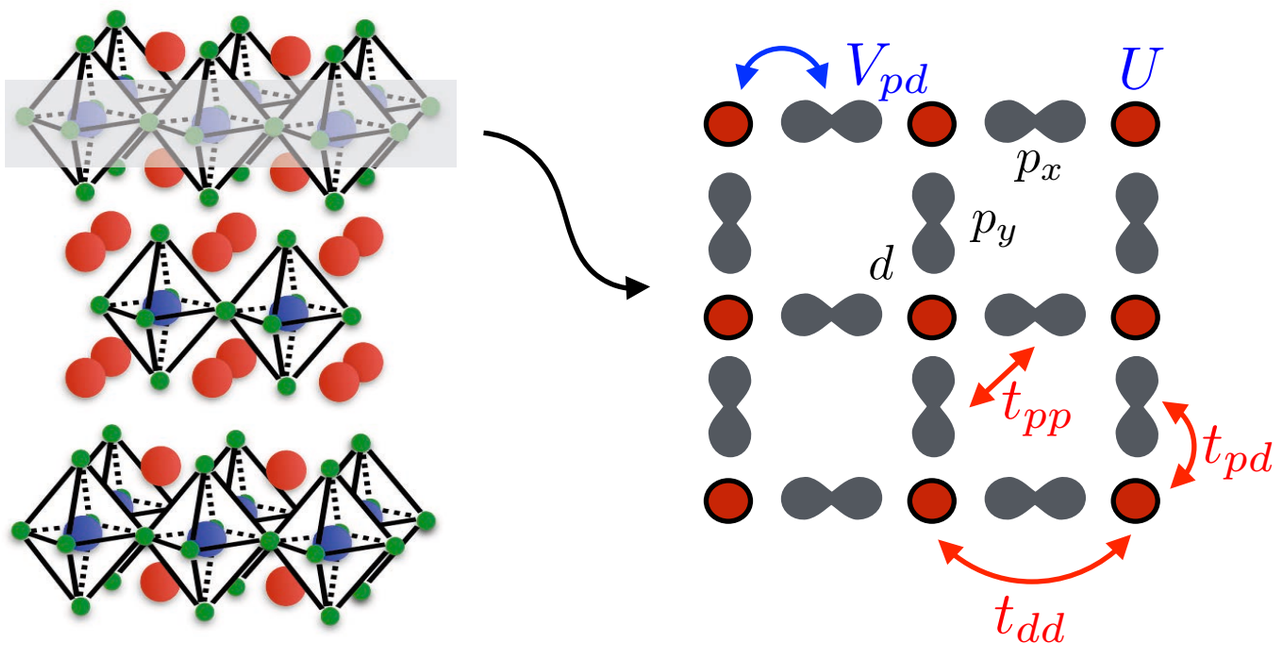
Photo-doping in a CT insulator

Photo-doping in a charge transfer insulator

Golez, Boenke, Eckstein, Werner,
PRB(R) 2019, PRB 2019

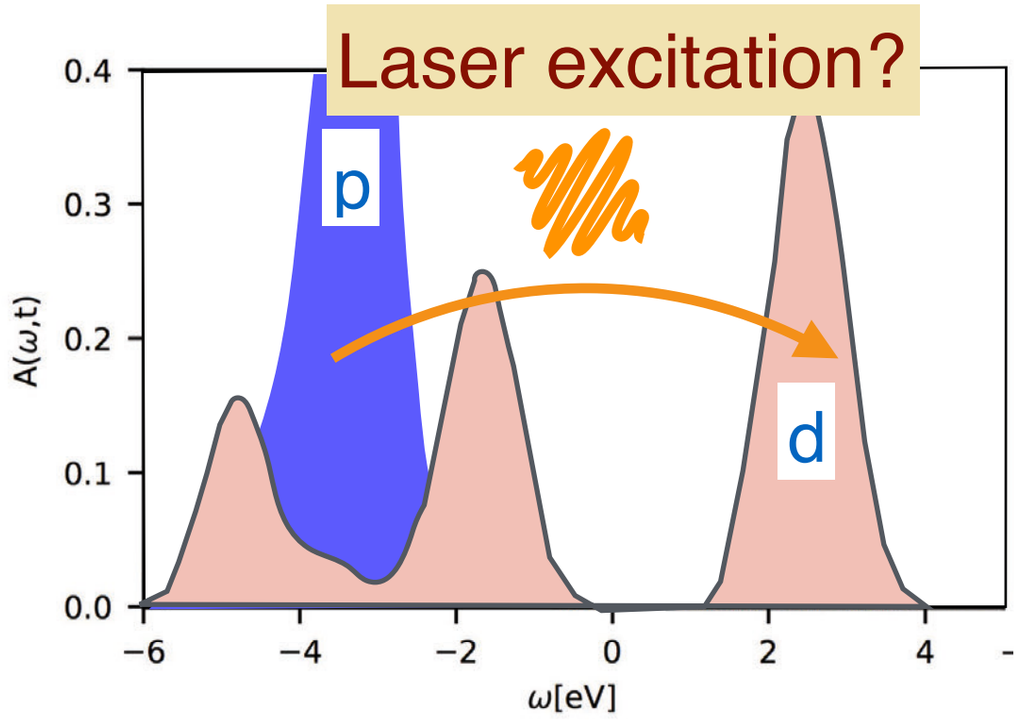
3-band Emery model

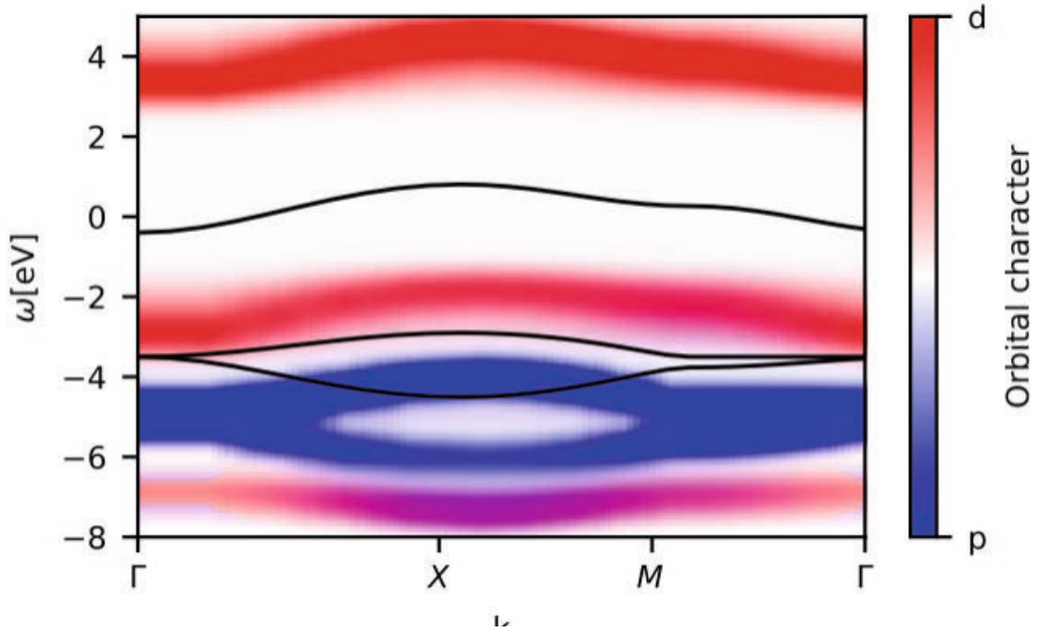
Tight-binding model CuO_2 plane in cuprate



$$U_{dd} = 8\text{eV}, t_{dd} = -0.1\text{eV},$$
$$V_{pd} = 2\text{eV}, \Delta_{pd} = -2\text{eV},$$
$$t_{pd} = 0.4\text{eV},$$

Equilibrium spectral function:

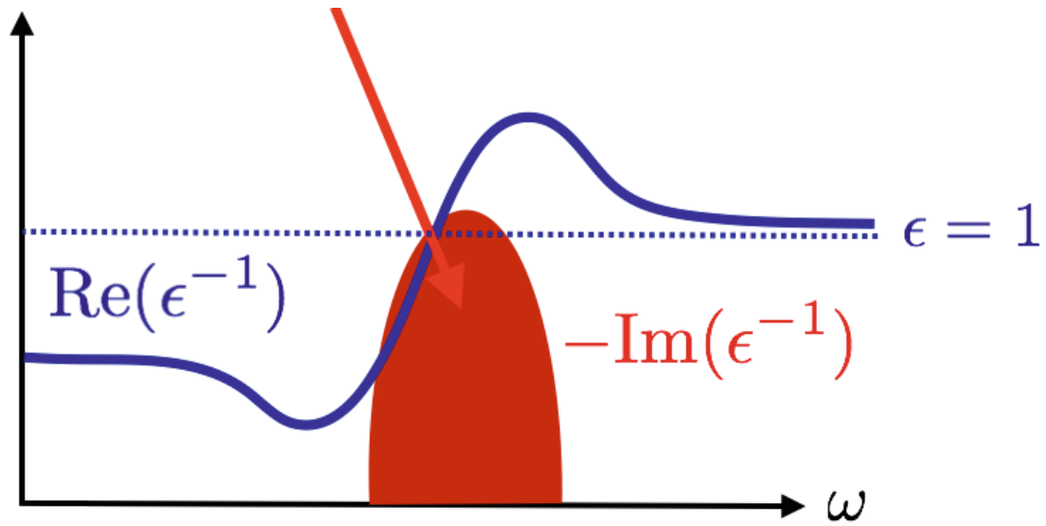




Manipulating electronic structure: The role of screening

$$W_q(\omega) = \frac{V_{bare,q}(\omega)}{\epsilon_q(\omega)}$$

spectrum of polarization modes
(el-hole excitation, plasmons)



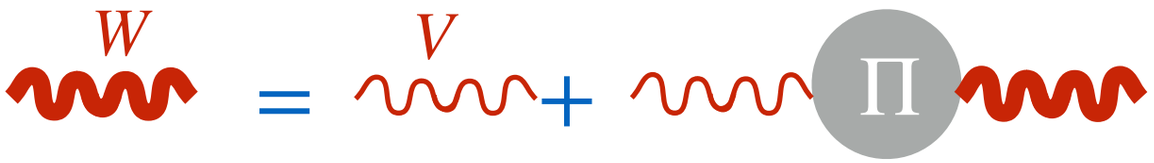
⇨ Needed for any quantitative understanding of the dynamics

⇨ Manipulate screened interactions out of equilibrium?

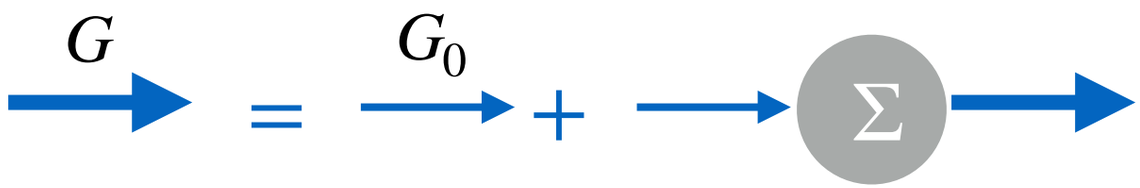
Treatment of electrons with self-consistently screened interaction?

GW+DMFT

- Self-consistent expansion in terms of fully screened interaction



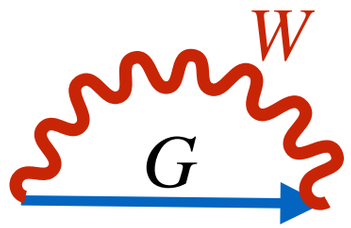
$$\Pi = \Pi[G, W]$$



$$\Sigma = \Sigma[G, W]$$

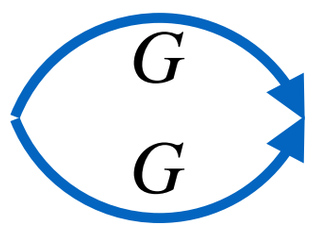
+ Diagrammatic approximation:

$$\Sigma[G, W] =$$



$$+\Sigma_{loc}[G_{loc}, W_{loc}] - d\,.\,c\,.\,.$$

$$\Pi[G, W] =$$



$$+\Pi_{loc}[G_{loc}, W_{loc}] - d\,.\,c\,.$$

GW+DMFT

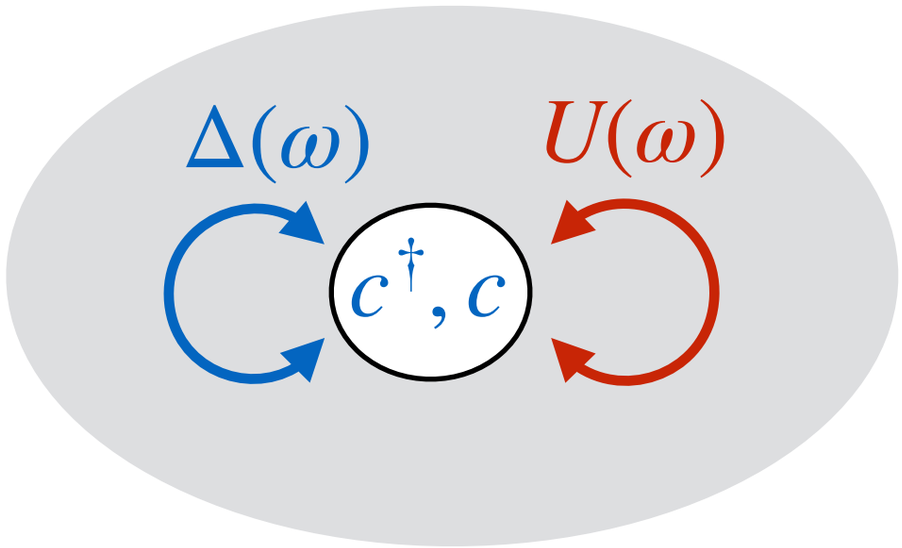
Biermann, Aryasetiawan, Georges PRL 2003

"GW" approximation Hedin 1965

GW+DMFT

- Evaluation of local contributions from auxiliary impurity problem:

  Impurity with self-consistent bath
  and self-consistent interaction $U(\omega)$



$\Rightarrow\ G_{imp},\ \chi_{imp} = \langle n(\tau)n(\tau')\rangle$

DMFT self-consistency for Δ and Σ plus self-consistency of interaction:

$$W_{imp} = U(\omega) + U(\omega)\chi_{imp}U(\omega)$$
$$= U(\omega) + U(\omega)\Pi_{imp}W_{imp}$$

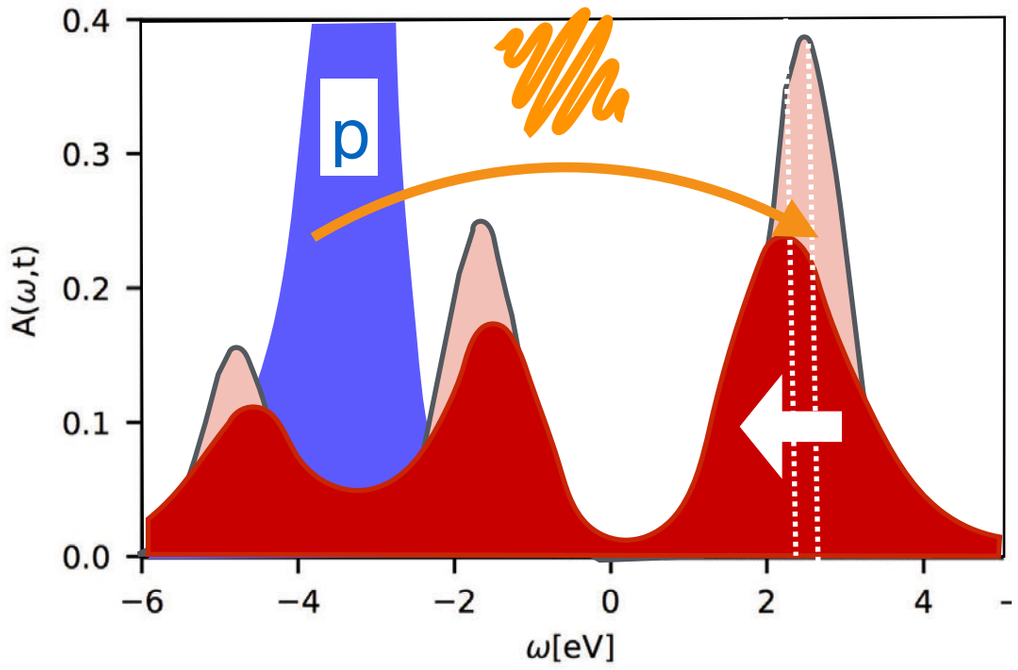
$$W_k = V_k + V_k\Pi_{imp}W_k$$

$$W_{loc} = \sum_k W_k \overset{!}{=} W_{imp}$$

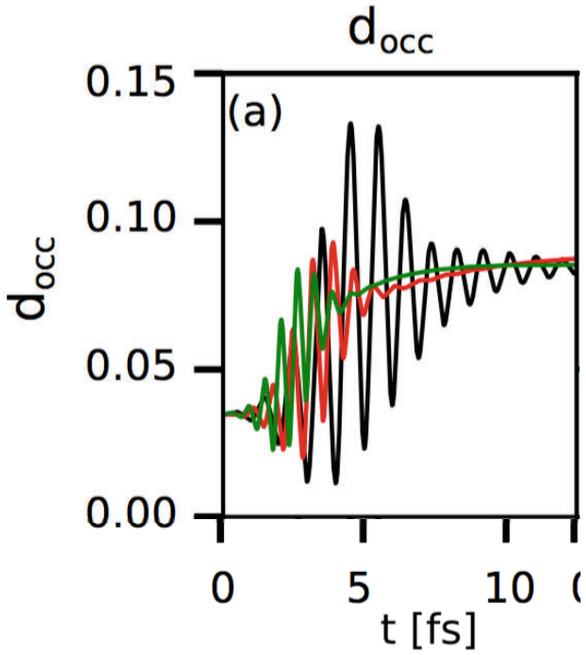
Photo-doping in a charge transfer insulator

Golez, Boenke, Eckstein, Werner,
PRB(R) 2019, PRB 2019

Spectral function:



Laser excitation:



~ 5% charge transfer ⇨ Modification of spectrum:

Band shifts and broadening (basically instantaneous)

Different possible origins:

- Dynamic screening (renormalization of “U”)
- Mean-field shifts due to inter-site interaction (HF)

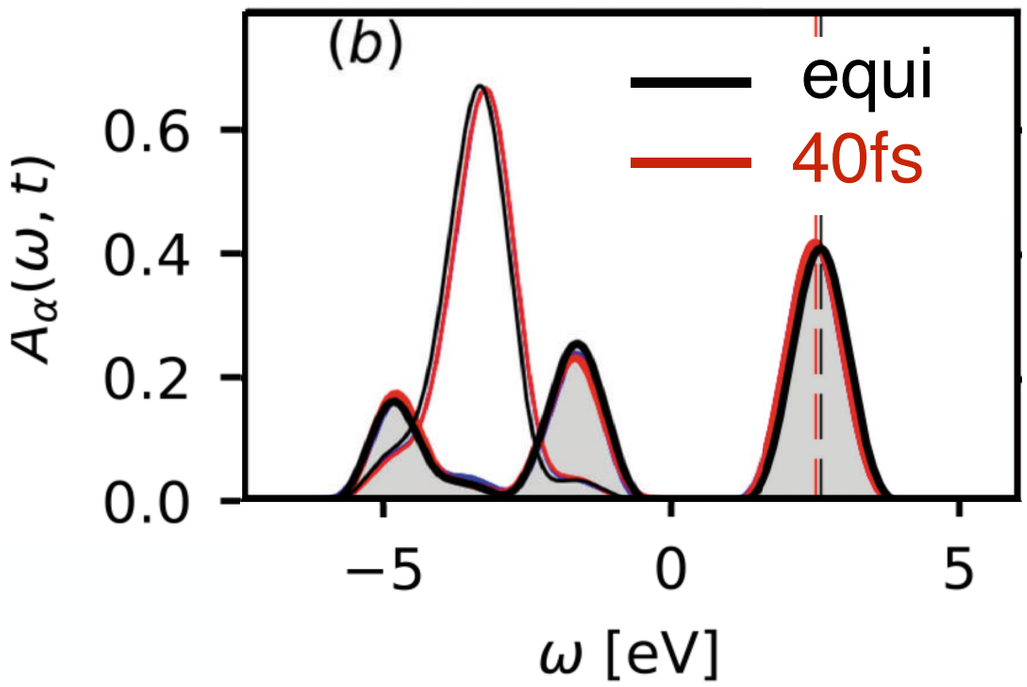
$$H_{pd} \rightarrow V_{pd} n_p \langle n_d \rangle + V_{pd} \langle n_p \rangle n_d + const\,.$$

} Distinguish?

Photo-doping in a charge transfer insulator

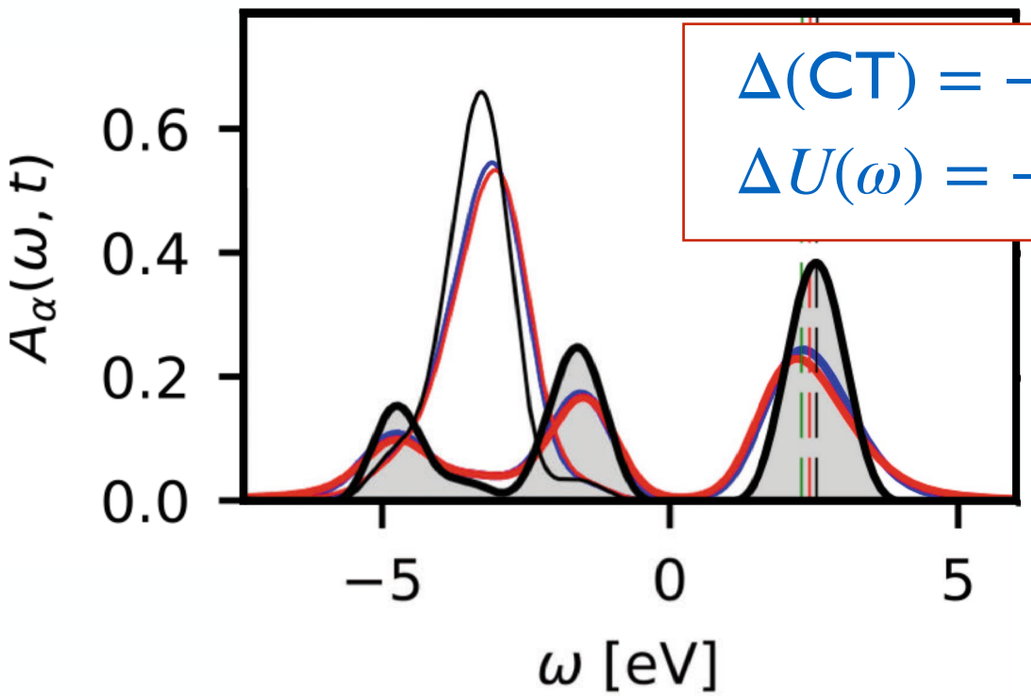
Golez, Boenke, Eckstein, Werner,
PRB(R) 2019, PRB 2019

Only mean-field like shifts (DMFT + Hartree Fock) $\Delta(\mathrm{CT}) = V_{pd}\langle n_p \rangle n_d$



$\Delta(\mathrm{CT}) = -150\mathrm{meV}$

Simulation including screening (GW+DMFT)



$\Delta(\mathrm{CT}) = -150\mathrm{meV}$
$\Delta U(\omega) = -140\mathrm{meV}$

Screening modes

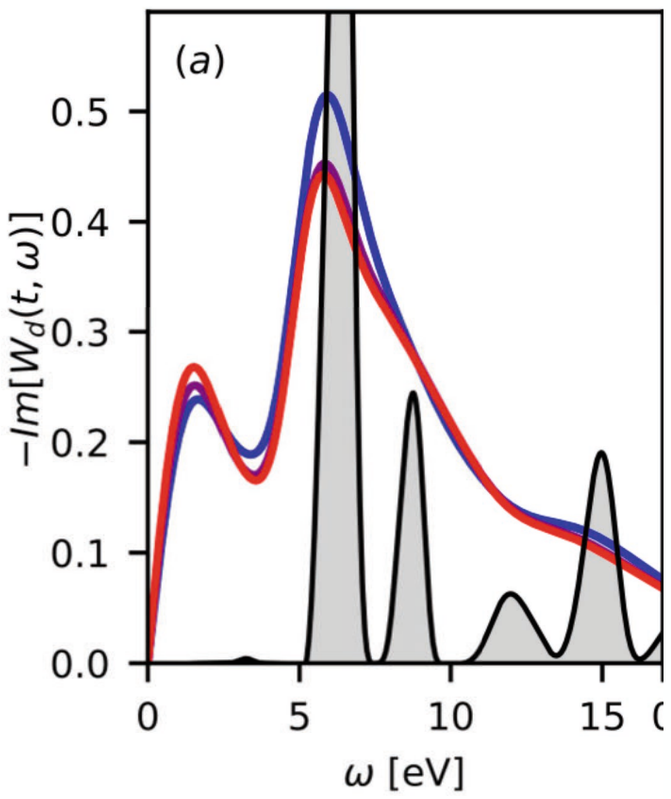
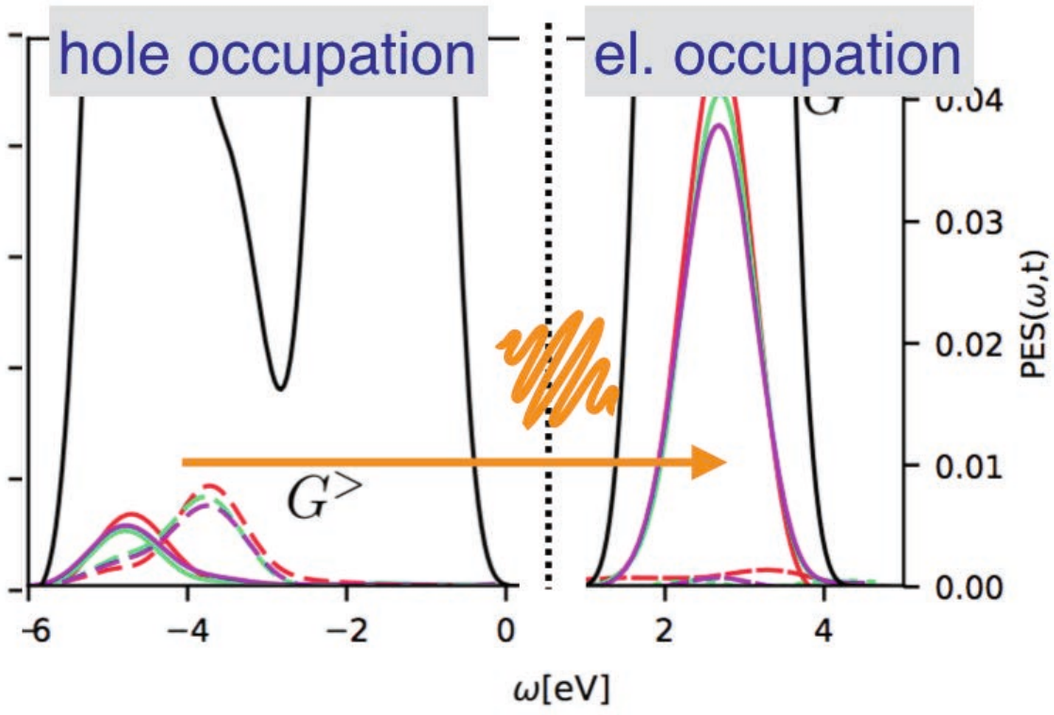


Photo-doping in a charge transfer insulator

Golez, Boenke, Eckstein, Werner,
PRB(R) 2019, PRB 2019

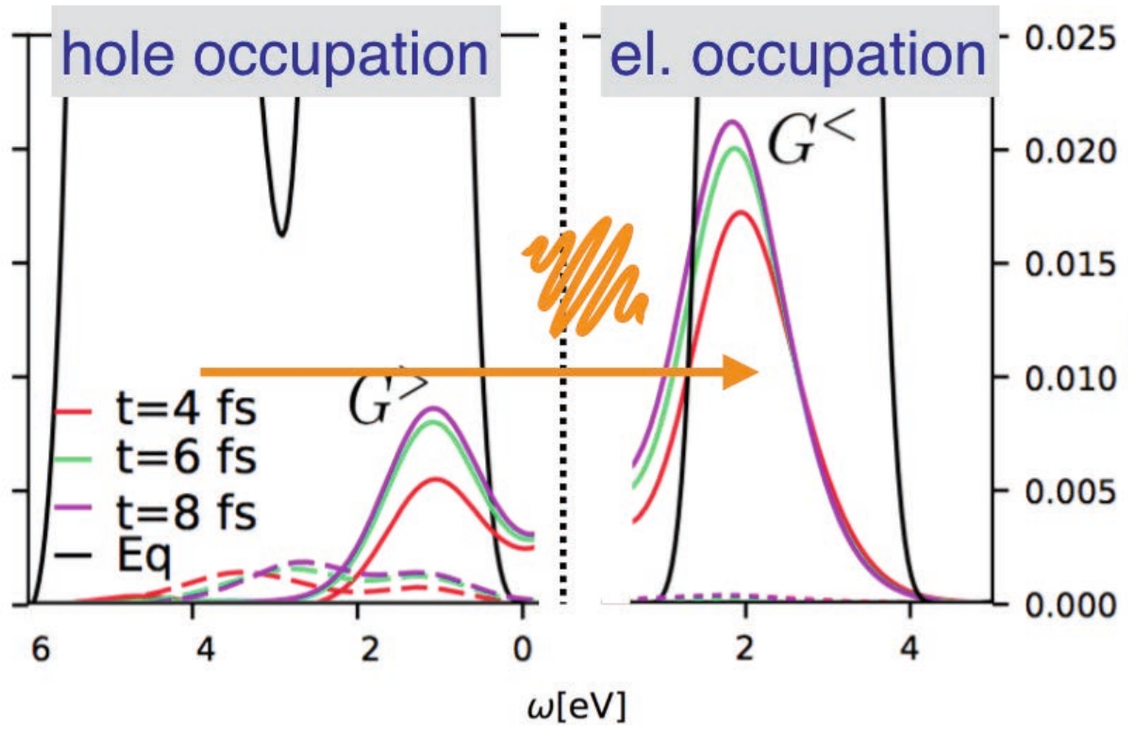
Effect of dynamical screening on population dynamics:

DMFT+Hartree Fock:



⇨ No population dynamics

DMFT+GW:



⇨ Ultrafast relaxation of electron and hole populations

(electron boson scattering)

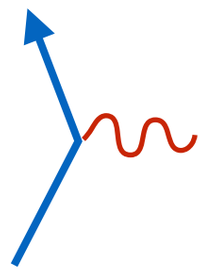
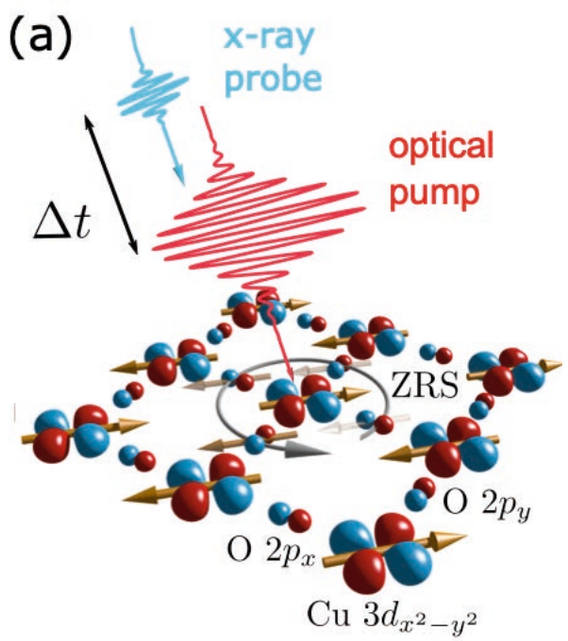
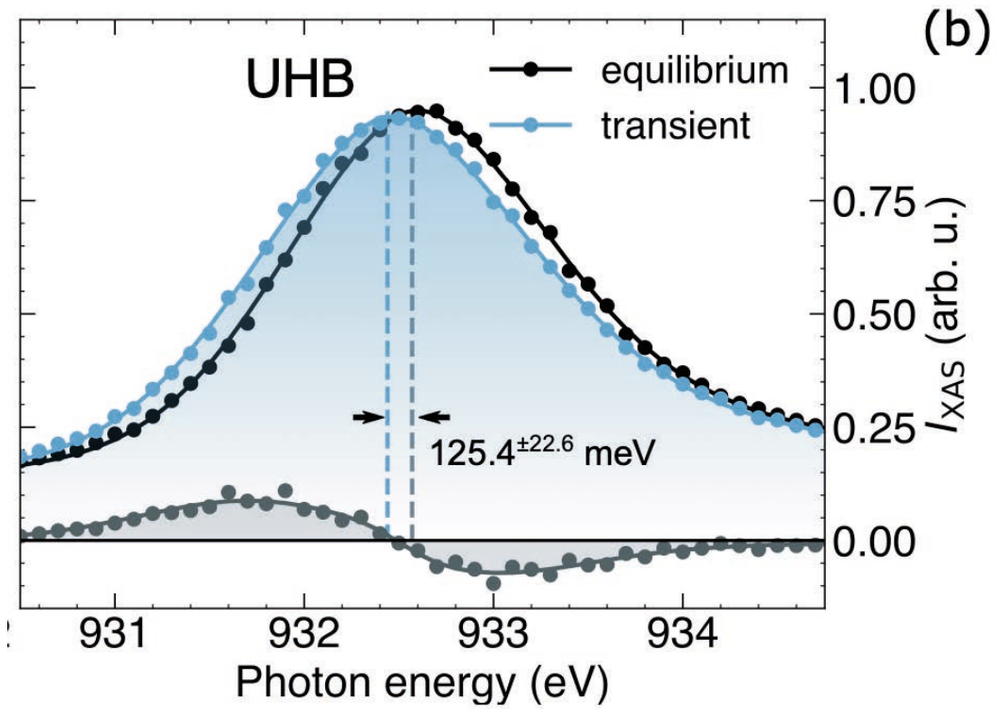


Photo-doping in a charge transfer insulator

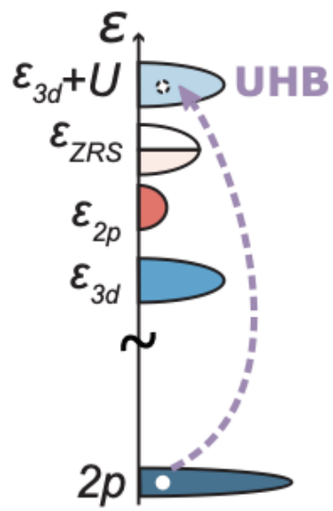
Optical excitation on cuprate LSCO, XAS probe

Baykusheva et al., Phys. Rev. X **12**, 011013 (2022)





Cu edge



… but Xrays are different from ARPES:

Final state: strongly bound (localized) core-valence exciton:
Sensitive to dynamic screening … more specific theory needed

Summary / Outlook

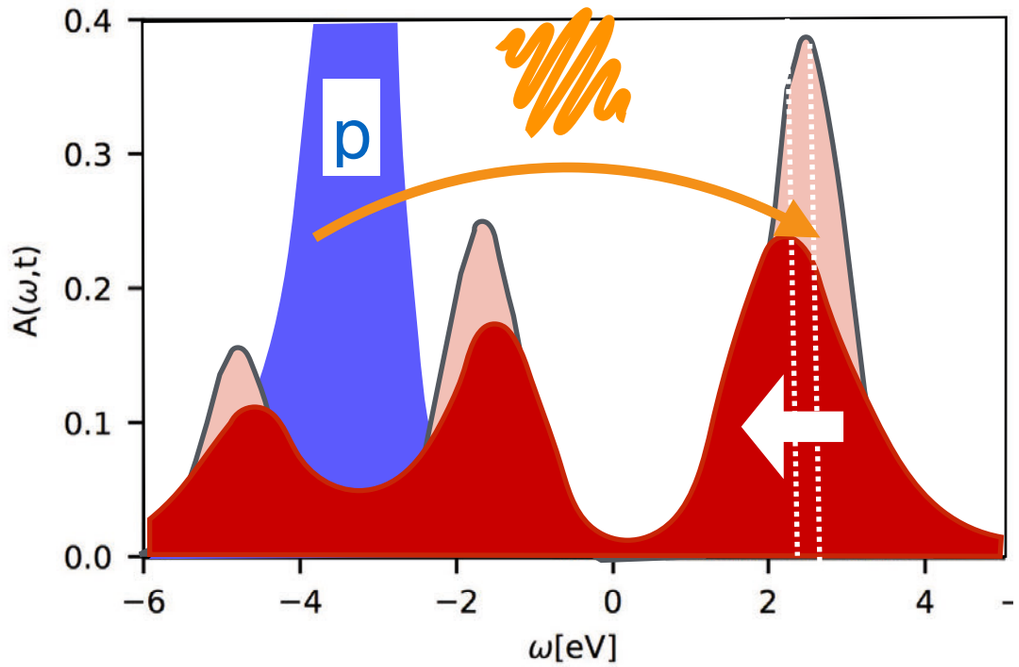
Keldysh formalism:

Formulation of many-body theory in real time

Coupled equations for time-dependent spectrum and occupation

Non-equilibrium DMFT (+GW)

Electronic structure of correlated systems out of equilibrium



Outlook, future (technical) developments:

Non-equilibrium Green's function simulations at long times
⇨ realistic many-band simulations

Non-perturbative impurity solvers

Electron boson systems, electron lattice dynamics