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UNIVERSITÄT
ERLANGEN-NÜRNBERG

NATURWISSENSCHAFTLICHE
FAKULTÄT

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

Martin Eckstein

”Quantum Matter: Computation Meets Experiments”

Aspen, March 12, 2020

Controlling materials out of equilibrium

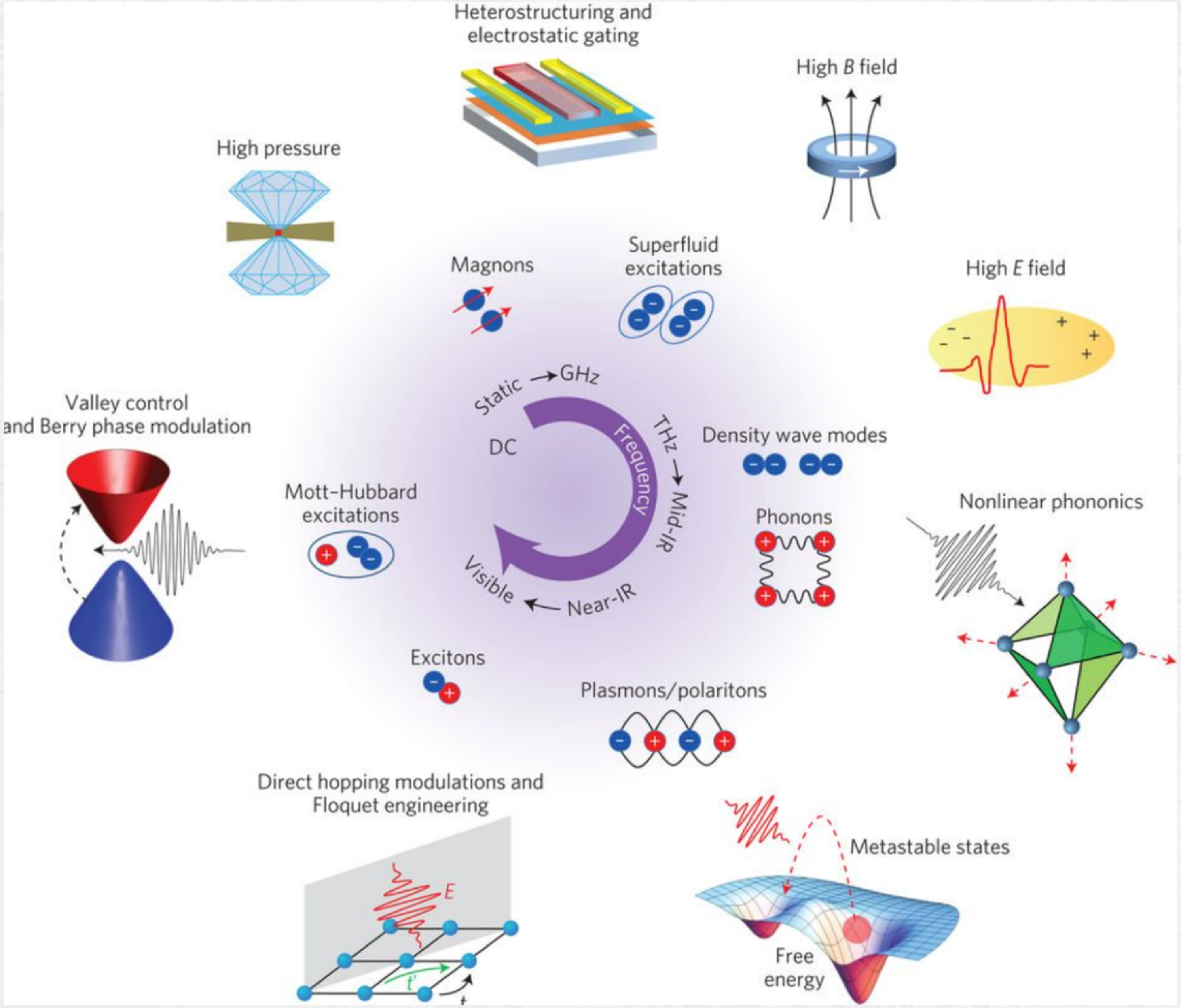
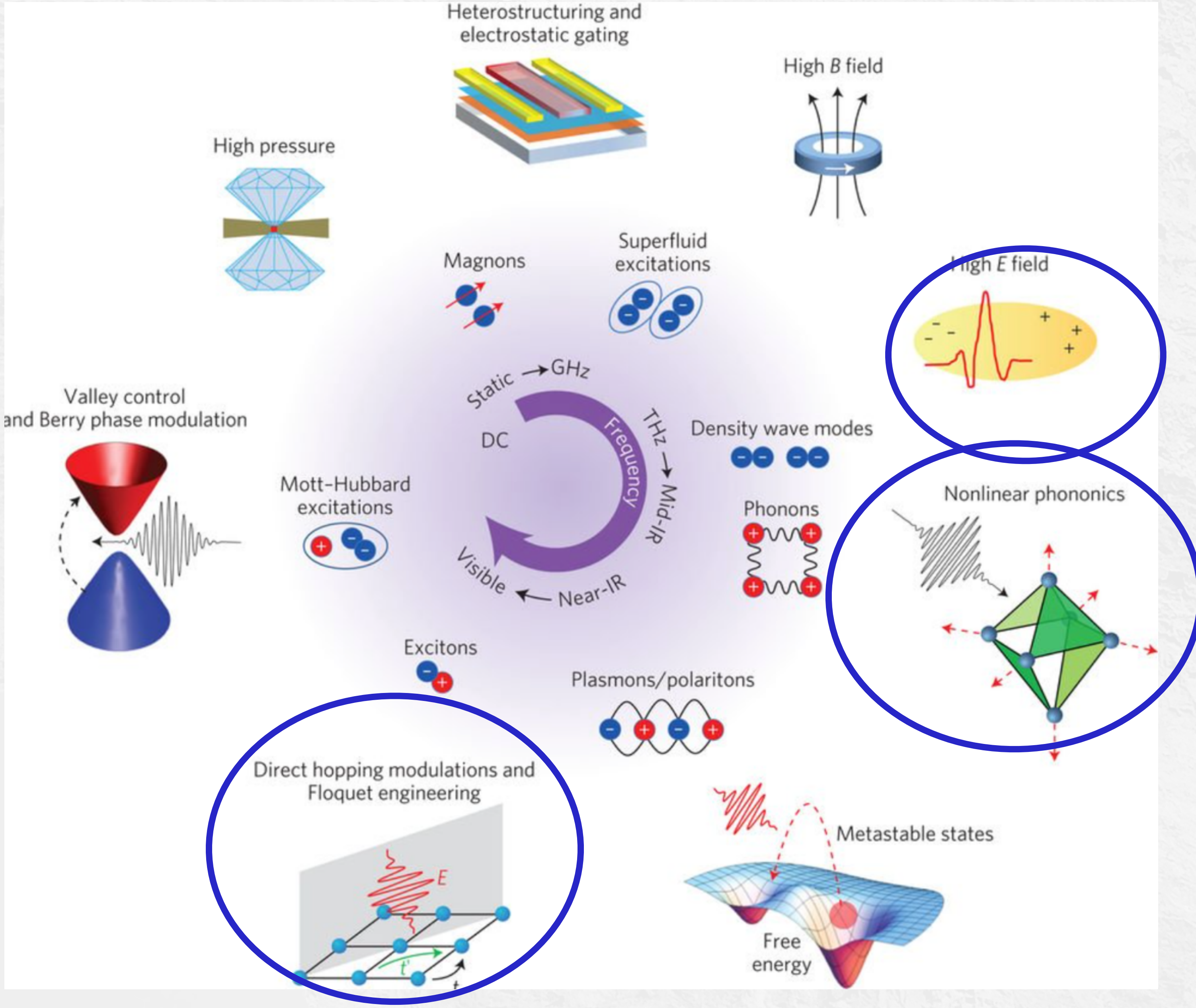


image from Basov et al., Nature Materials 2017

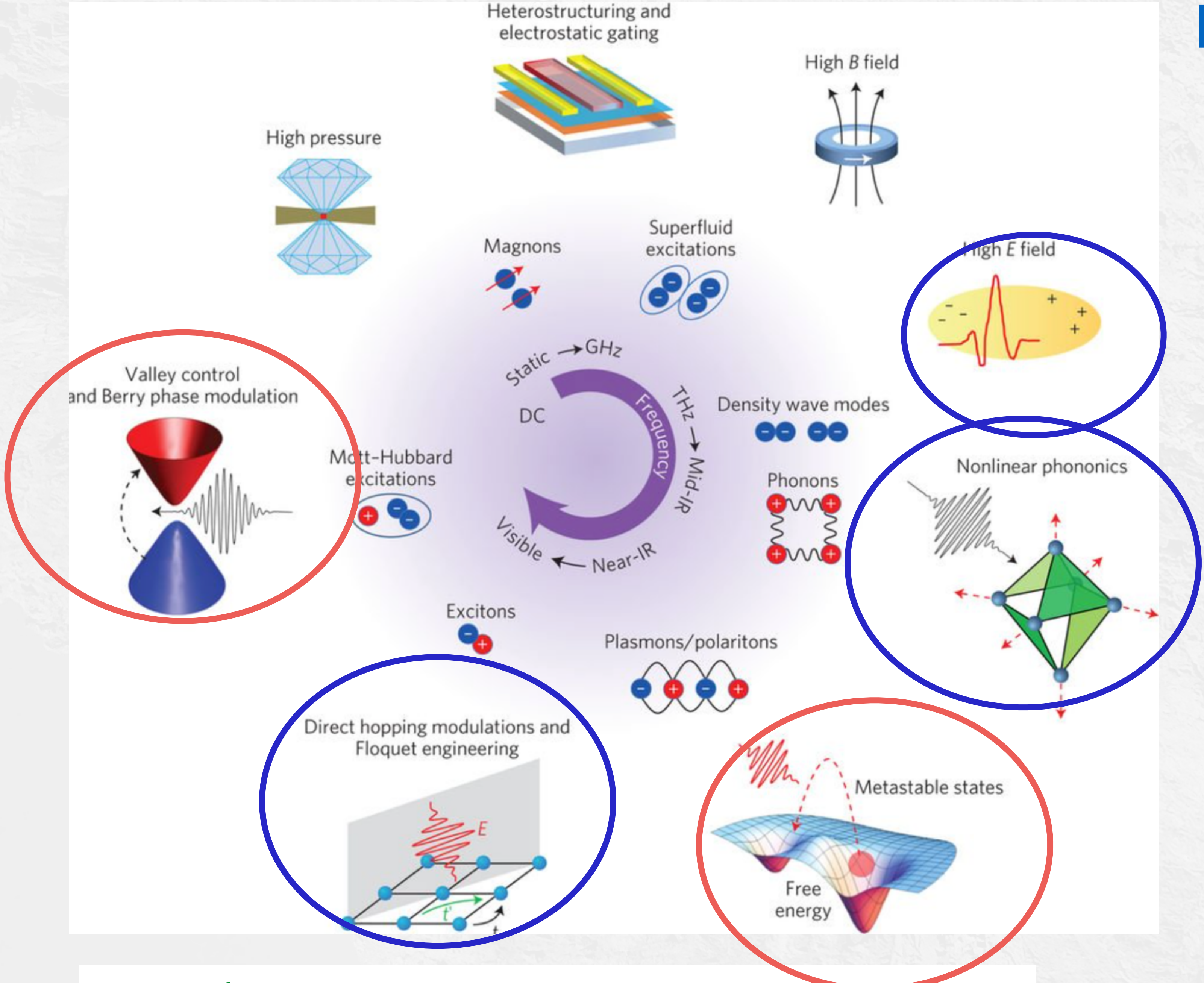
Controlling materials out of equilibrium



Field-induced phenomena

image from Basov et al., Nature Materials 2017

Controlling materials out of equilibrium



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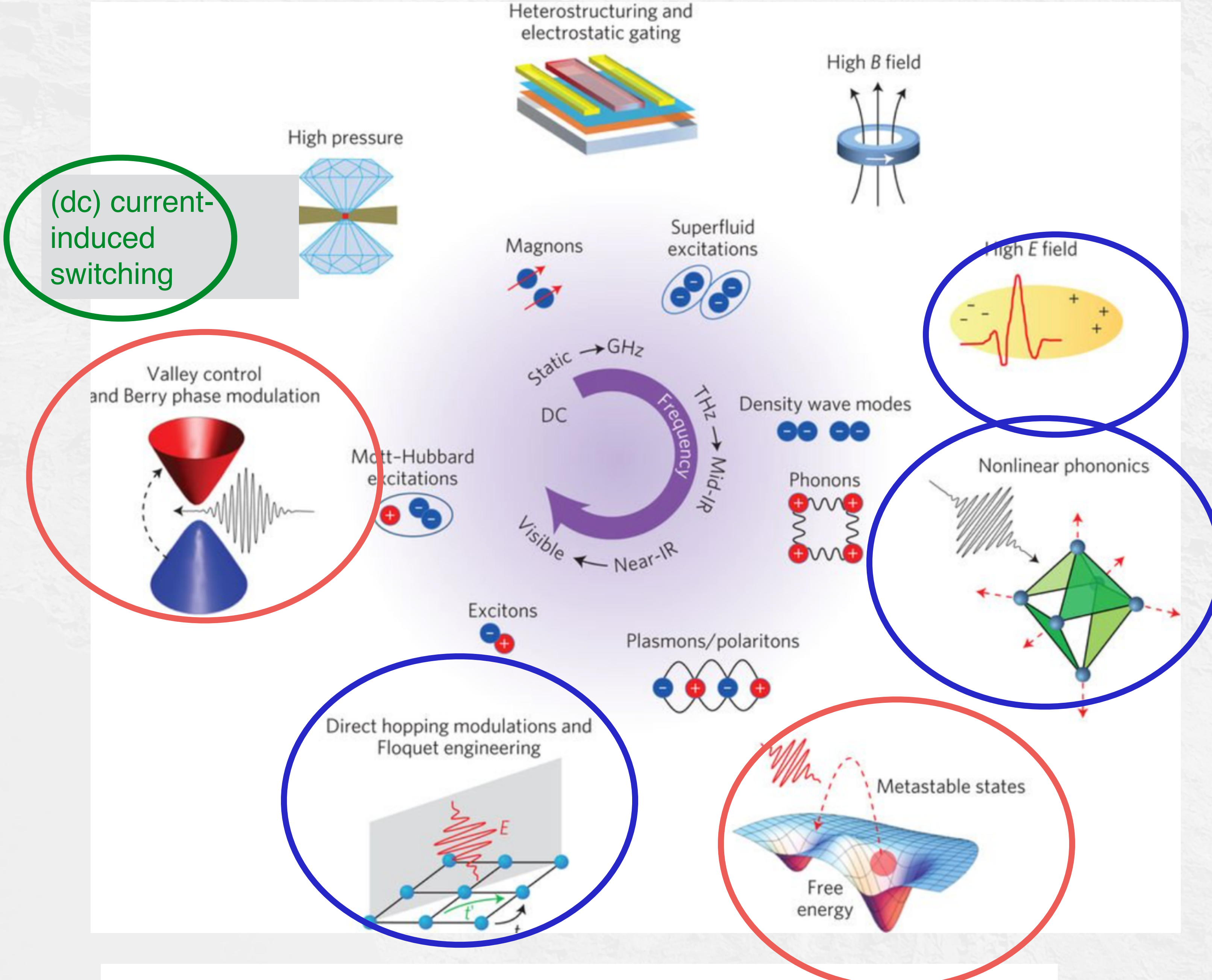
Field-induced phenomena

Distributional engineering

Controlling materials out of equilibrium



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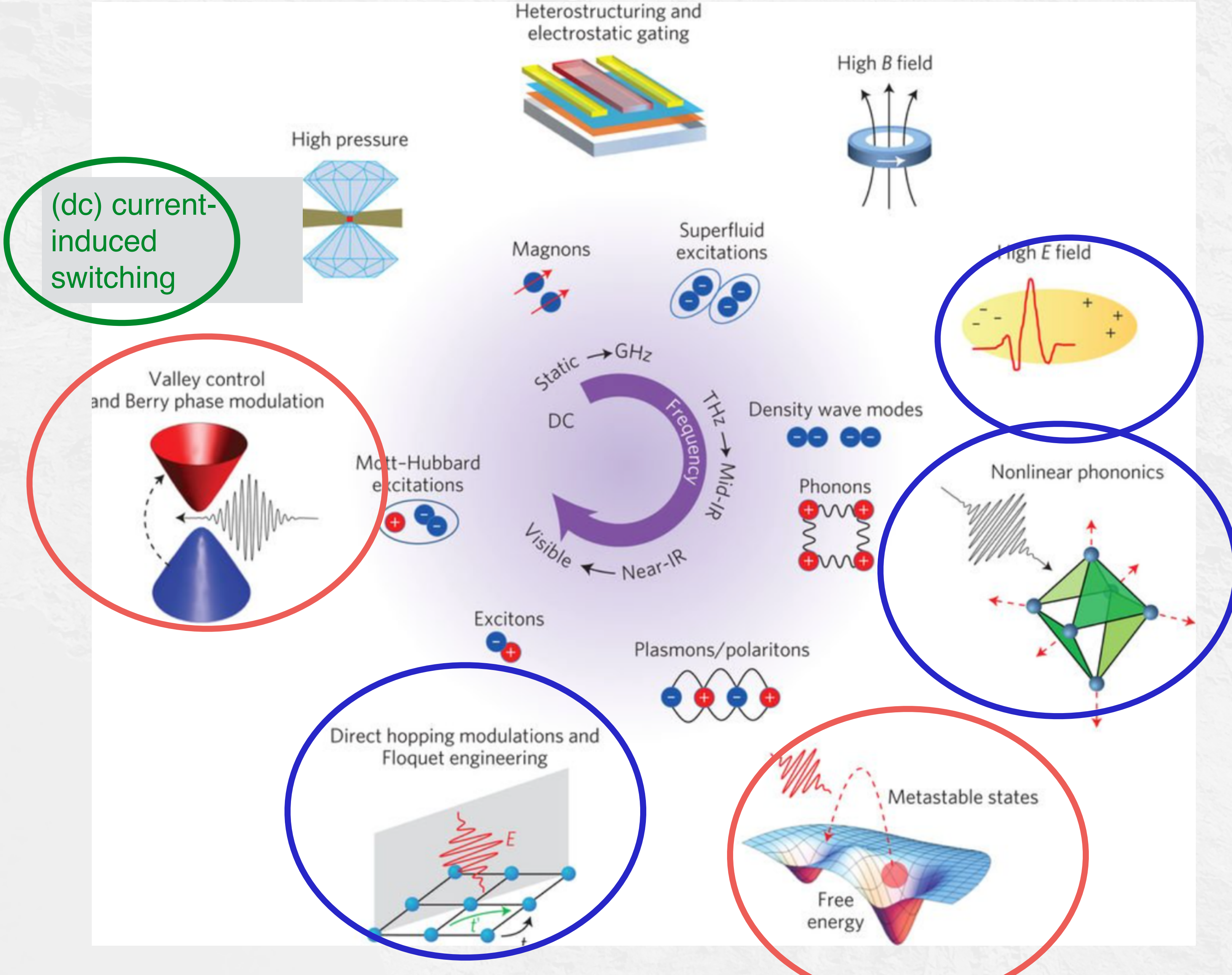
Non-equilibrium steady states

Controlling materials out of equilibrium



Field-induced phenomena

Distributional engineering

Non-equilibrium steady states

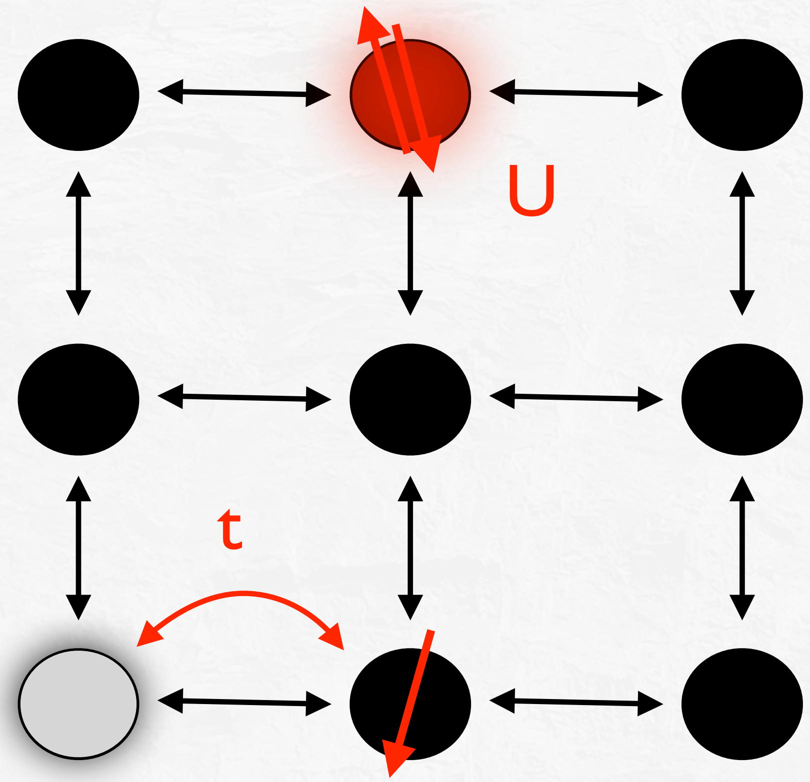
image from Basov et al., Nature Materials 2017

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.

Real-time dynamics of correlated electrons

Hubbard model

$$H = -t \sum_{\langle ij \rangle, \sigma=\uparrow,\downarrow} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



plus electric fields (Peierls substitution)

$$t_{ij} \to t_{ij} e^{i\phi_{ij}(t)}$$

$$\phi_{ji}(t) = e\vec{A}(t)(\vec{r}_j - \vec{r}_i)$$

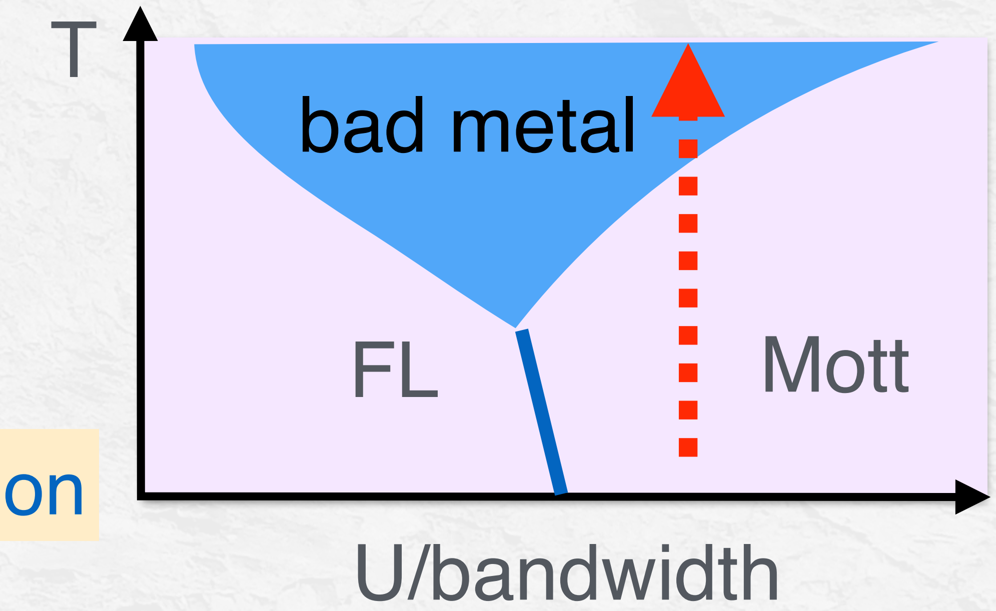
(plus possibly dissipation, phonons)

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



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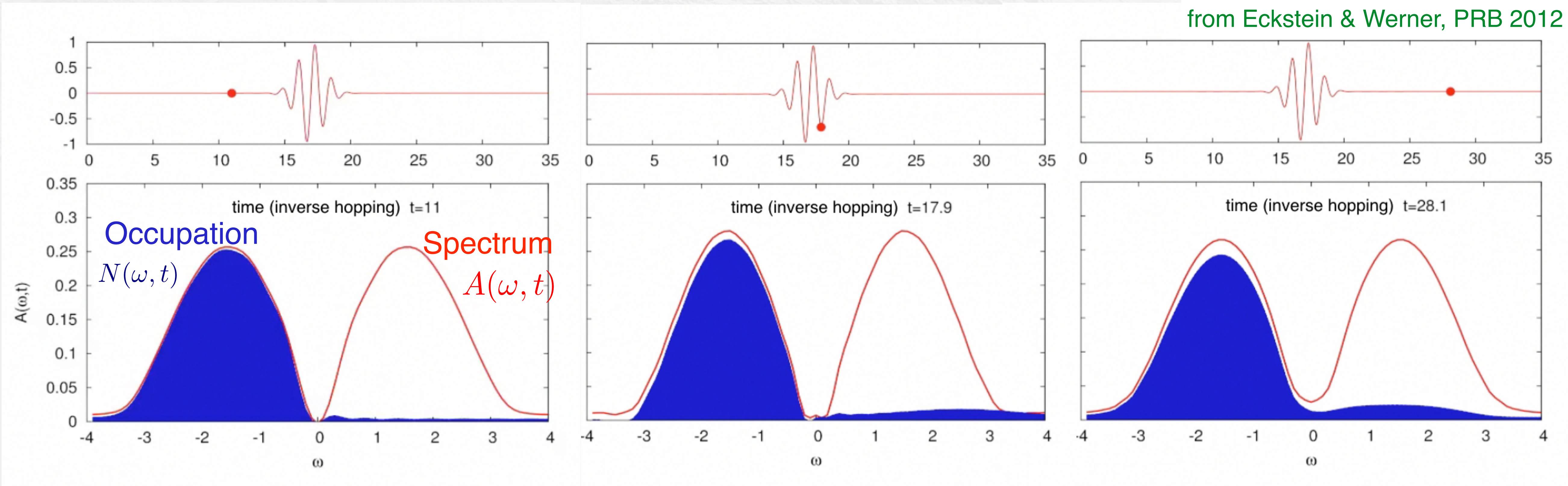


charge transfer, field-induced band narrowing

Relaxation

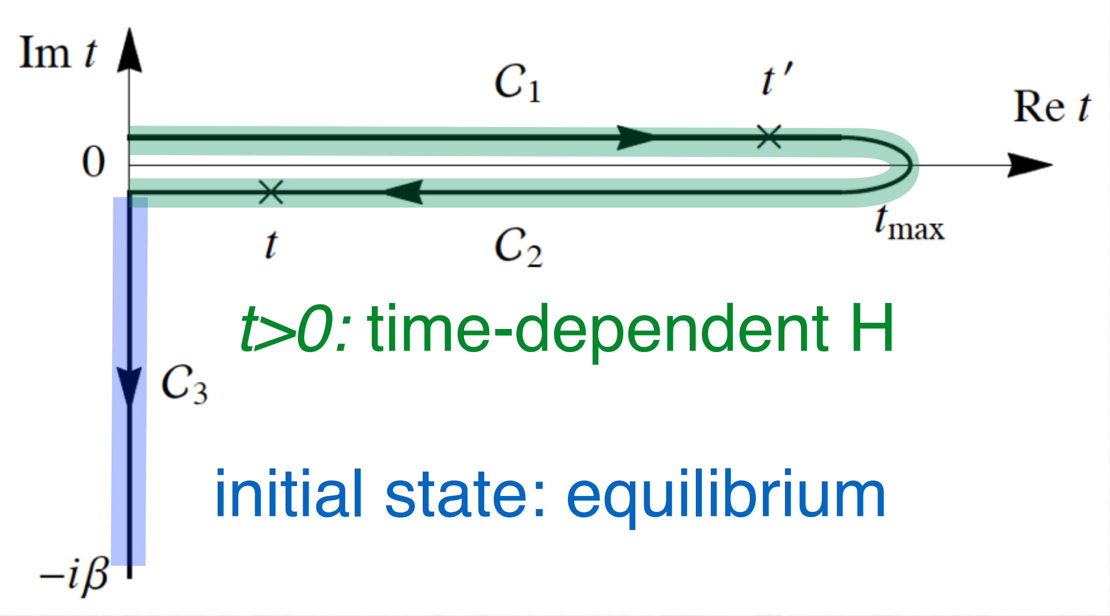
Non-equilibrium Green's functions

Keldysh formalism:

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

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

$$\Longleftrightarrow \quad G(t,t') = -i\langle T_{\mathcal{C}}\, c(t) c^\dagger(t')\rangle$$



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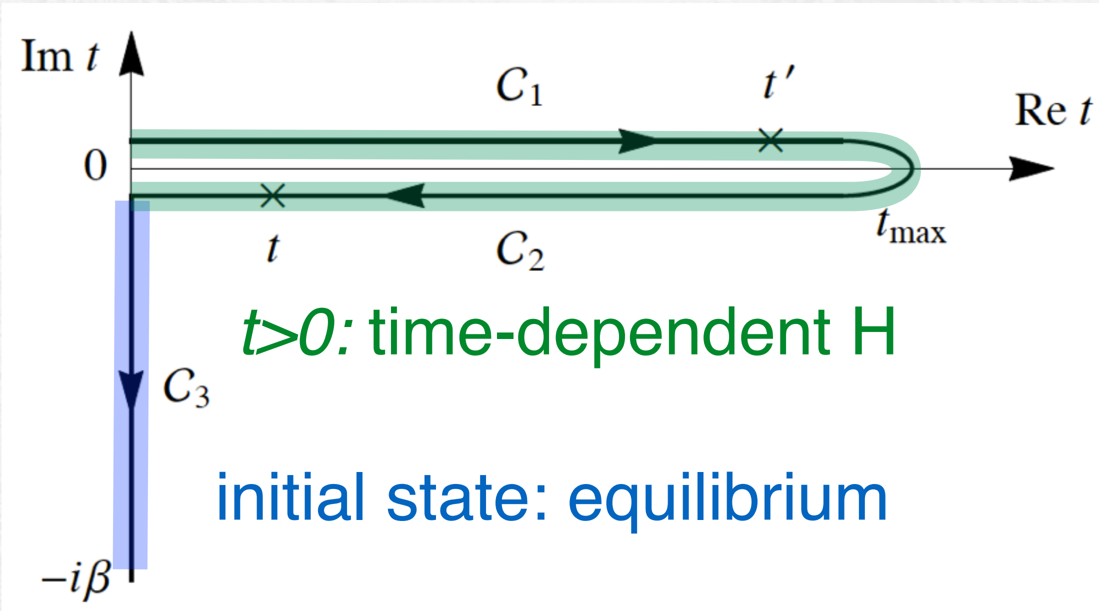
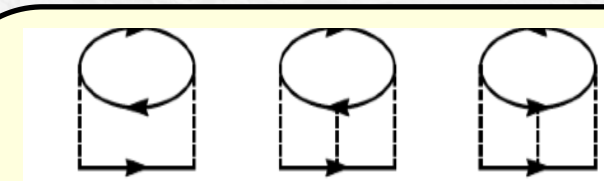
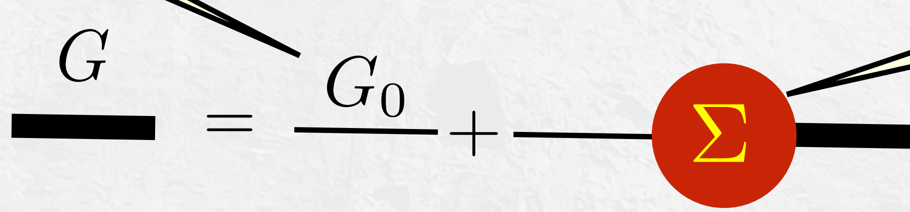


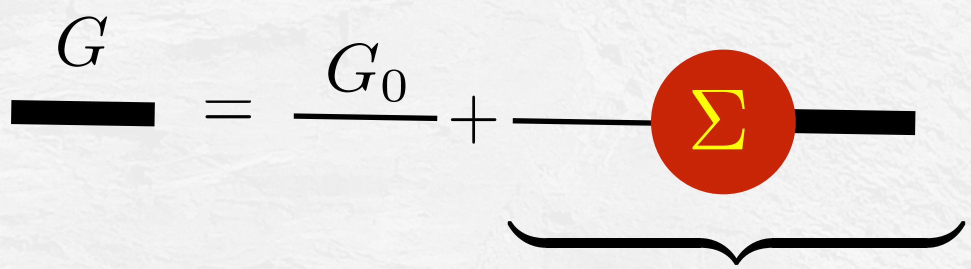
time-dep. mean-field solution


, DMFT, ...



Non-equilibrium Green's functions

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



$$\int_{\text{previous times}} dt_1 dt_2 G_0(t,t_1)\Sigma(t_1,t_2)G(t_2,t)$$

open source code: nessi.tuxfamily.org

Non-equilibrium Green's functions

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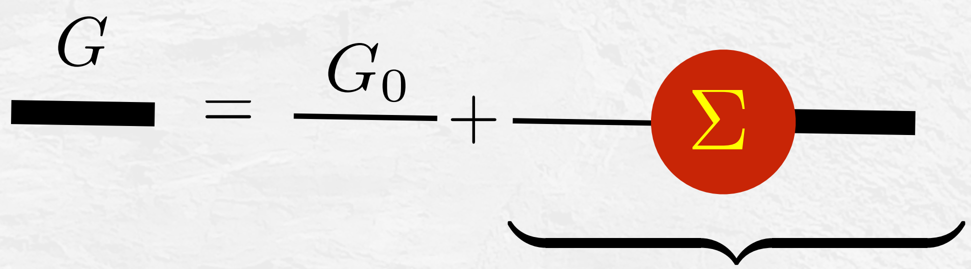

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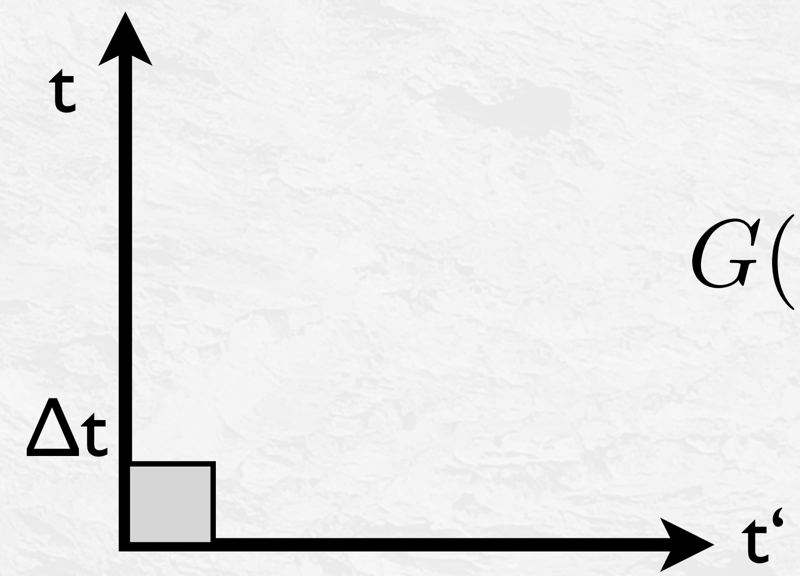

$G(t,t')$

„causal" time-propagation:

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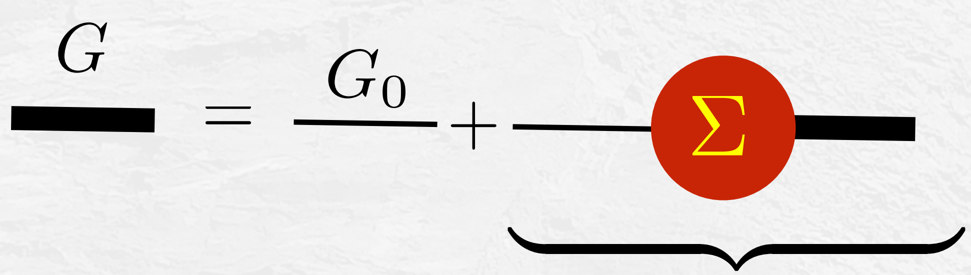

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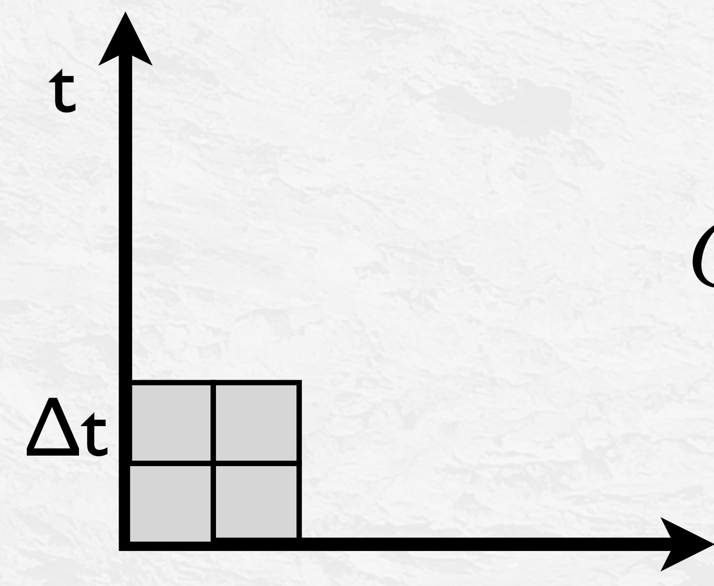

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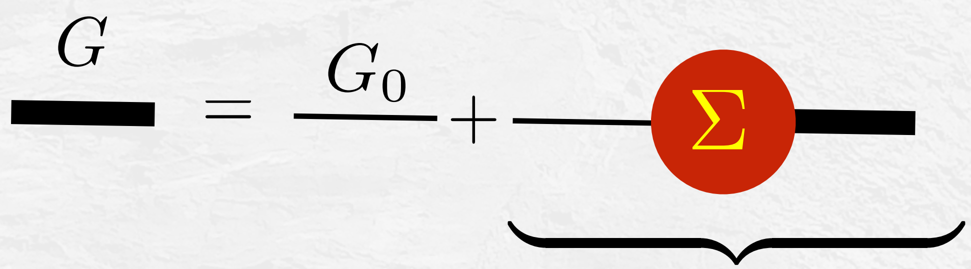

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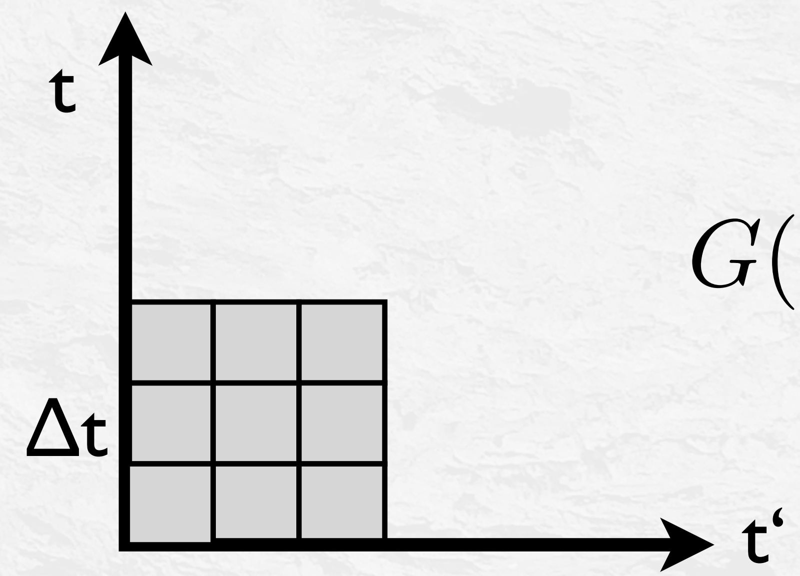

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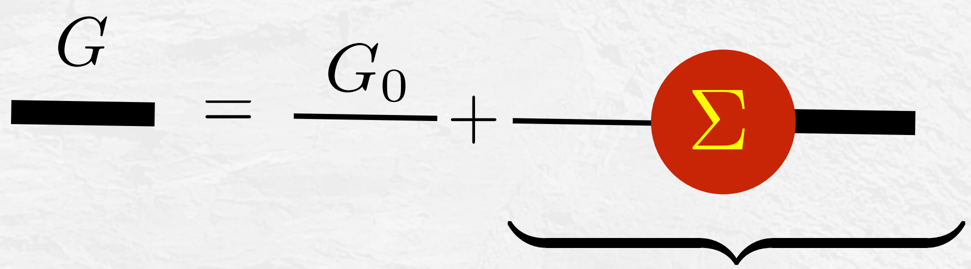

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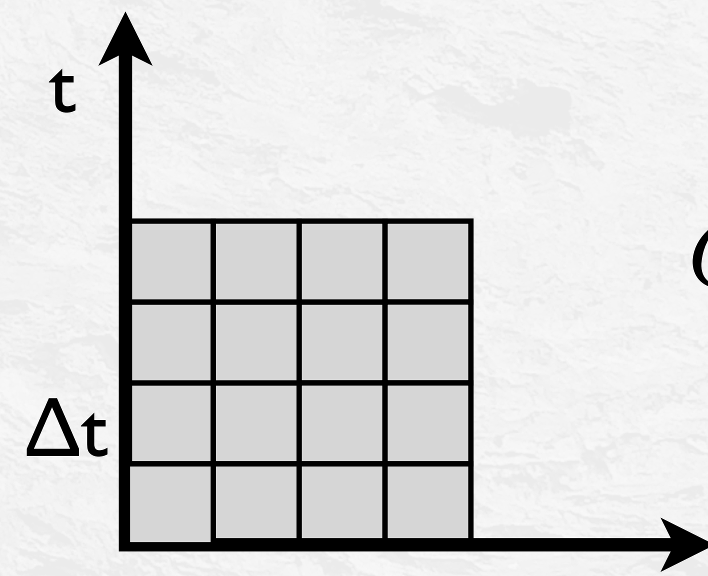

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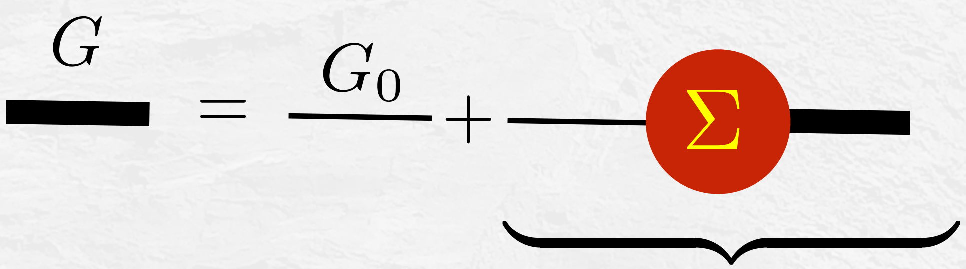

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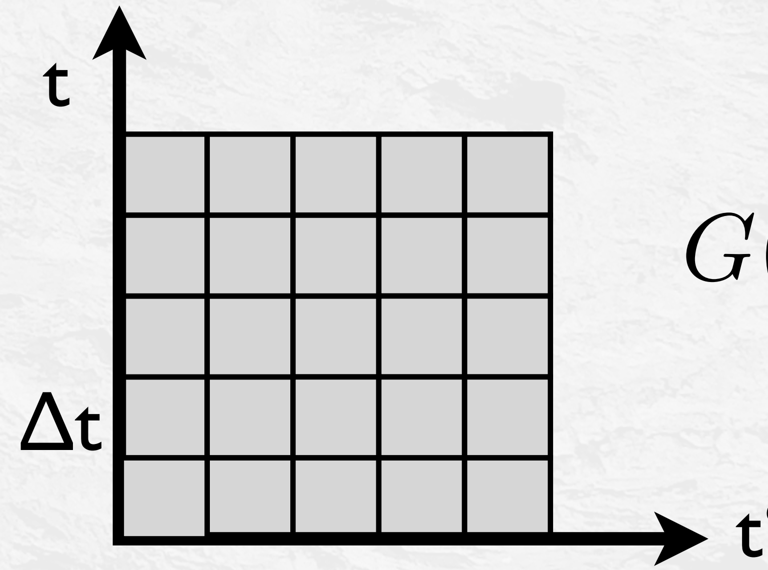

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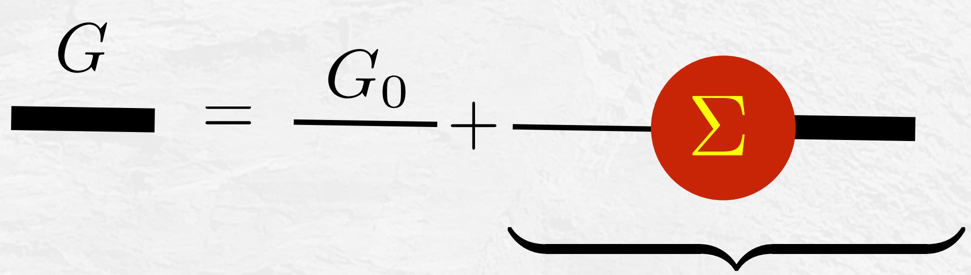

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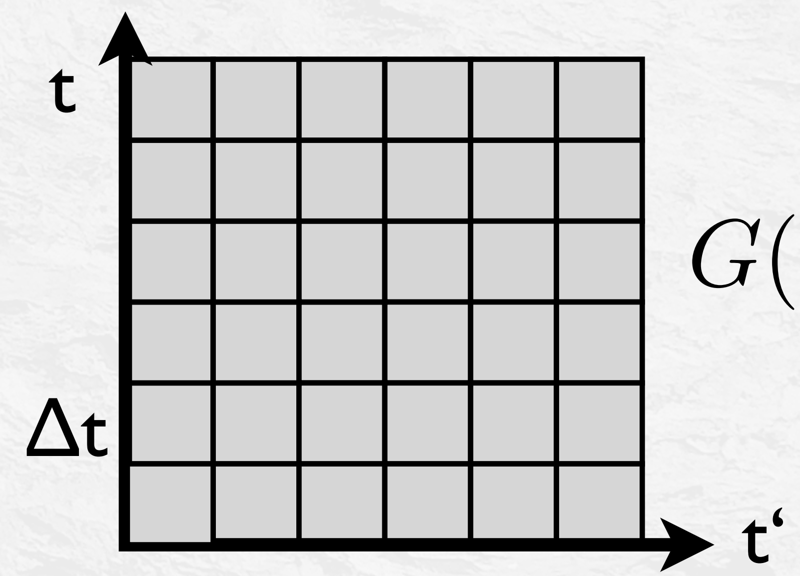

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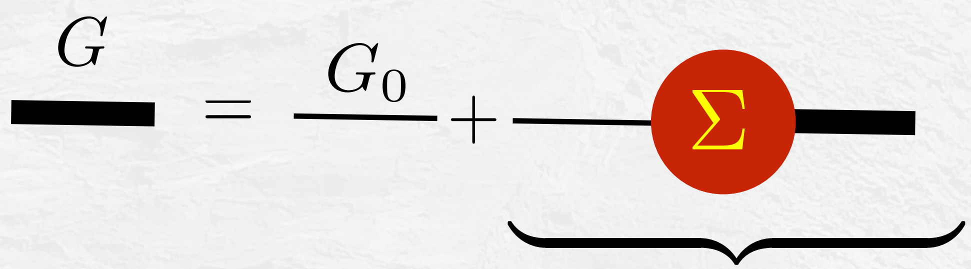

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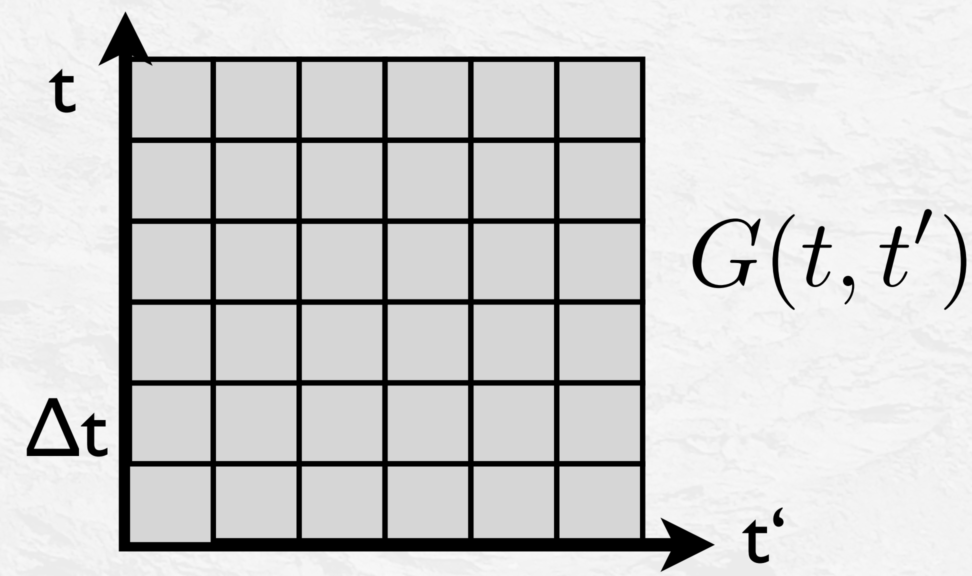

„causal“ time-propagation:

Memory: eg. GW simulation

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

Δt=0.01/W=0.06fs ⇨ >1TB for 100fs

open source code: nessi.tuxfamily.org

Non-equilibrium Green's functions

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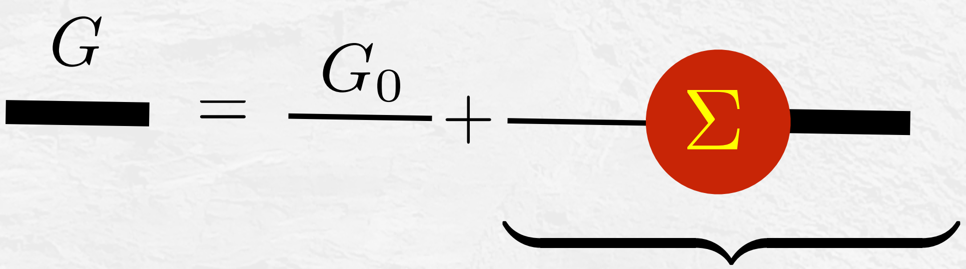

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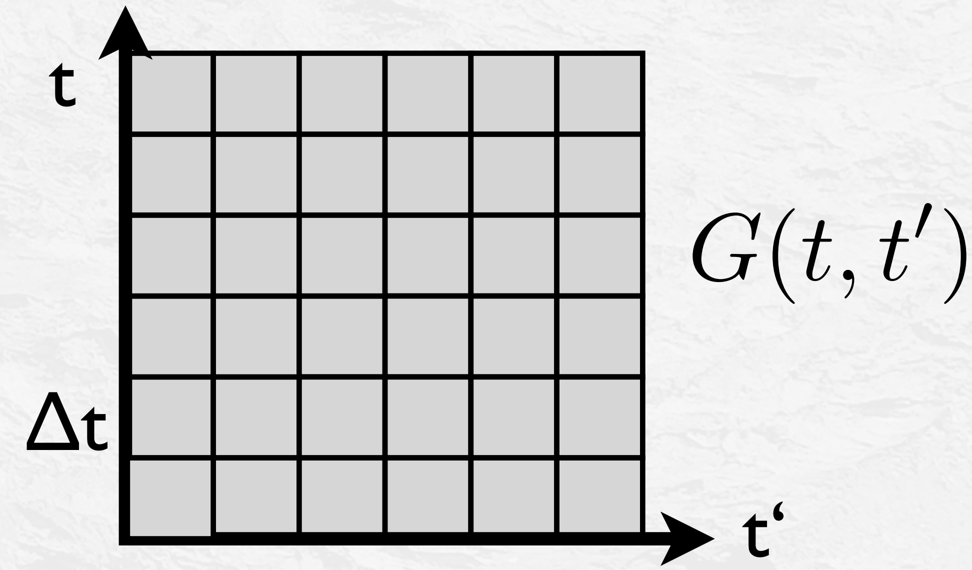


„causal" time-propagation:

Memory: eg. GW simulation

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

Δt=0.01/W=0.06fs ⇨ >1TB for 100fs

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

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

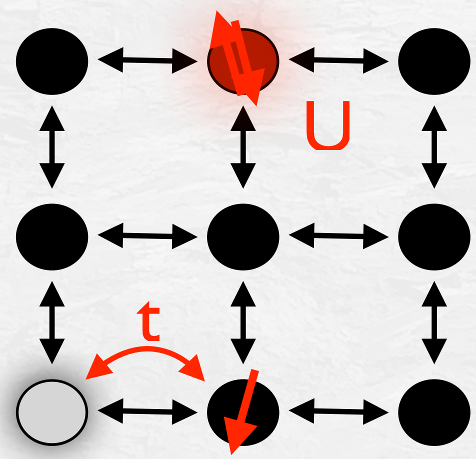
Schueler et al, PRB 2018,
Stahl et al., in preparation

open source code: nessi.tuxfamily.org

Non-equilibrium Green's functions

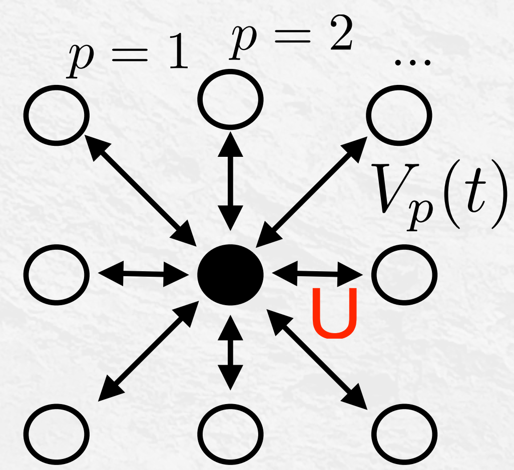
Computational challenges (II):

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



$$G_{imp} = G_{loc}$$

$$\Sigma_{imp} = \Sigma_{loc}$$



(numerically unbiased) impurity solvers:

MPS Wolf, et al. et Schollwöck, et al. (2014), Bauernfeid et al. et Evertz (2017)

CTQMC Gull et al. (2012, …), Cohen et al. (2018)

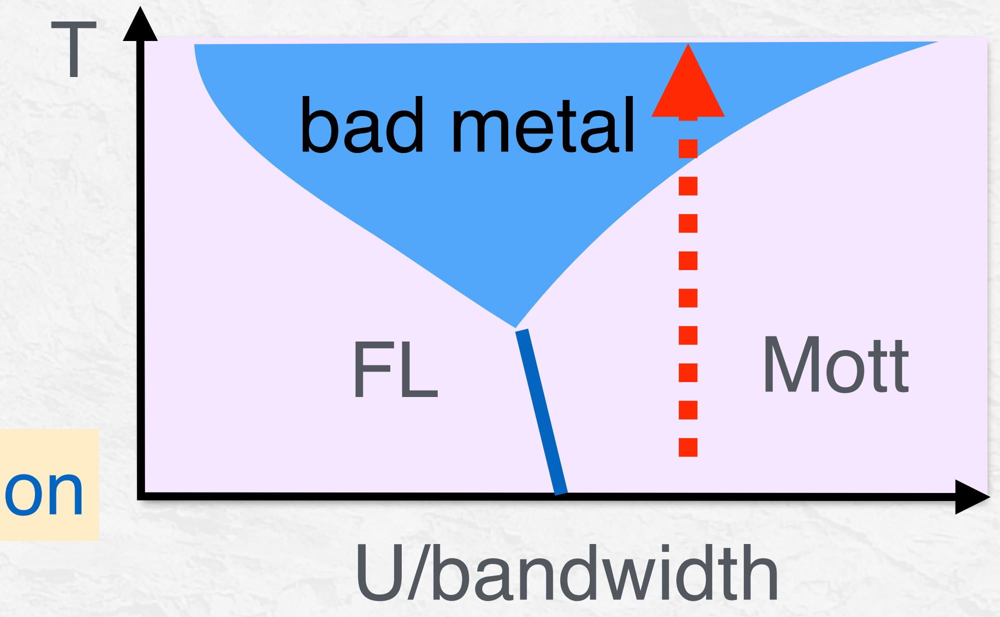
Keldysh QMC Parcollet e al.

Here: Still NCA

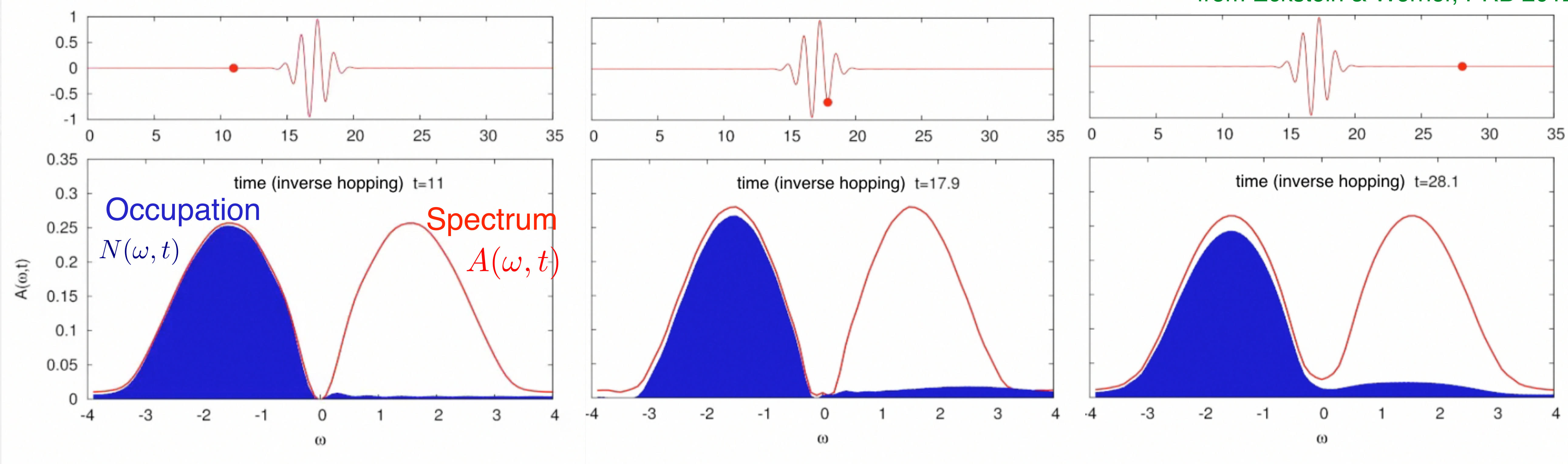
Real-time dynamics of correlated electrons

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

Intertwined evolution of spectrum and occupation



from Eckstein & Werner, PRB 2012



charge transfer, field-induced band narrowing

Relaxation

Real-time dynamics of correlated electrons

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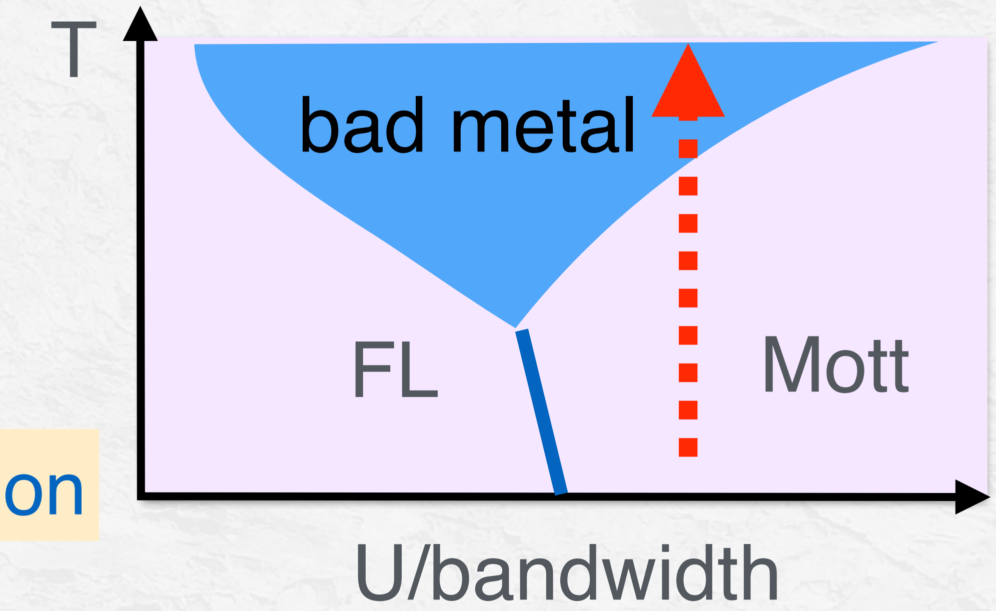

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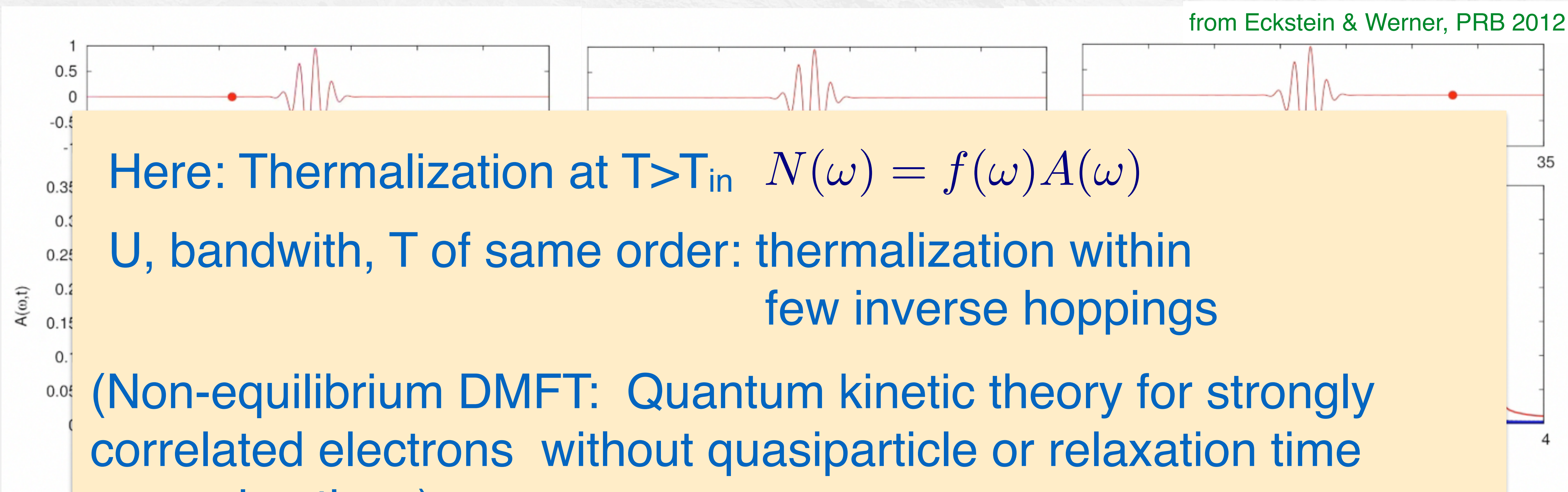


Here: Thermalization at T>T_{in} $N(\omega) = f(\omega)A(\omega)$

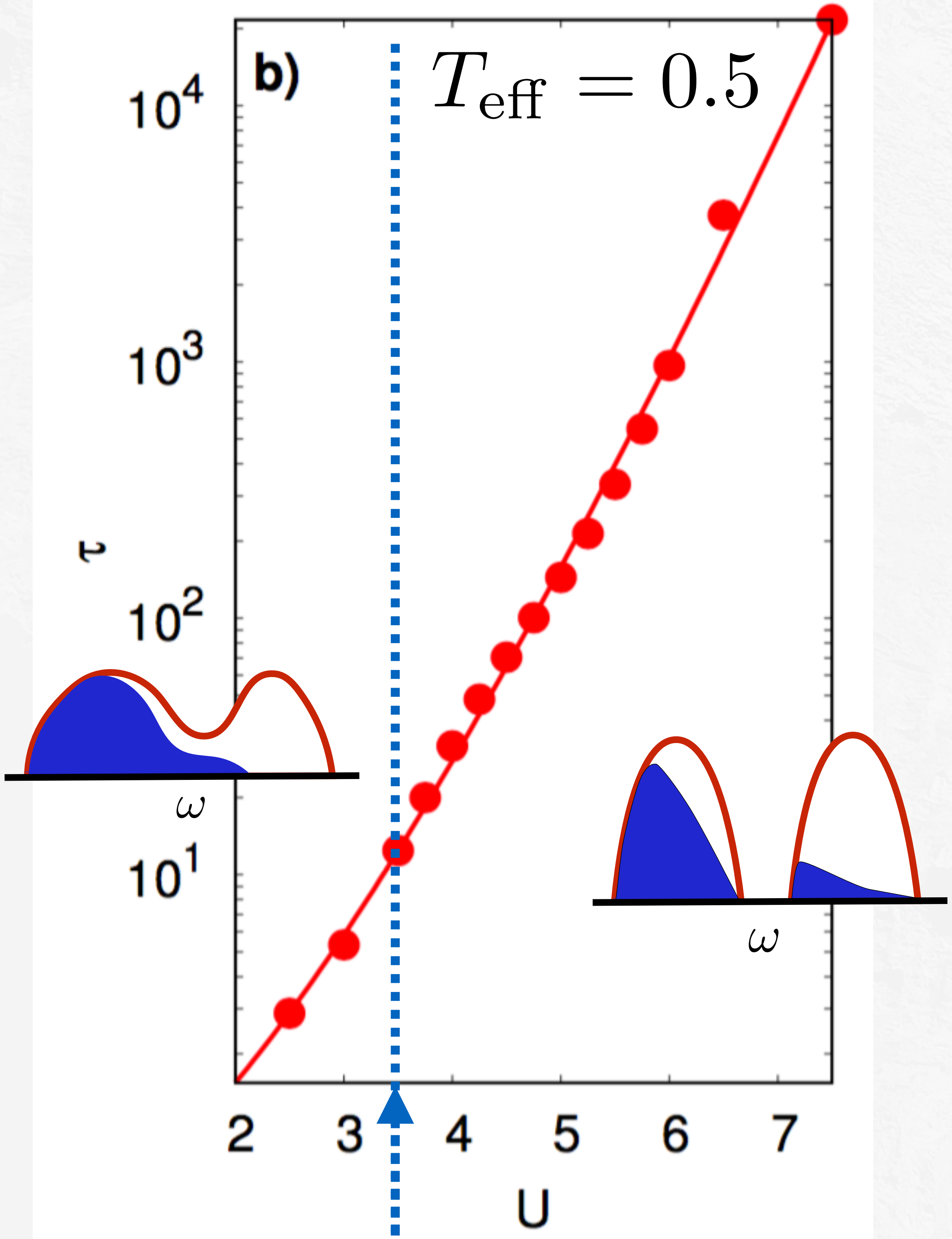
U, bandwith, T of same order: thermalization within few inverse hoppings

(Non-equilibrium DMFT: Quantum kinetic theory for strongly correlated electrons without quasiparticle or relaxation time approximations)

induced band narrowing

Lifetime of photo-doped states

[time] = 1/hopping



Mott transition

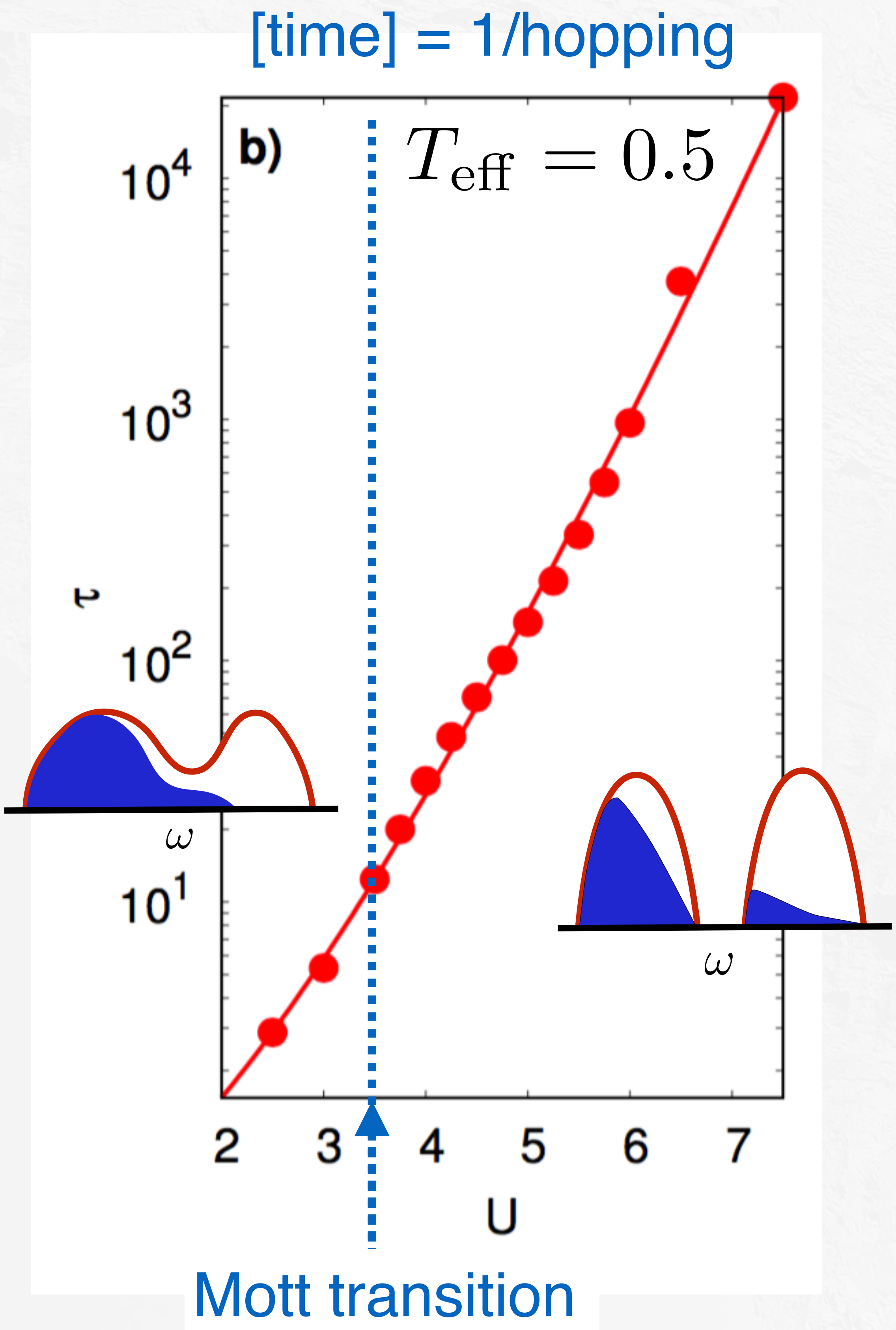
Thermalization of the double occupancy after (pretty much any kind of) excitation:

$$d(t) = d(T_{\text{eff}}) + Ae^{-t/\tau}$$

T_{eff} determined by total energy

Eckstein & Werner, PRB (2010)

Lifetime of photo-doped states



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

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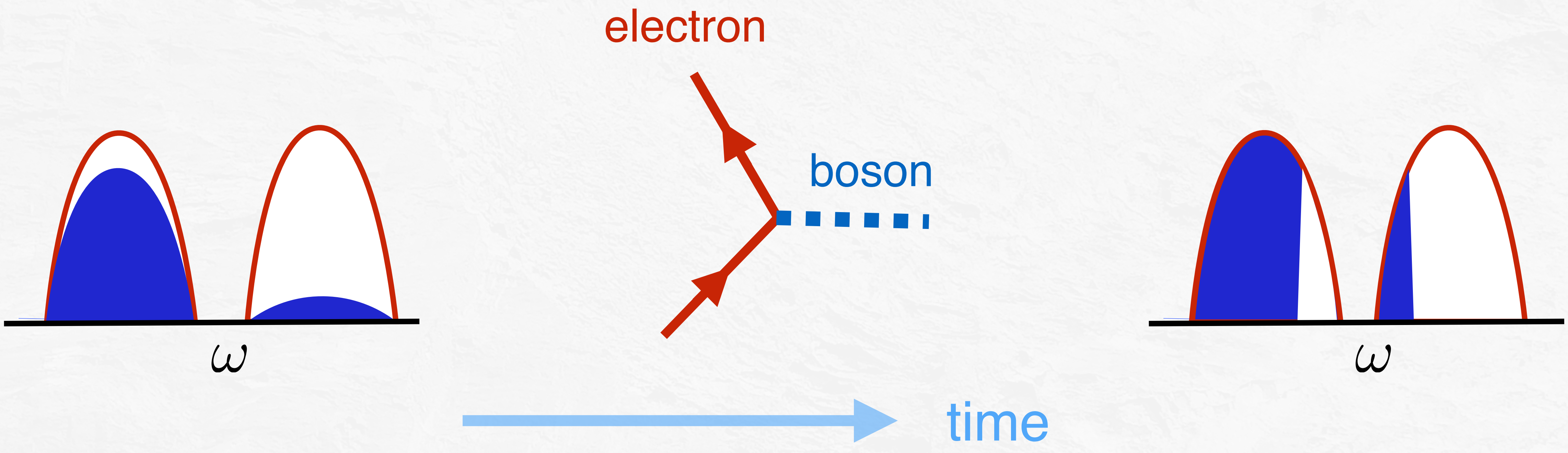
Long-lived non-thermal population in the upper Hubbard band

Eckstein & Werner, PRB (2010)

Internal relaxation



⇨ Phonons (quasi external heat bath)

⇨ Non-local charge and spin fluctuations (beyond DMFT)

⇨ Short range (Cluster DMFT) Eckstein & Werner, Sci. Rep 2016

⇨ Long range (extended DMFT, DMFT+GW)

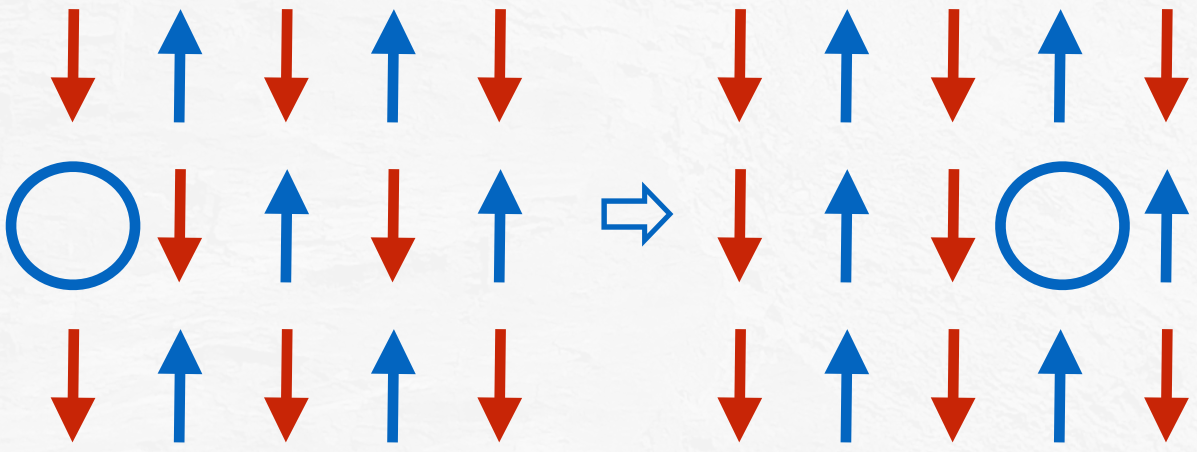
Golez, Eckstein, Werner PRB 2015, PRL 2017, PRB 2019

Golez, Boenke Eckstein, Werner PRB 2019

Bittner, **Golez**, Eckstein, Werner PRB 2019

(Short-range) spin correlations

Eckstein & Werner, Sci. Rep 2016



Hole motion:

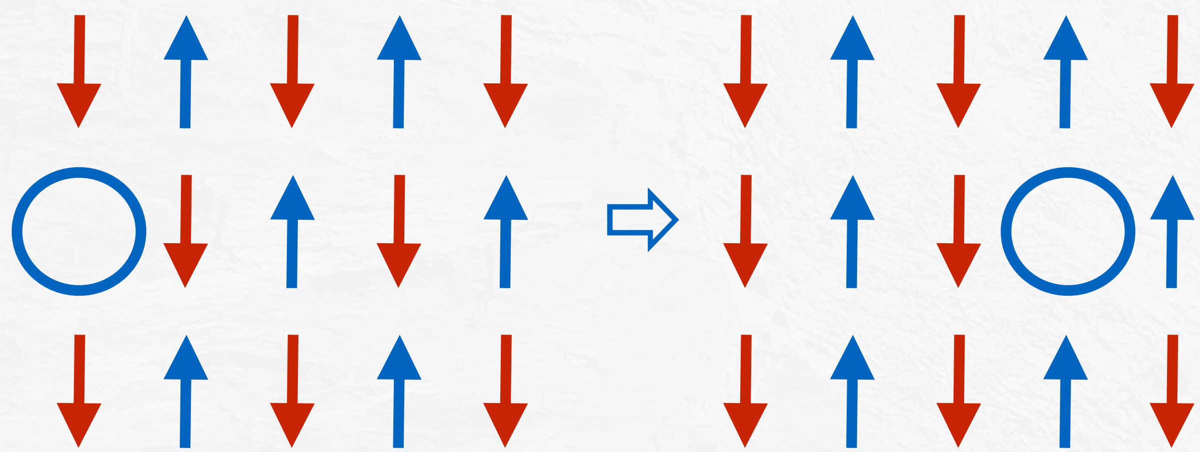
⇨ defects in AFM order

⇨ transfer of energy to spin

Active also in paramagnetic phase?

(Short-range) spin correlations

Eckstein & Werner, Sci. Rep 2016



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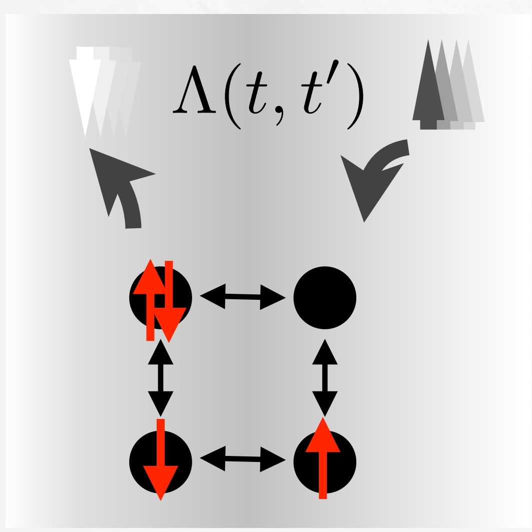
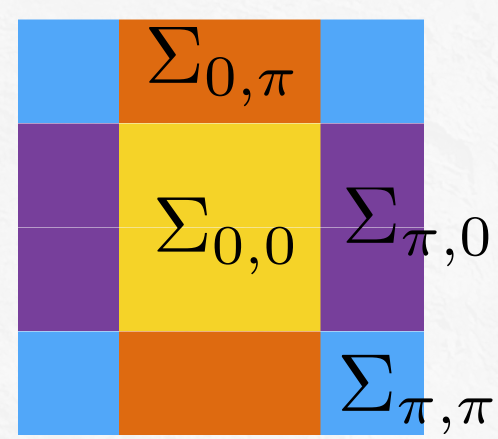
single band 2d Hubbard model U/t=12; 2x2 cluster DCA, NCA solver

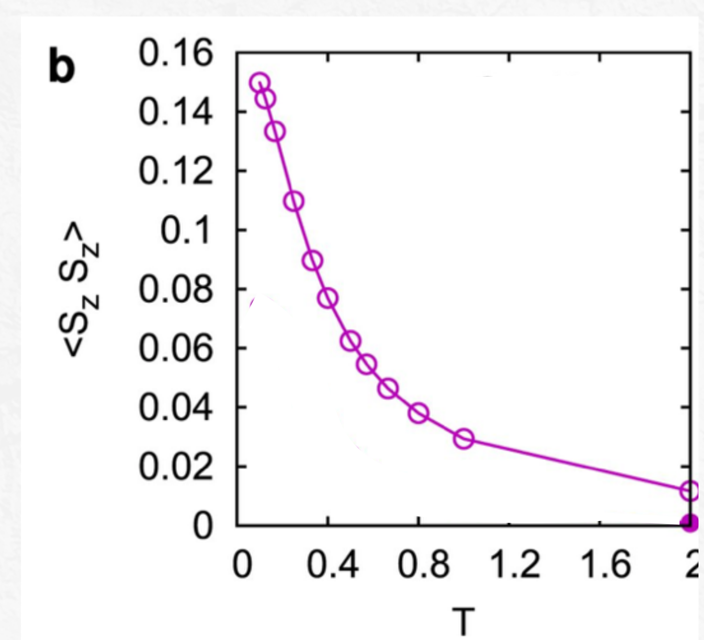


⇨



$$\Sigma_{\boldsymbol{k}}(t,t') = \sum_{\boldsymbol{K}} \Sigma_{\boldsymbol{K}}(t,t')\theta_{\boldsymbol{K}}(\boldsymbol{k})$$

NN Spin correlations (in the cluster)



(Short-range) spin correlations

Eckstein & Werner, Sci. Rep 2016

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

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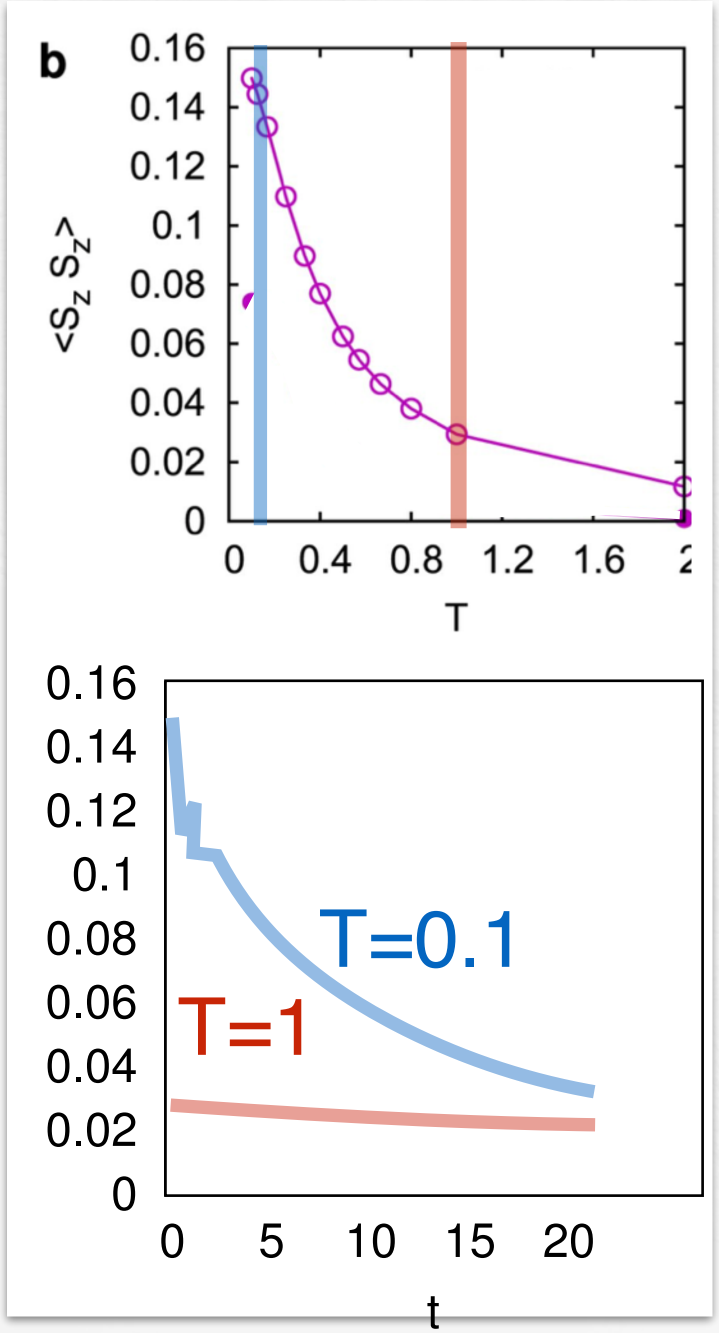
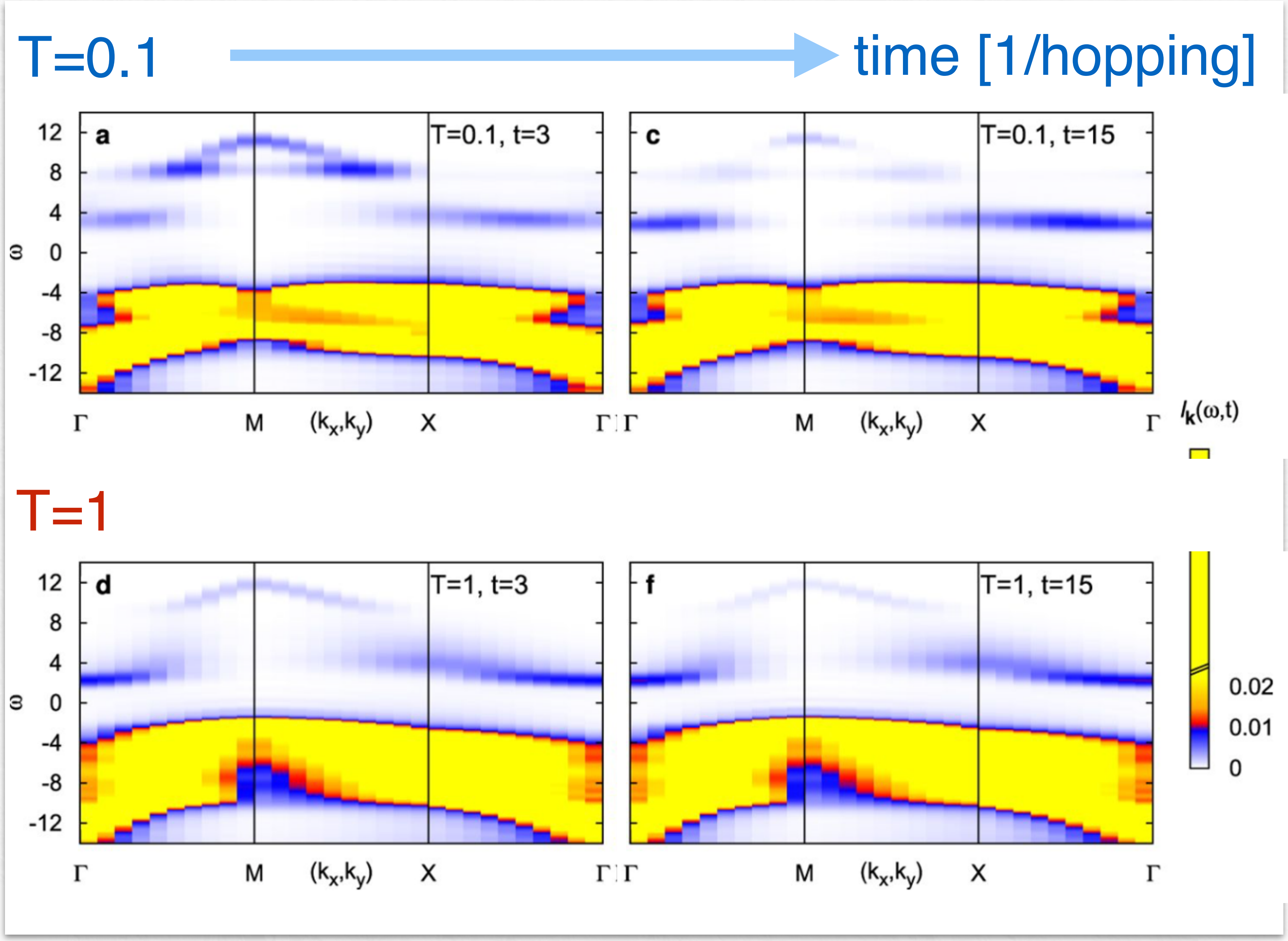


Occupied DOS $N_k(\omega, t)$



strong spin correlations: inter Hubbard band relaxation within few 1/hopping

(Short-range) spin correlations

Eckstein & Werner, Sci. Rep 2016

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

NN Spin correlations

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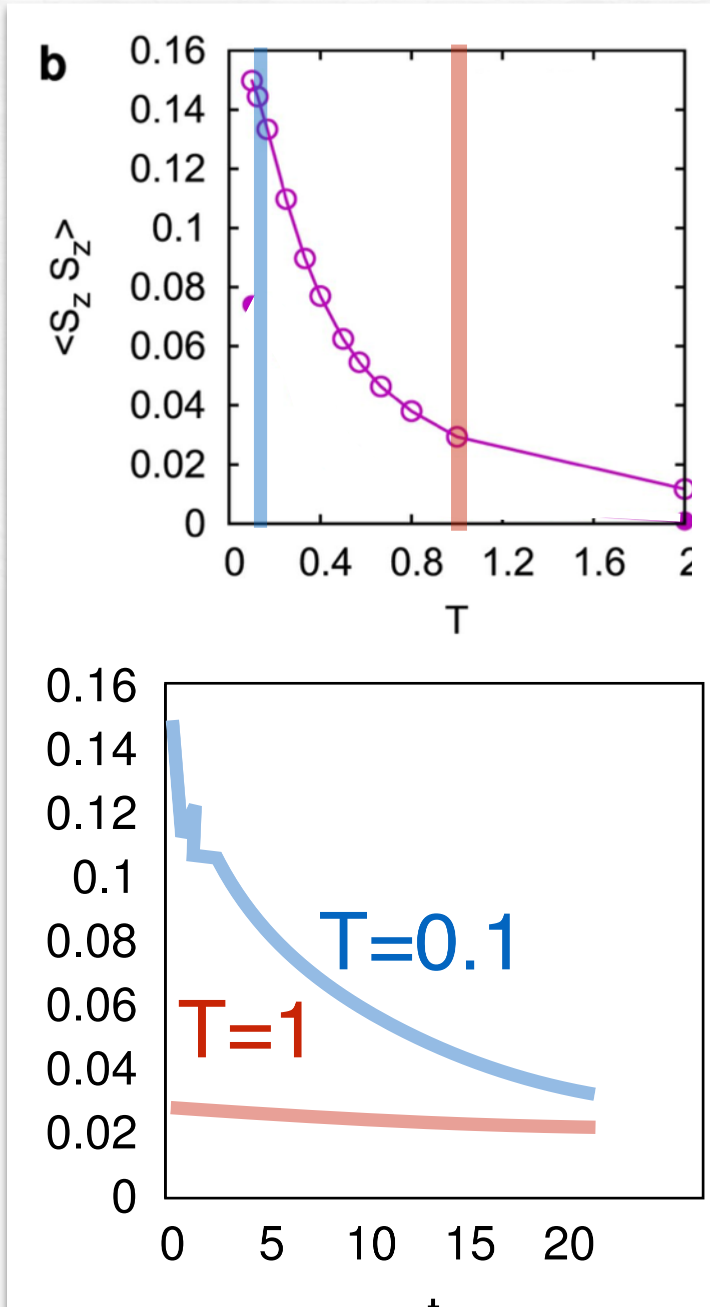
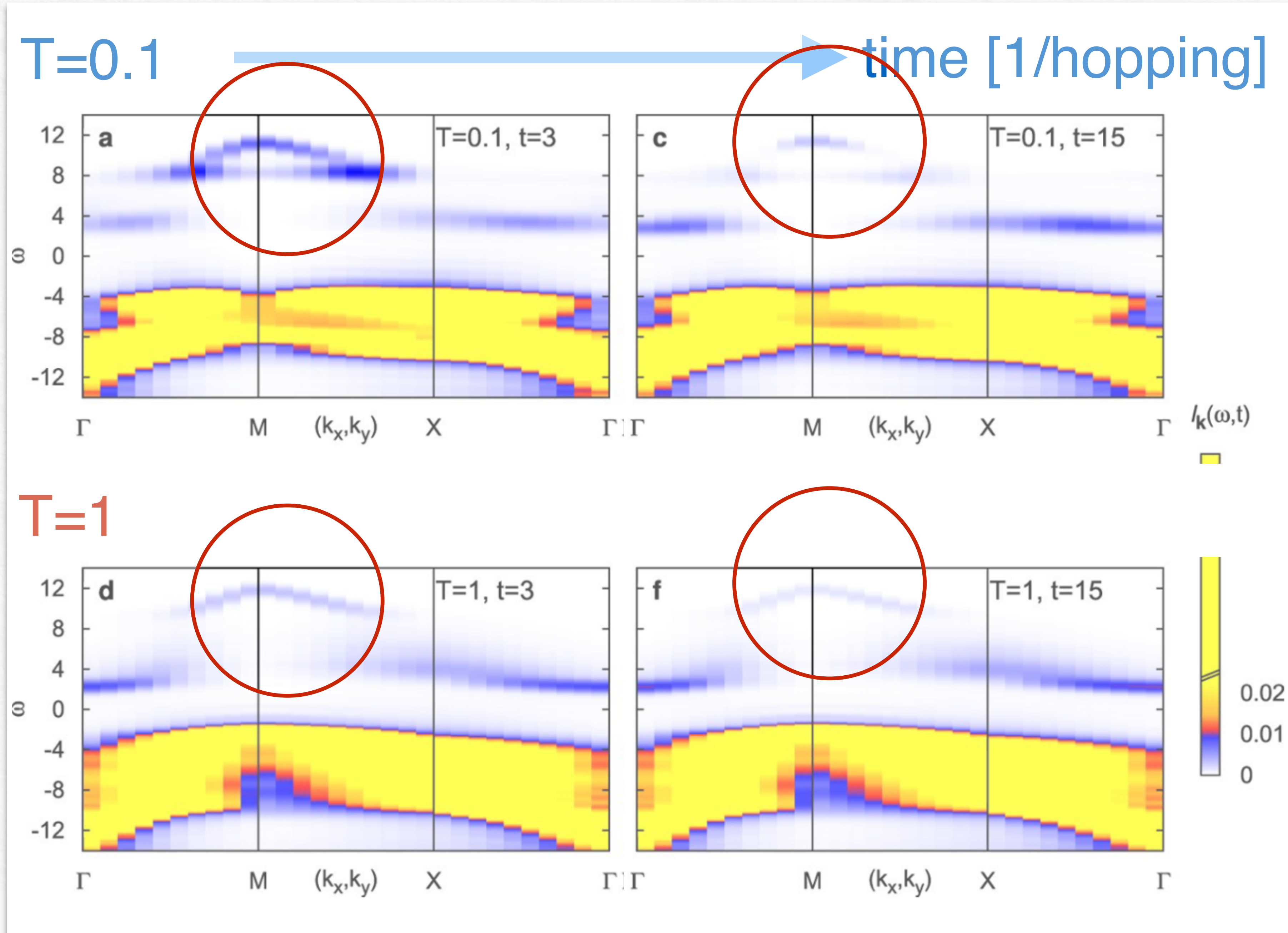
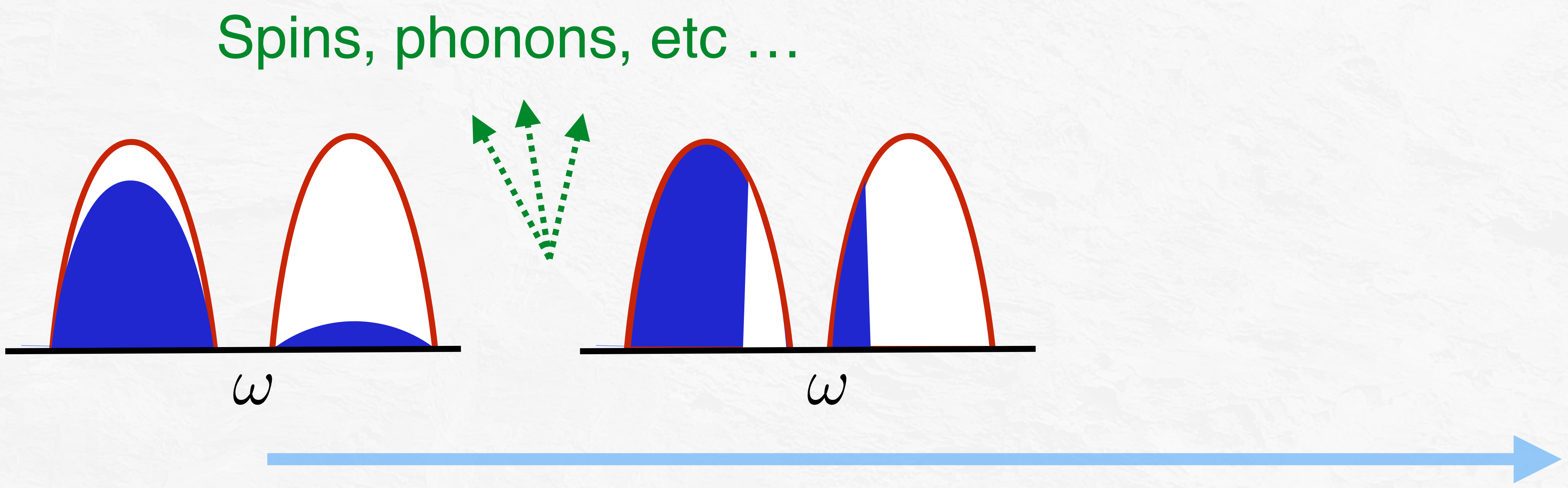




strong spin correlations: inter Hubbard band relaxation within few 1/hopping

Long-lived steady states



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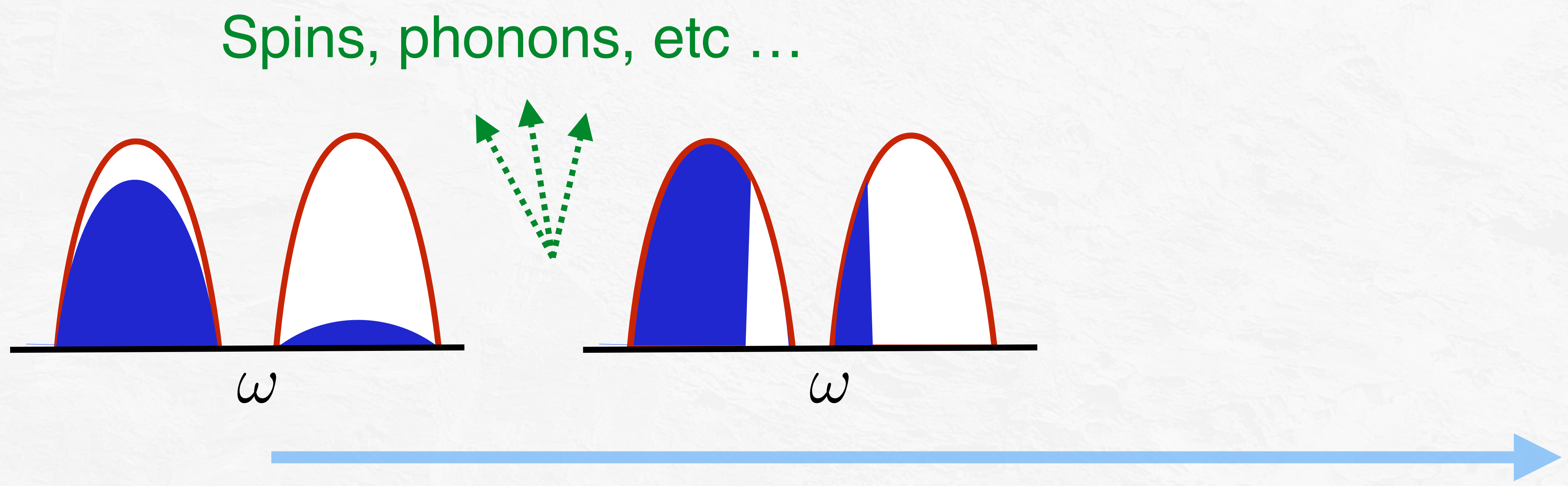
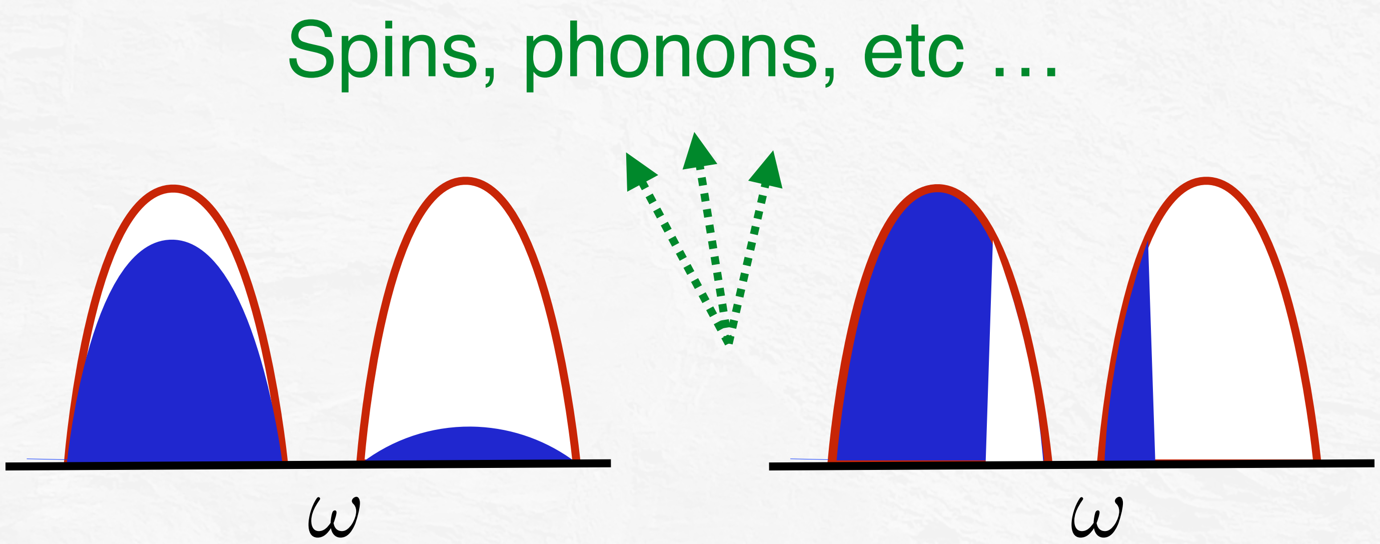


Pre-thermal states

Quasi-steady states due to relaxation bottlenecks and constraints

Long-lived steady states

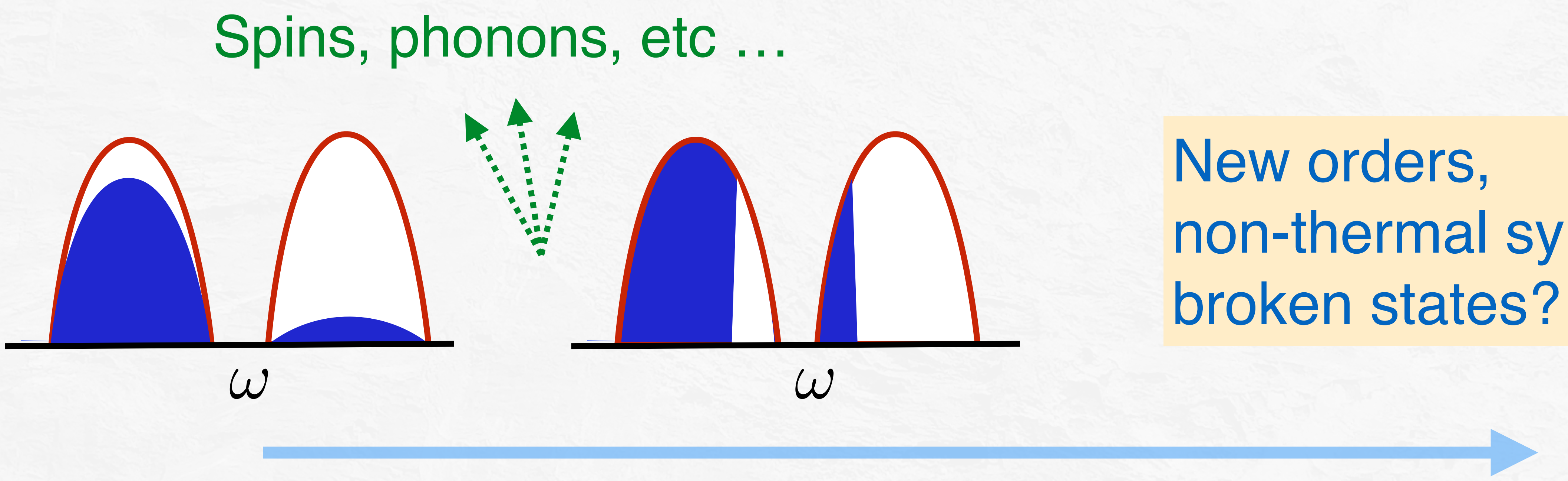


New orders,
non-thermal symmetry
broken states?

Pre-thermal states

Quasi-steady states due to
relaxation bottlenecks and constraints

Long-lived steady states



New orders,
non-thermal symmetry
broken states?

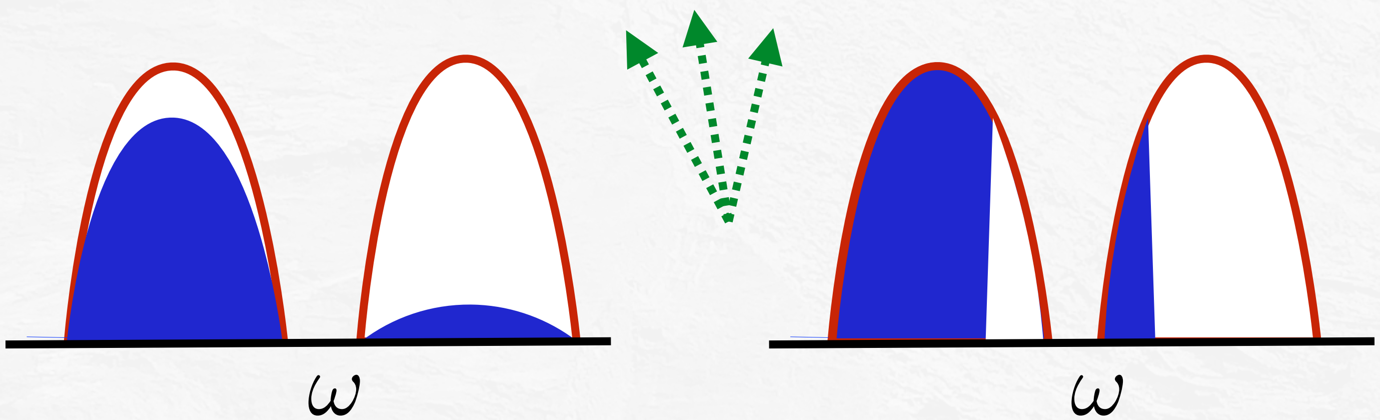
Pre-thermal states

Quasi-steady states due to
relaxation bottlenecks and constraints

⇨ 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

Long-lived steady states

Directly targeting a non-equilibrium steady state

dissipation + continuous driving (bias) ⇨ Non-equilibrium steady state

$$G(t,t') \to G(t-t', t_{av} = (t+t')/2) \to \quad G(\omega) = A(\omega)F(\omega)$$

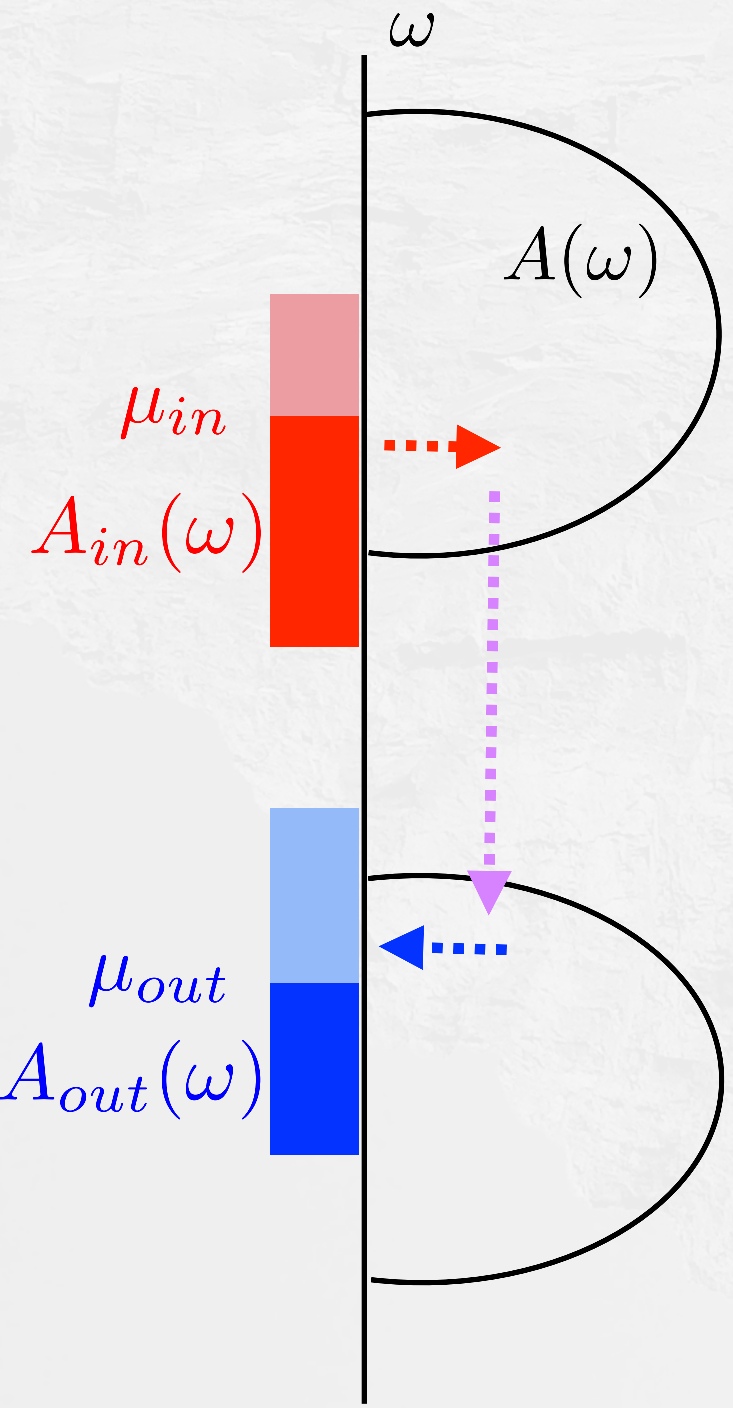
Keldysh formalism:

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

Interaction & dissipation

Photo-doping by bath

Non-equilibrium steady state:



Two bath coupled to every site of the lattice:

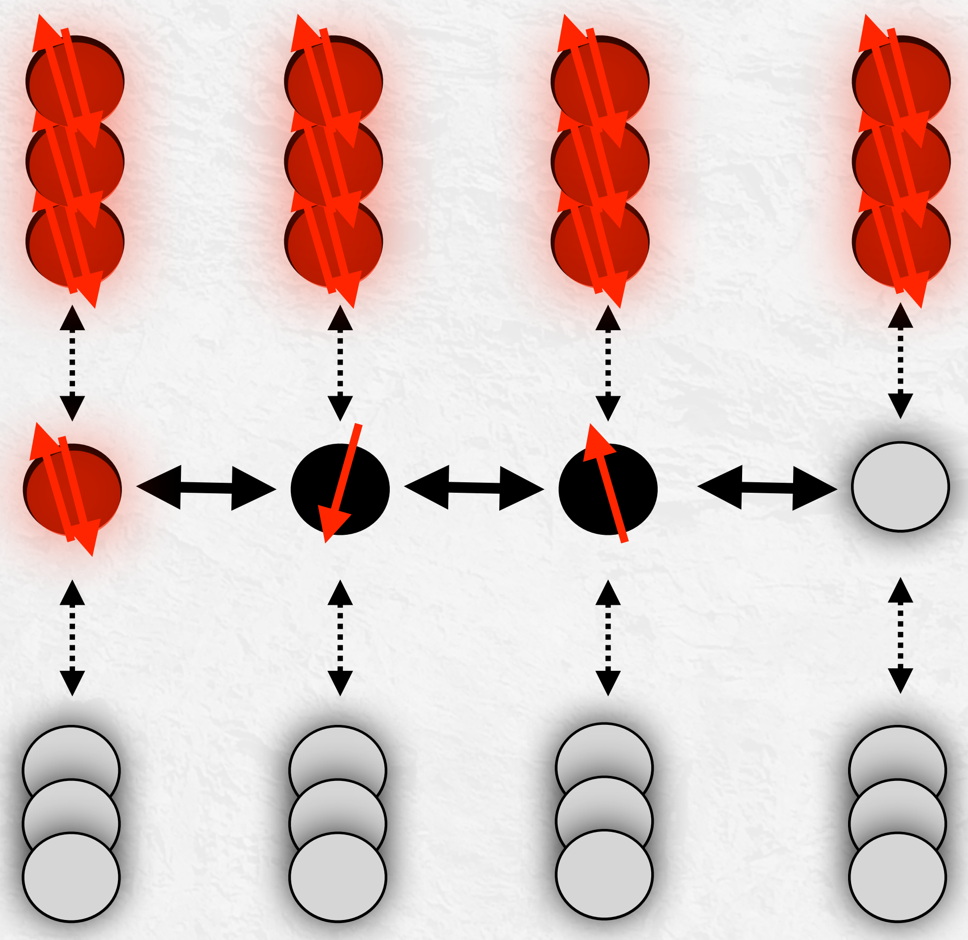
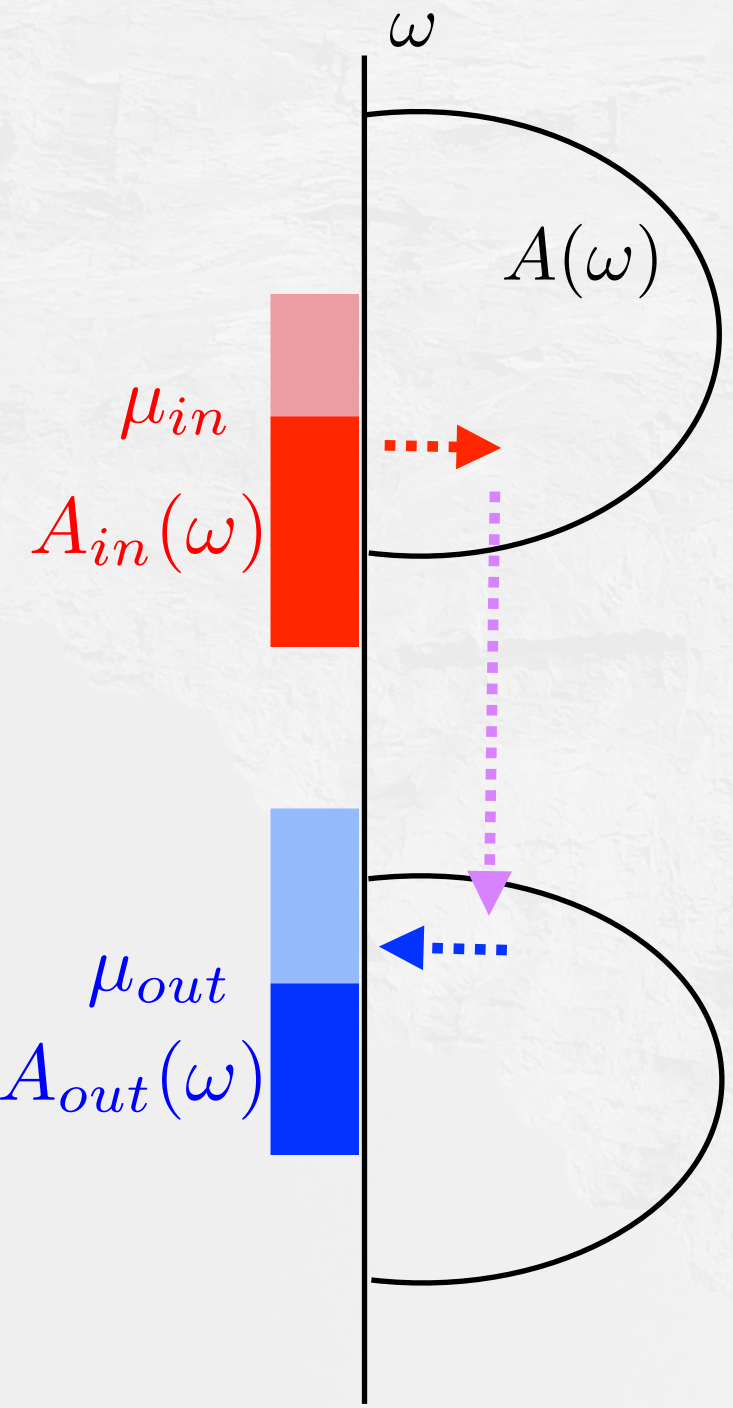


Photo-doping by bath

Non-equilibrium steady state:



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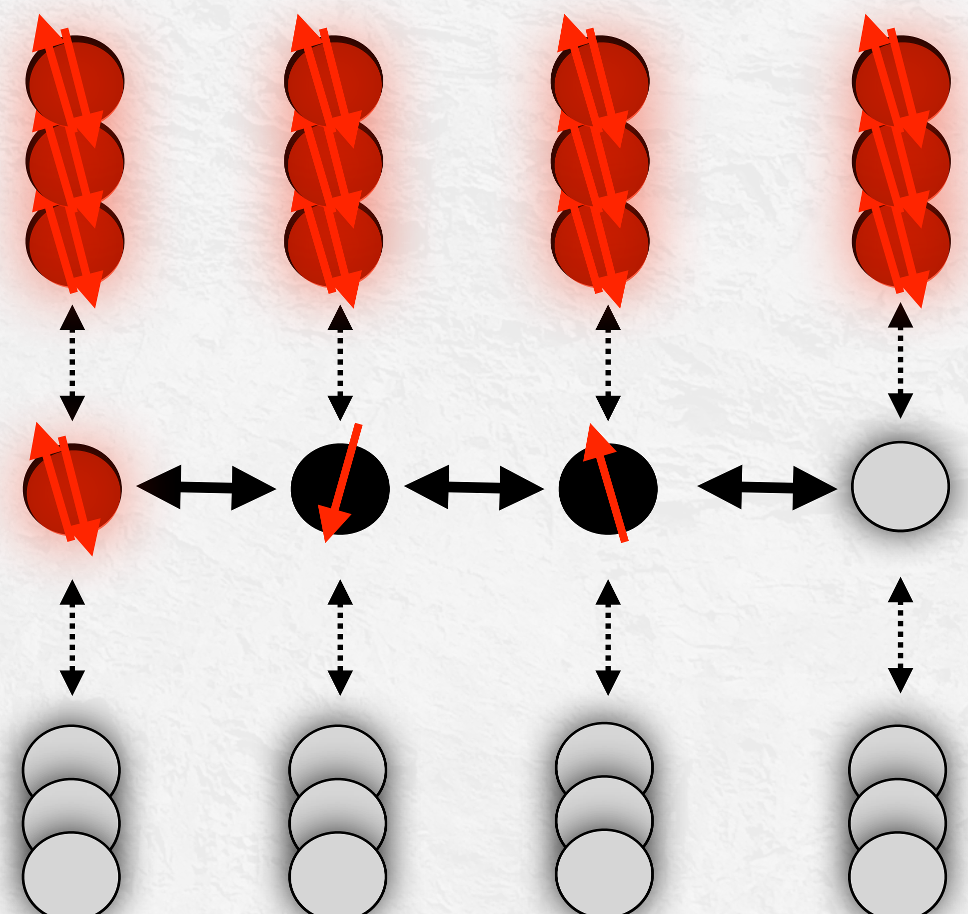


Steady state with occupation in upper band largely independent of bath details if state is controlled by few bottlenecks

Photo-doping by bath

Non-equilibrium steady state:

Two bath coupled to every site of the lattice:

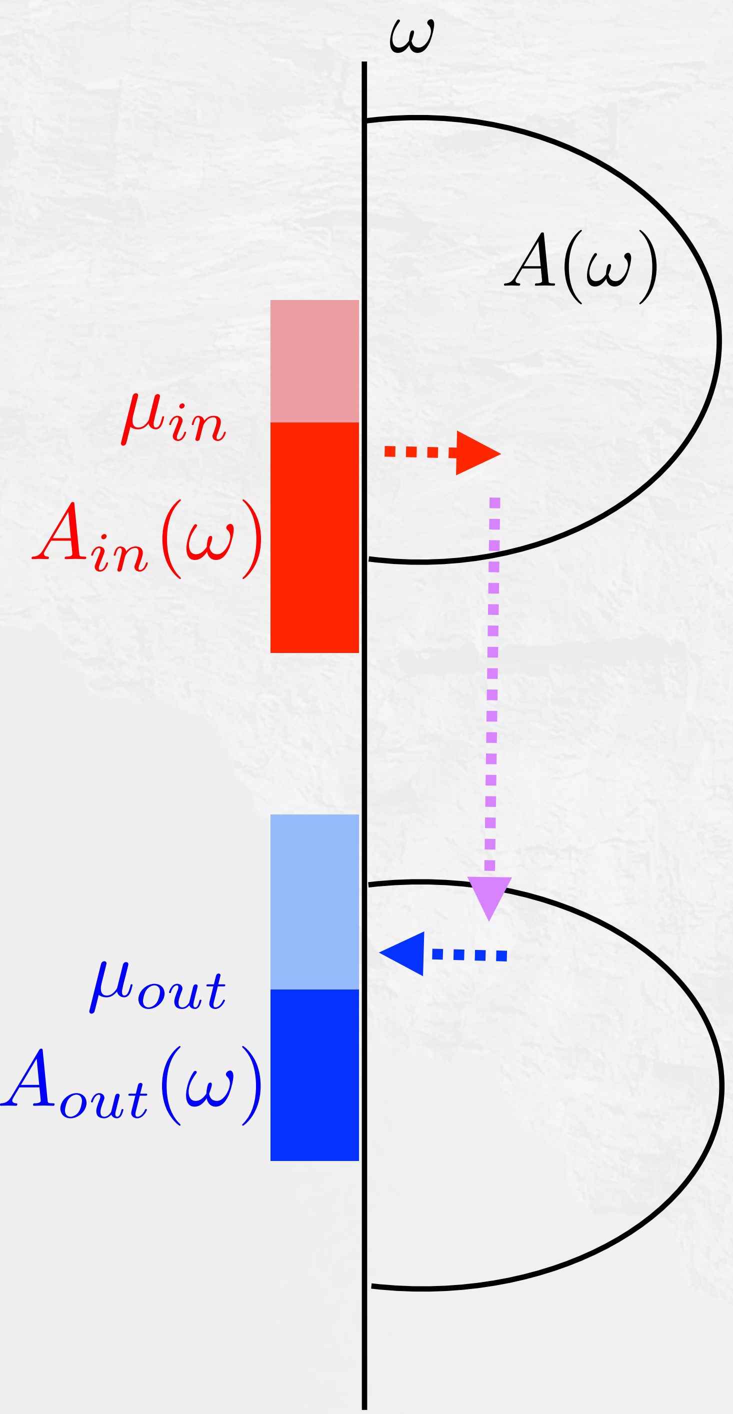
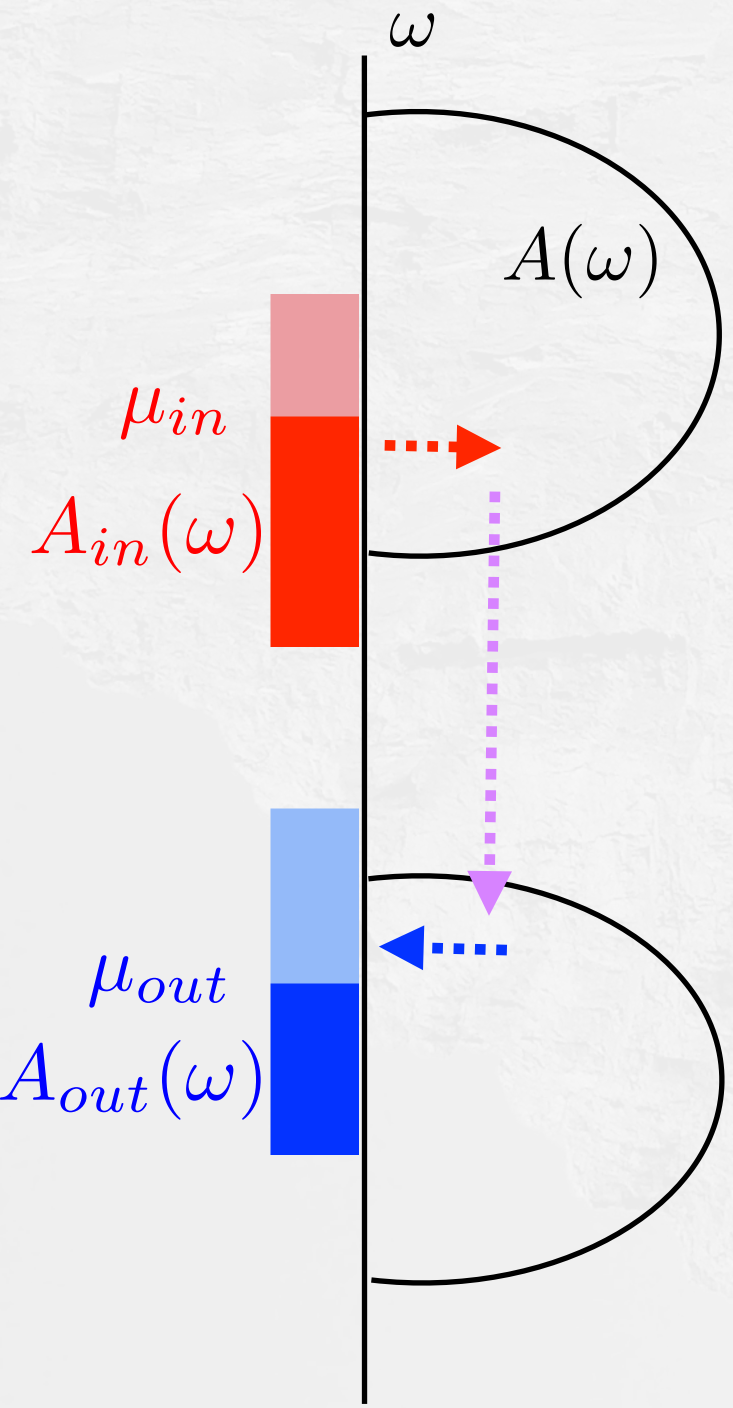


Photo-doping by bath

Non-equilibrium steady state:



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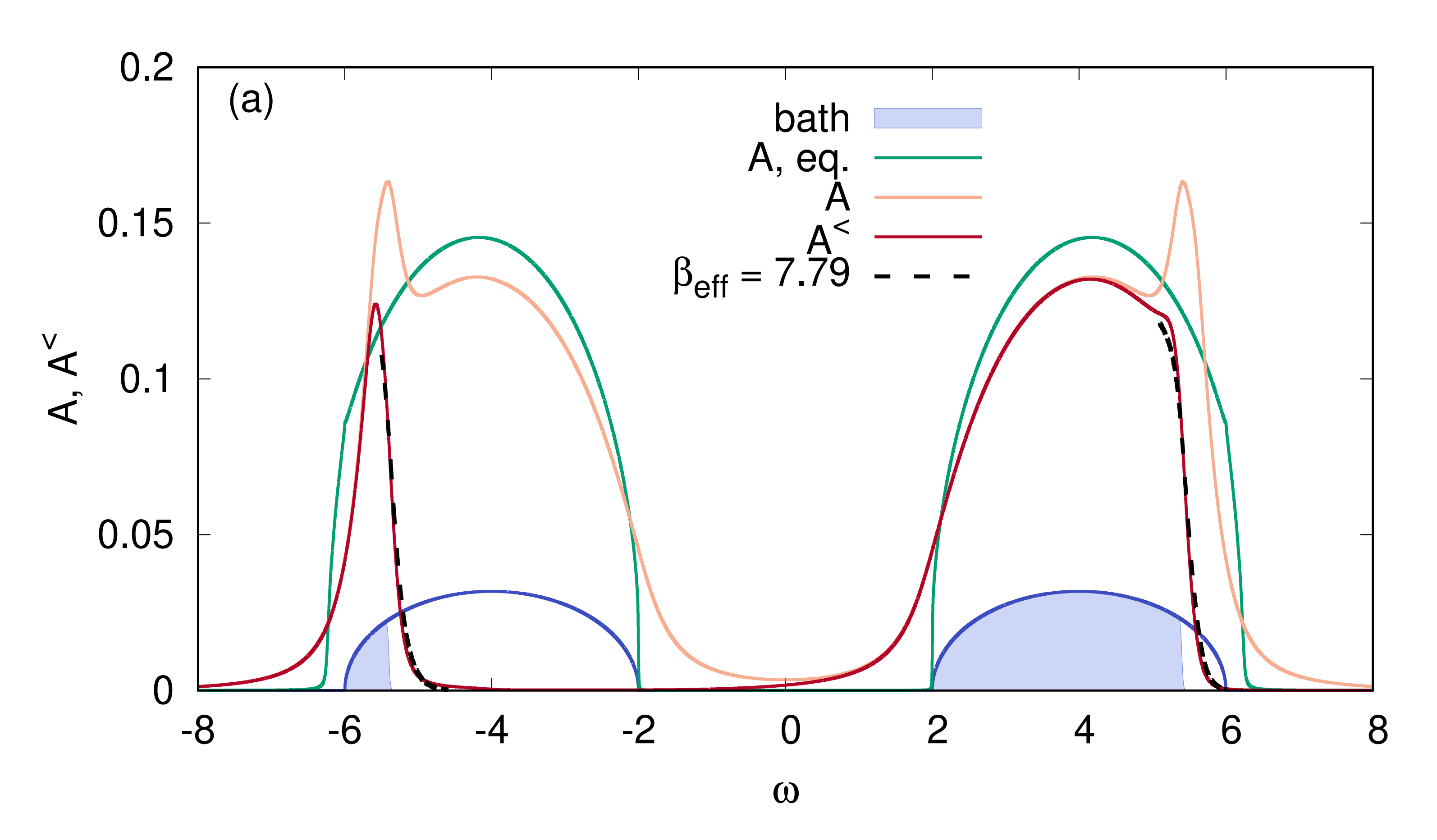
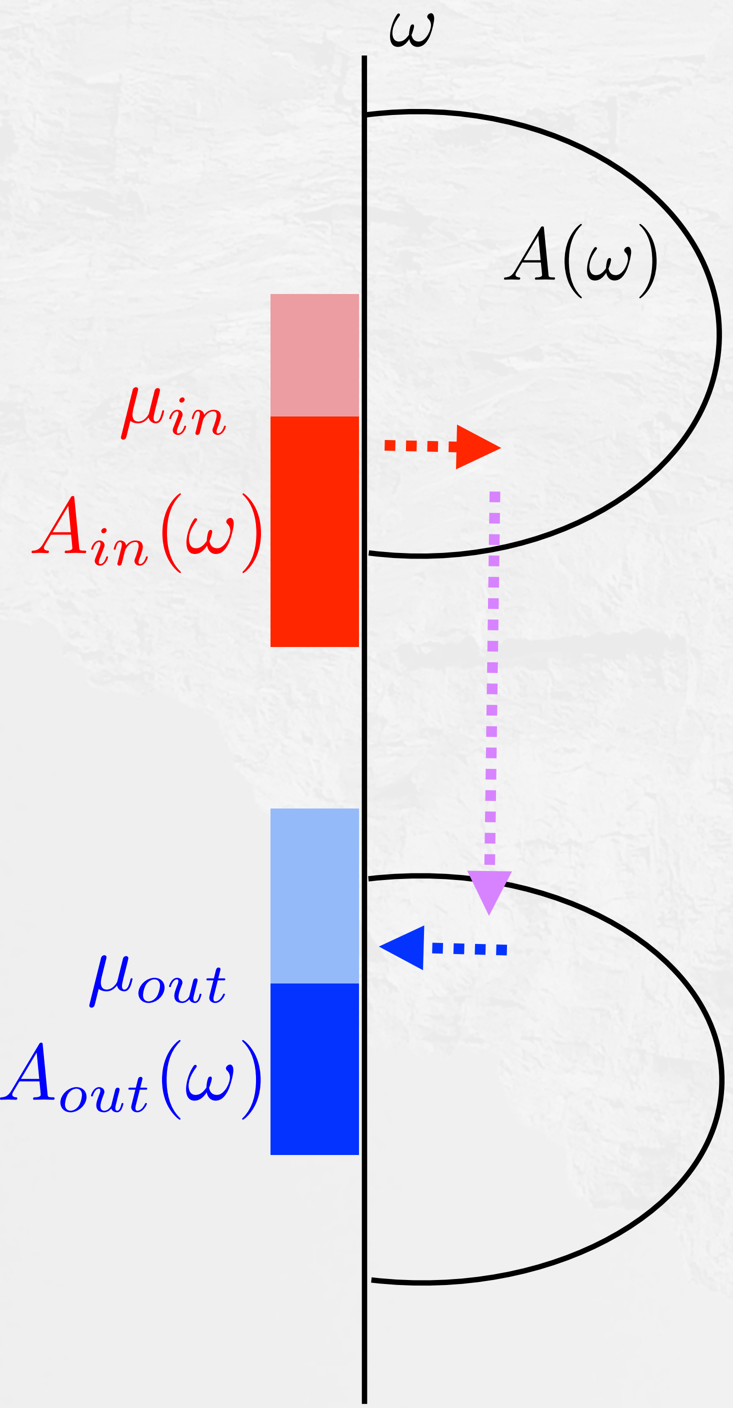


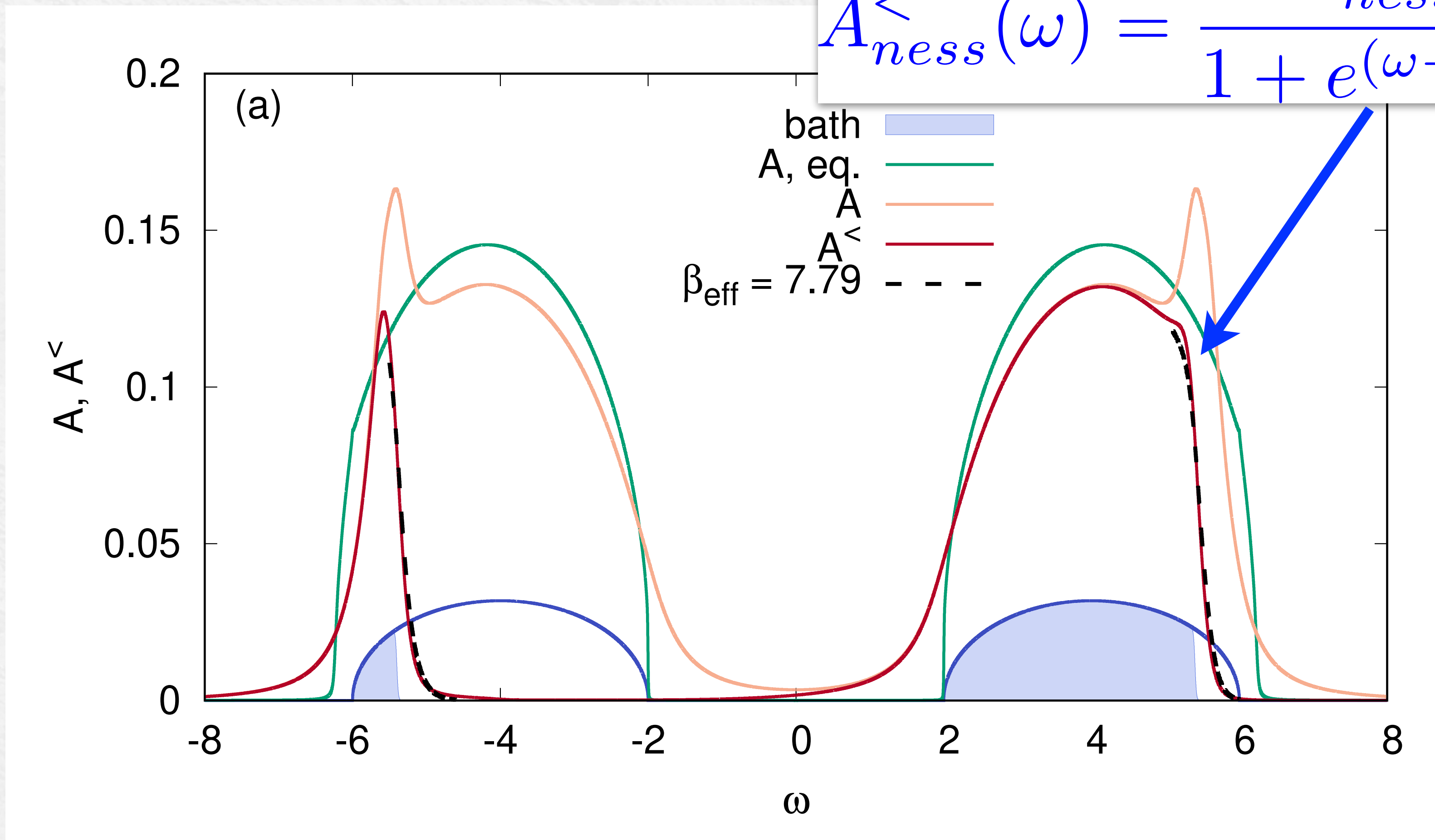
Photo-doping by bath

Non-equilibrium steady state:



Two bath coupled to every site of the lattice:

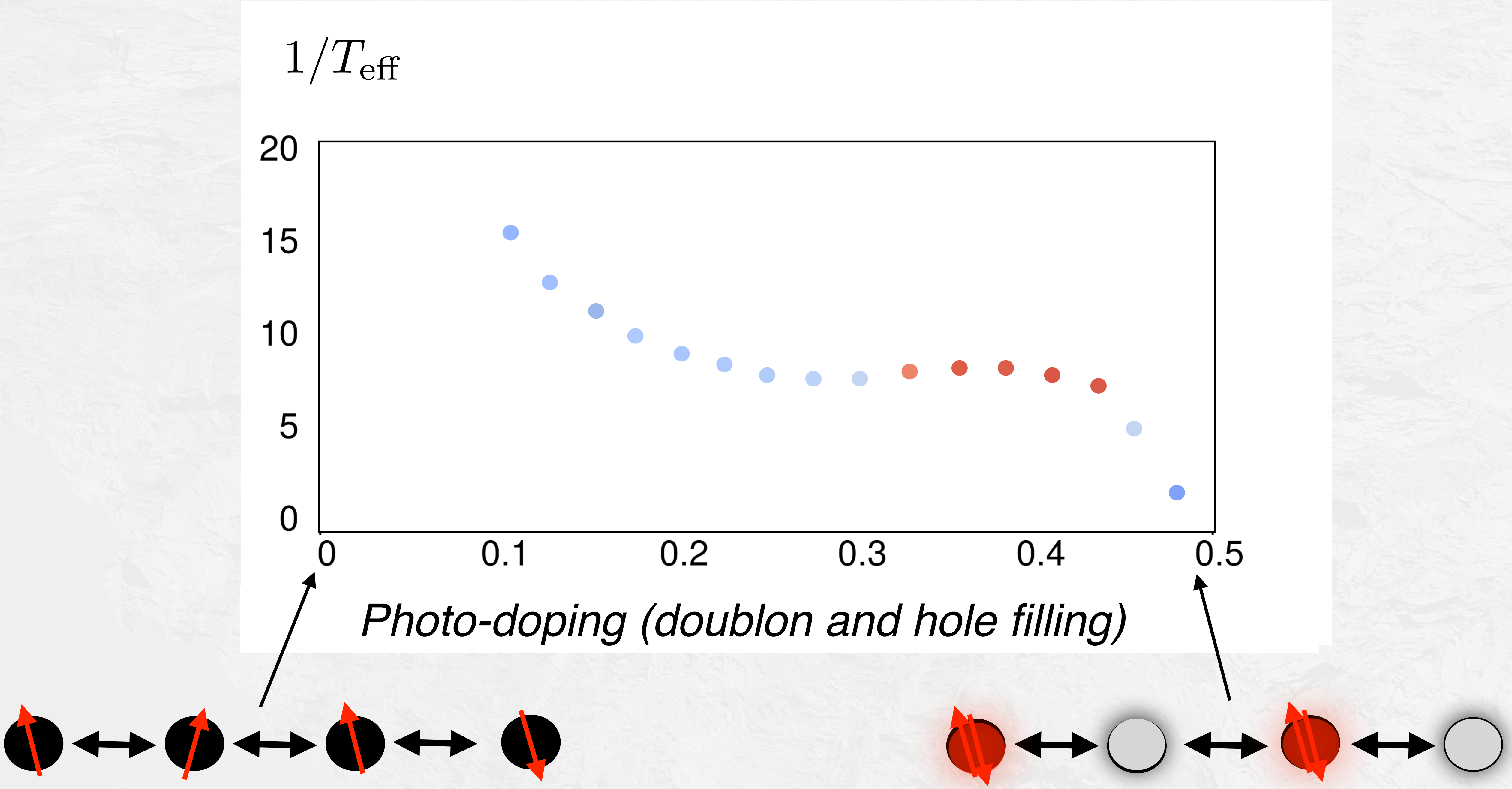
$$A^{<}_{ness}(\omega) = \frac{A_{ness}(\omega)}{1 + e^{(\omega-\mu)/T_{\mathrm{eff}}}}$$



State characterized by two (self-consistently determined) state variables: Temperature, double occupancy (photo-doping)

Phase-diagram of photodoped system

Scan for different μ_{bath}:



Phase-diagram of photodoped system

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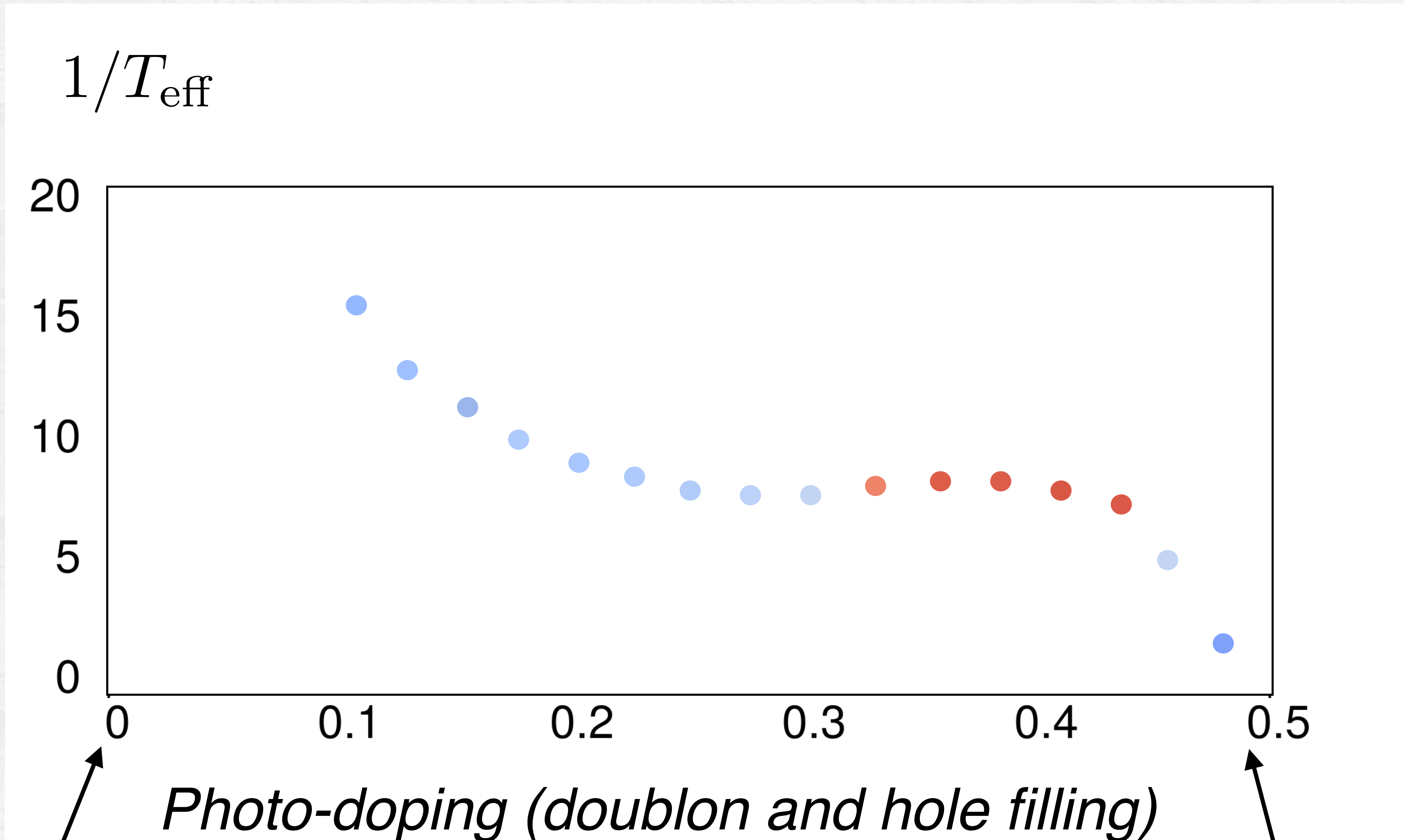
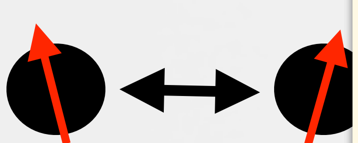
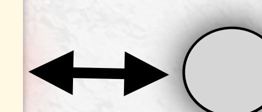




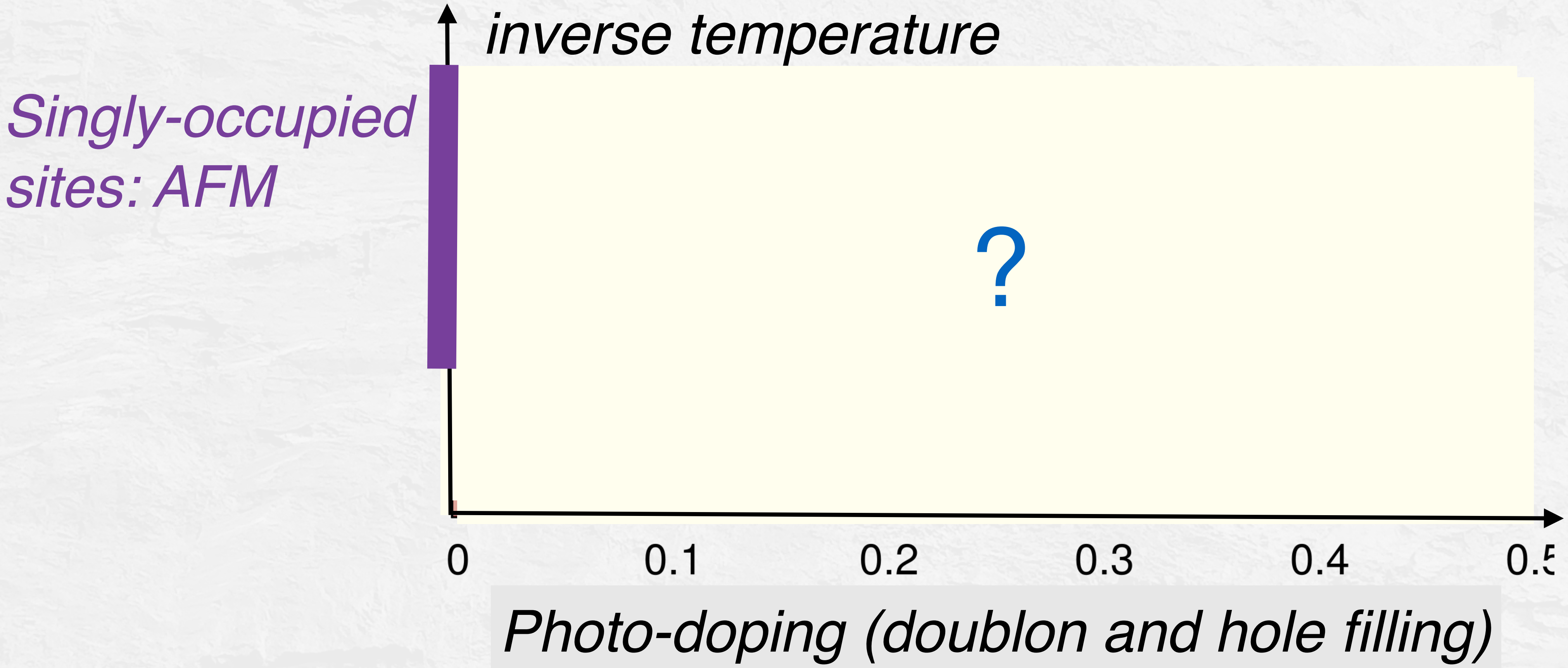
Instabilities?

AFM, CDW, SC, …

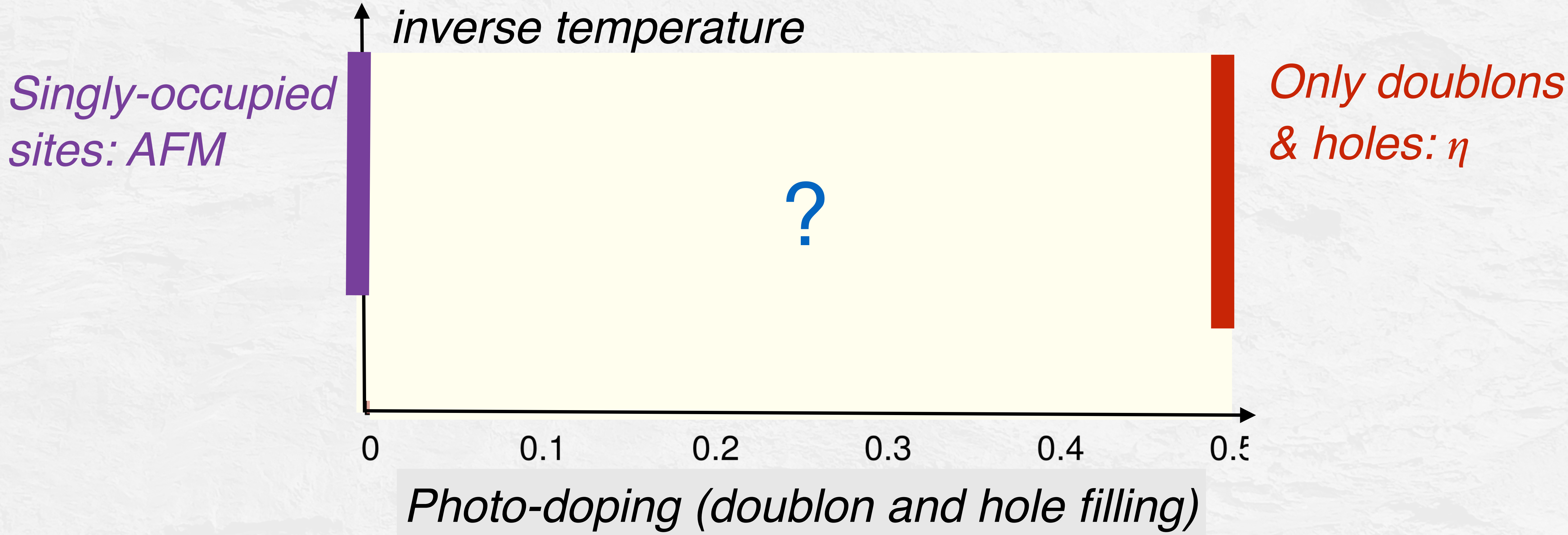
$$H = H_{\text{System+Bath}} + h_\eta \sum_j \eta_j^x \qquad \chi_\eta = \frac{\langle \eta_j^x \rangle_{\text{ness}}}{h_\eta}\Big|_{h\to 0}$$



η-pairing superconductivity



η-pairing superconductivity



Large U limit: only doubly occupied/empty sites

SU(2)c symmetry

$$H^{\mathrm{eff}} = \sum_{\langle ij \rangle} J_{\mathrm{ex}} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \cdots$$

Long-range η-pairing: $\langle \eta_j^+ \rangle \equiv \Delta$

⇨ Staggered phase Superconductivity

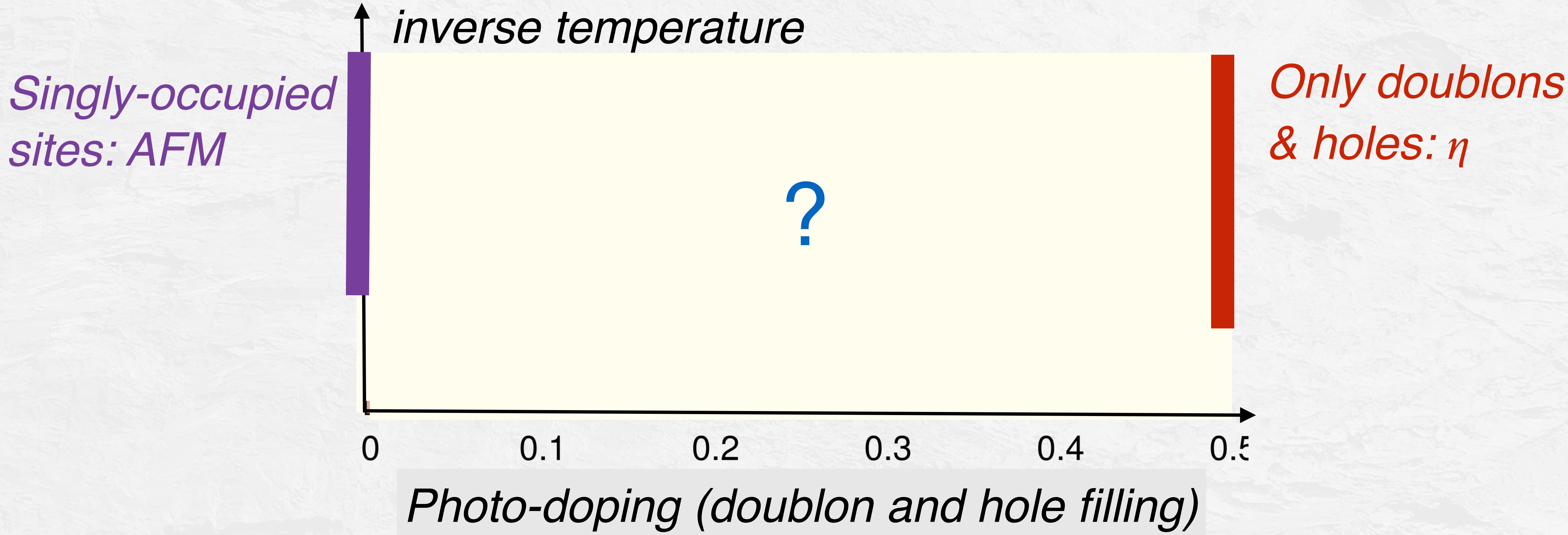
(exact excited state on bipartite lattice,

Yang, PRL **63**, 2144 (1989)

$$\eta_j^+ = (-1)^j c_{j\uparrow}^\dagger c_{j\downarrow}^\dagger$$
$$\eta_j^- = (-1)^j c_{j\downarrow} c_{j\uparrow}$$
$$\eta_j^z = \frac{1}{2}(n_j - 1)$$

$$\begin{pmatrix} |\uparrow\downarrow\rangle \\ |0\rangle \end{pmatrix}$$

η-pairing superconductivity

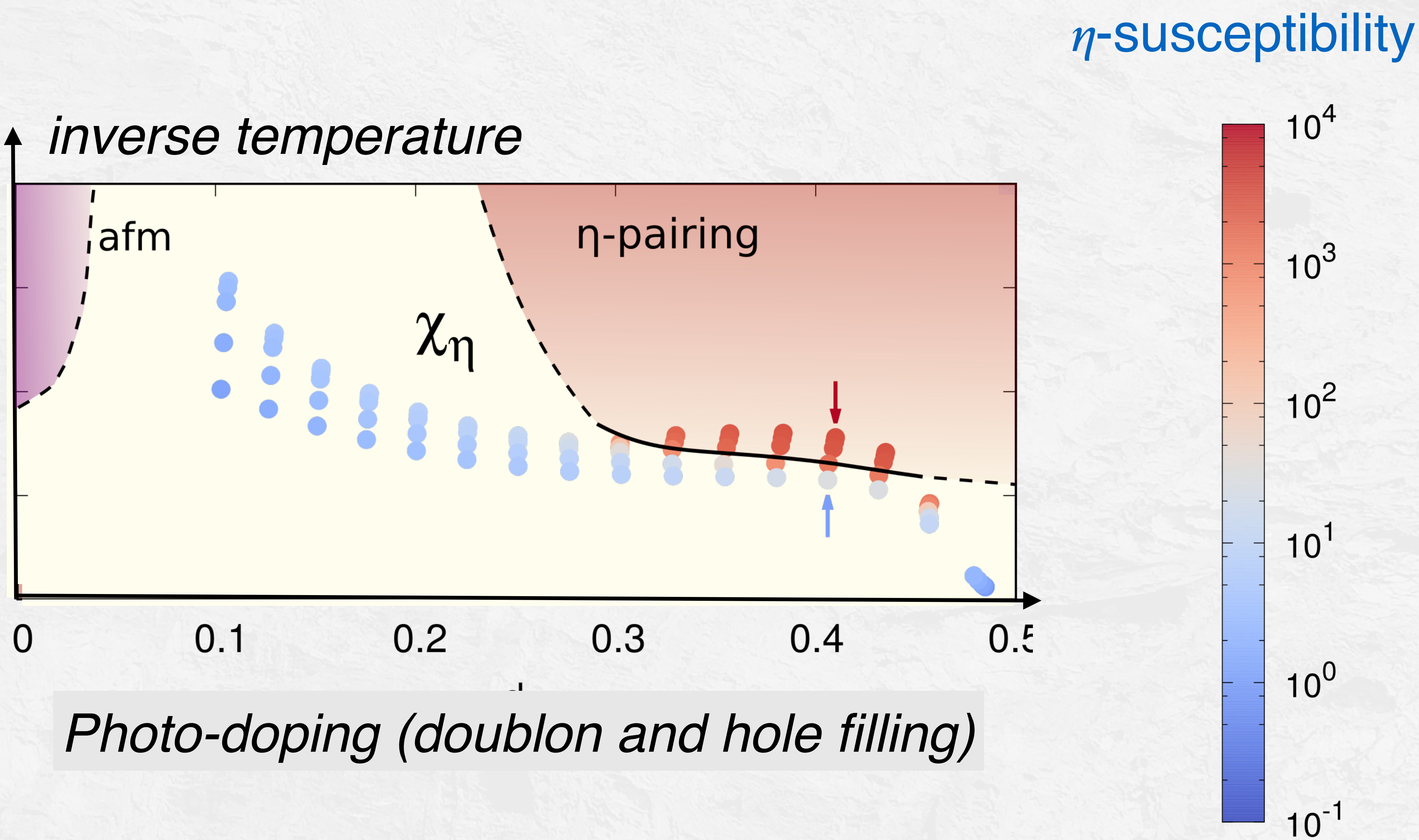


⇨ photo-excitation to populate doublons?

finite size (ED): Enhancing η-correlations Kaneko et al, 2018,2019

Pumping doublons d>1/4 by driving Peronacci, Schiro, Parcollet 2019

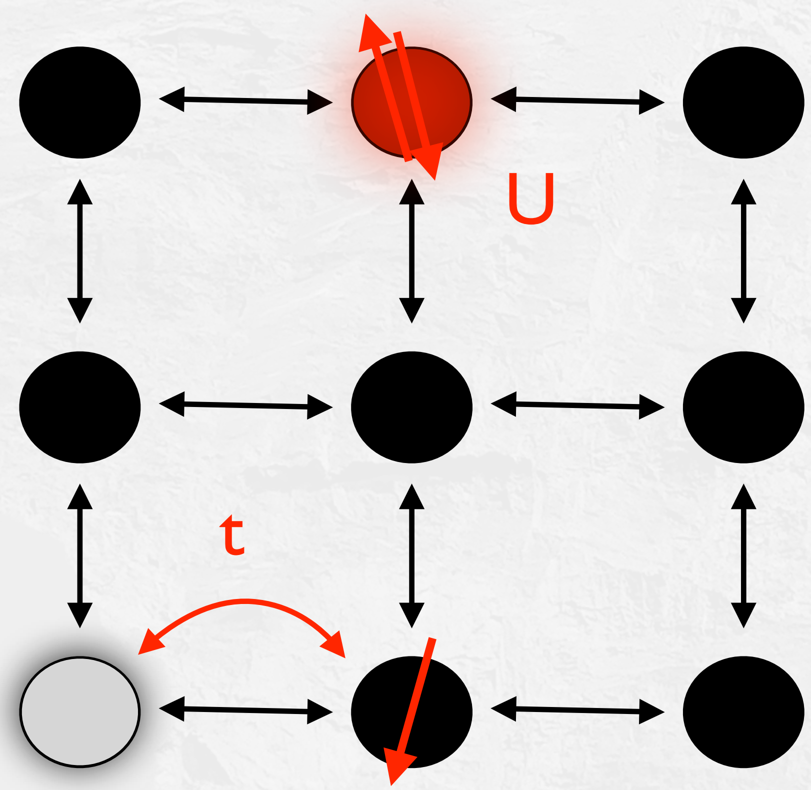
η-pairing superconductivity

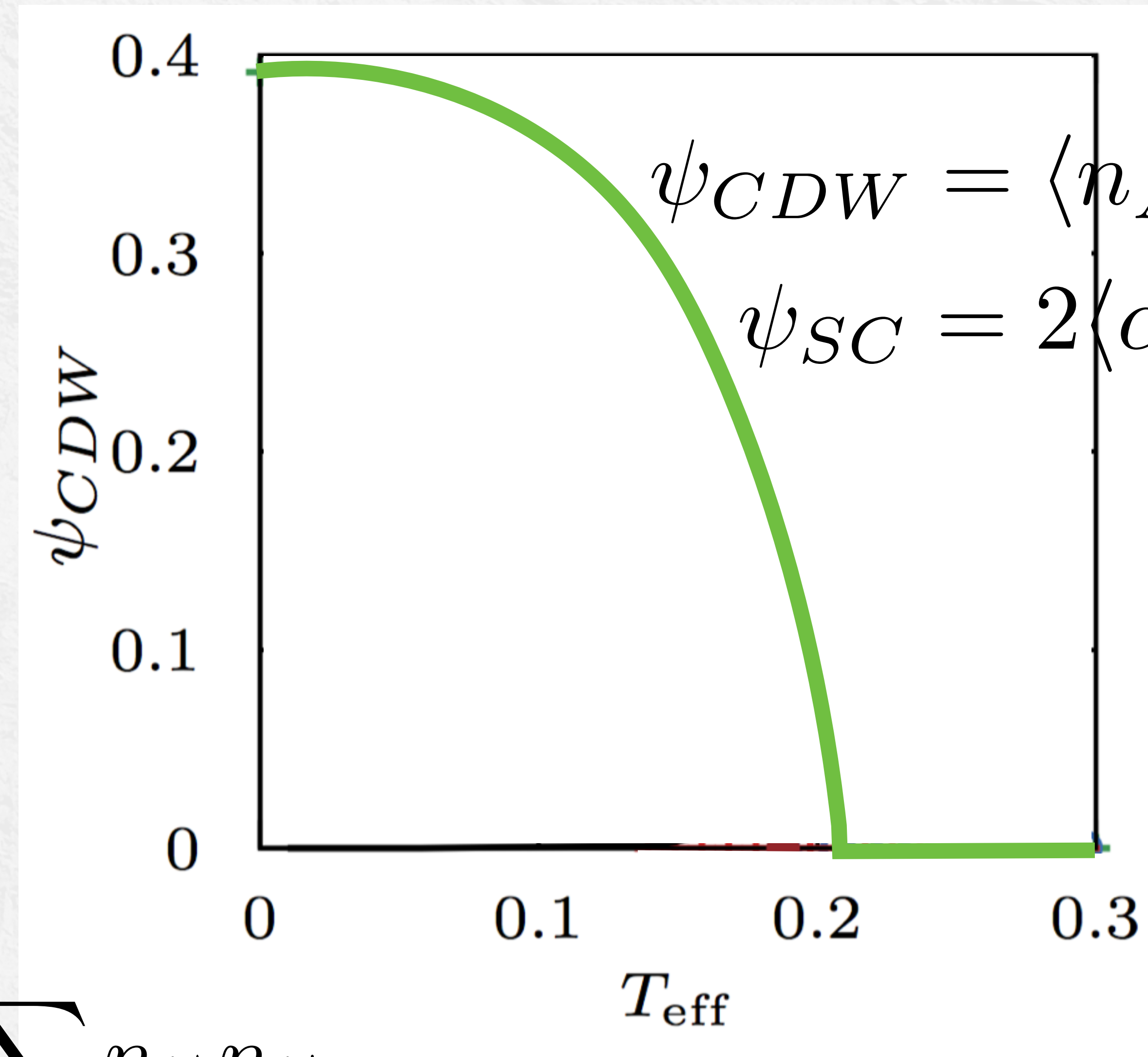


Extended region of non-thermal superconductivity in the Hubbard model a real steady state

CDW in attractive Hubbard model

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

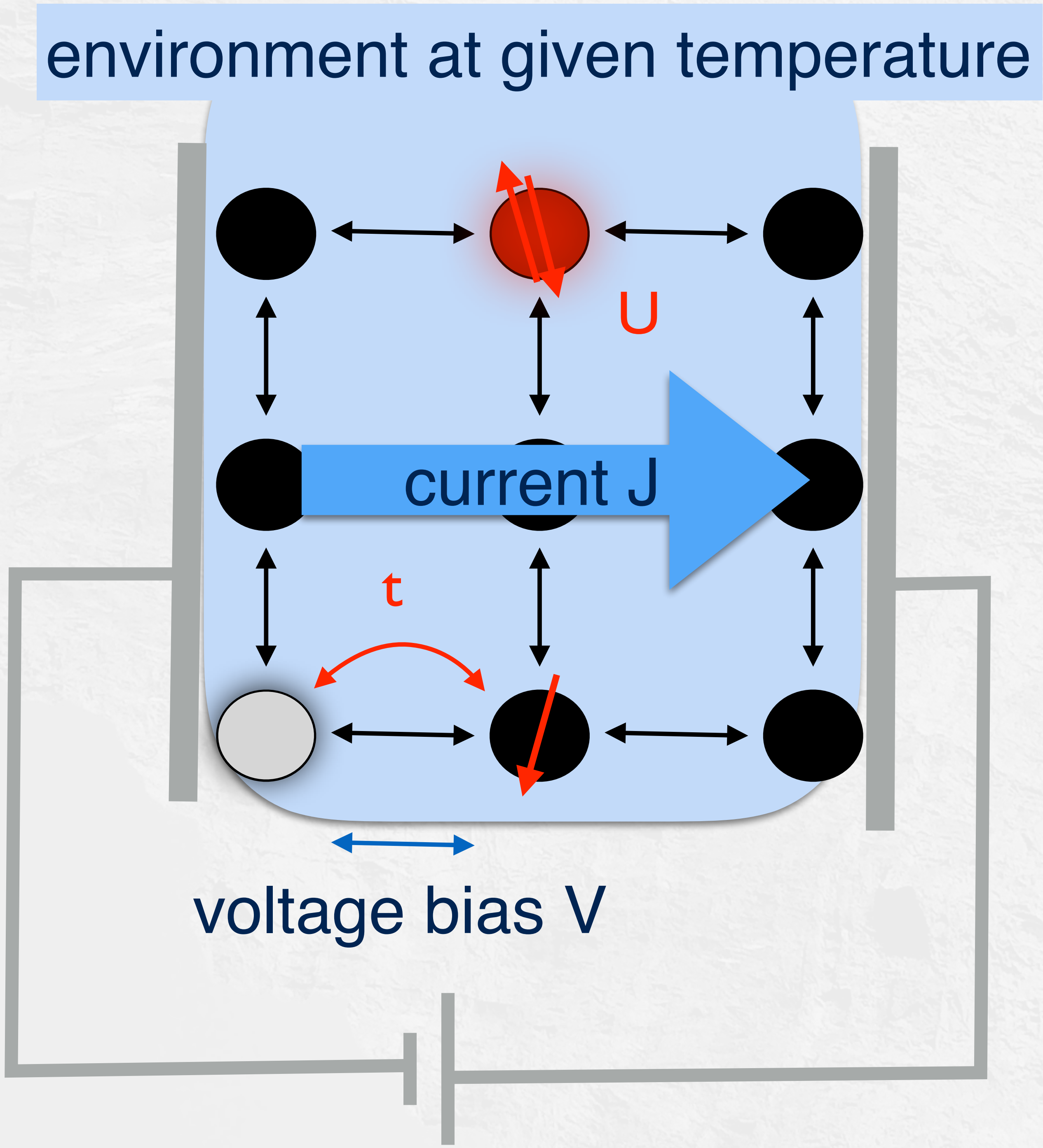




$$H = -t \sum_{\langle ij \rangle, \sigma=\uparrow,\downarrow} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

Can one stabilize superconductivity under non-equilibrium conditions?

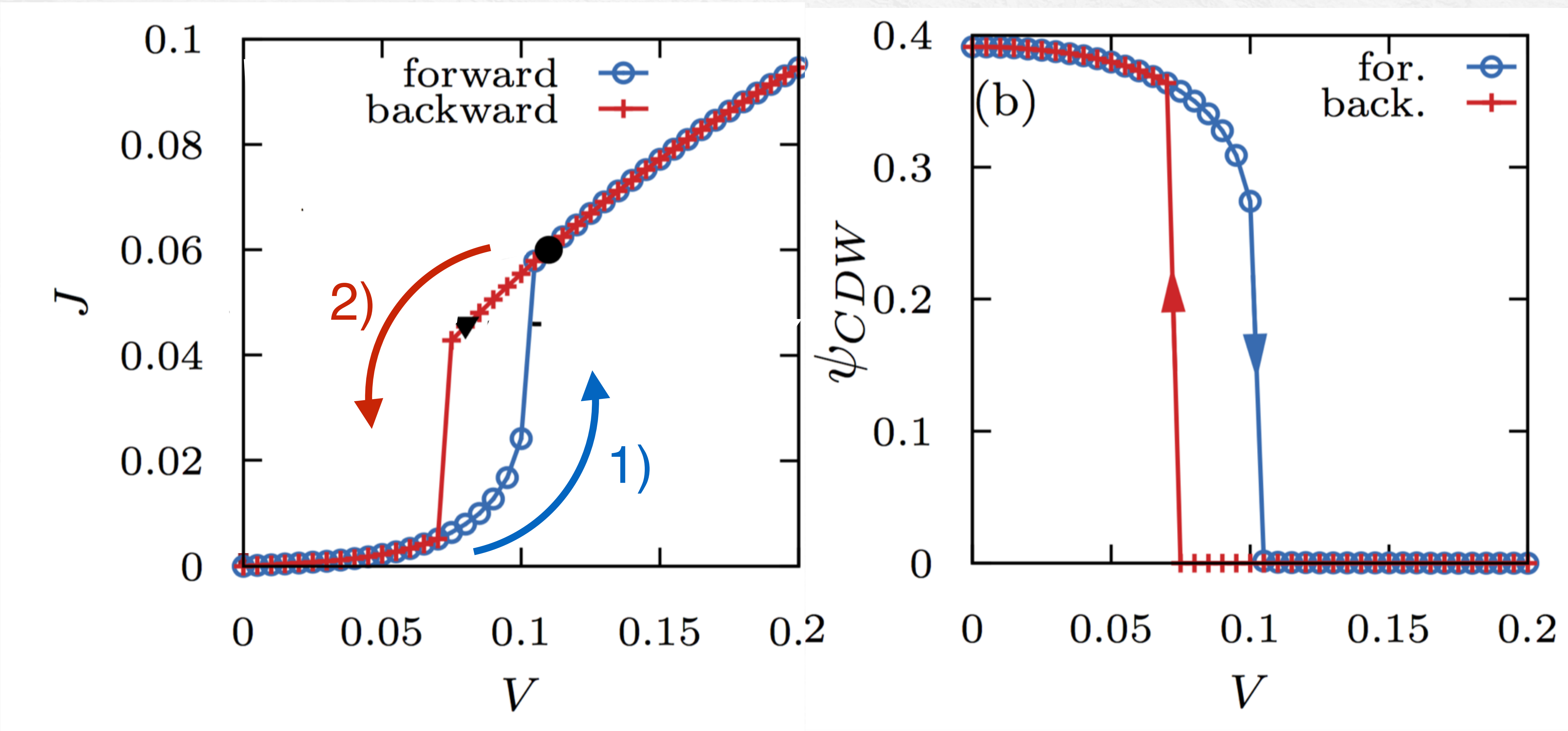
Model Hamiltonians



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

I-V Characteristics

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



Coexisting steady state solutions ⇨ first-order transition

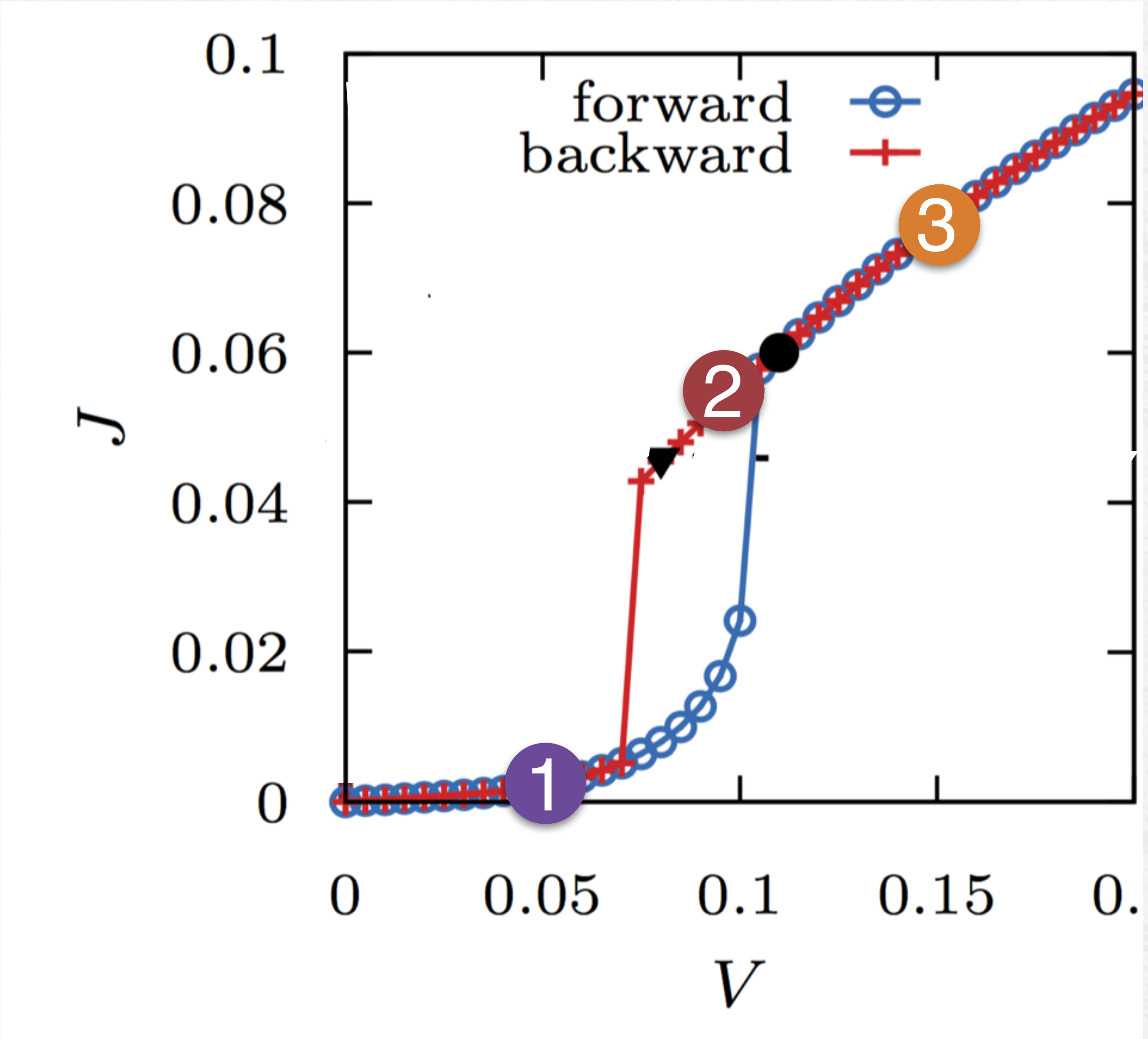
1) Melting of CDW through charge excitations

2) Metal unstable against infinitesimal CDW fluctuations:

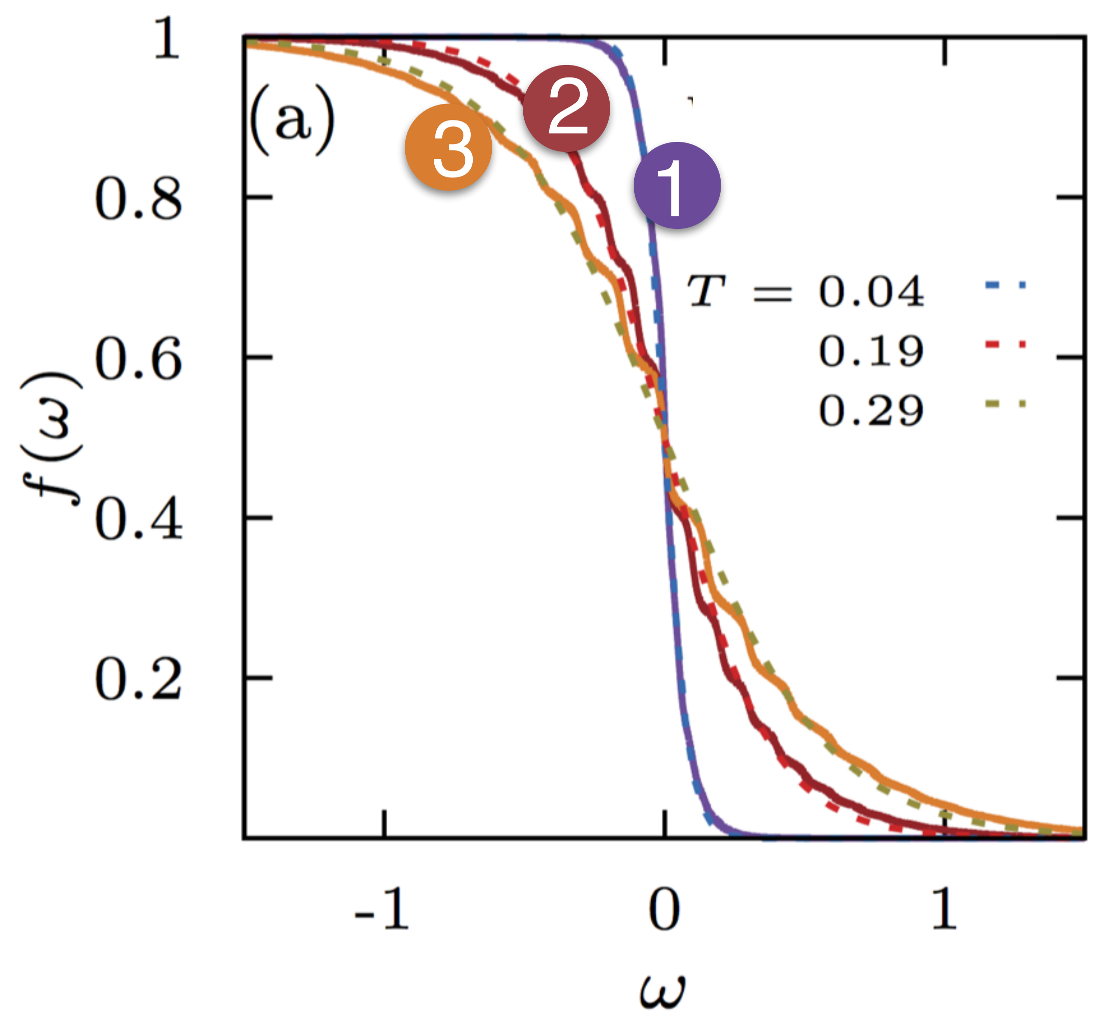
⇨ close to thermal mechanism?

I-V Characteristics

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



Distribution functions:



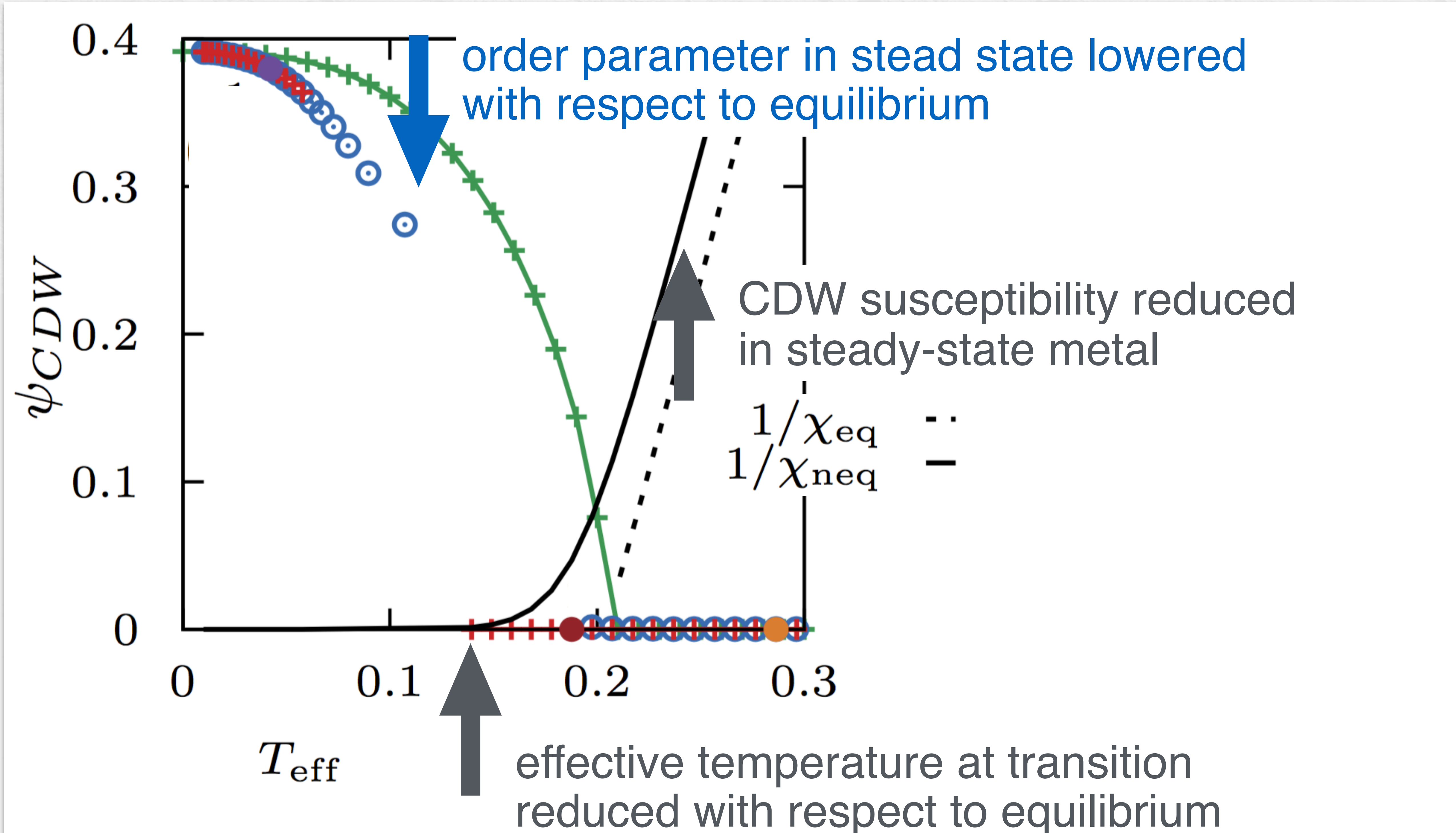
Only slight deviations from Fermi function (due to el.-el. scattering)

⇨ in the following analysis, first use T_{eff} to characterize steady state

I-V Characteristics

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

CDW in steady state as function of effective temperature

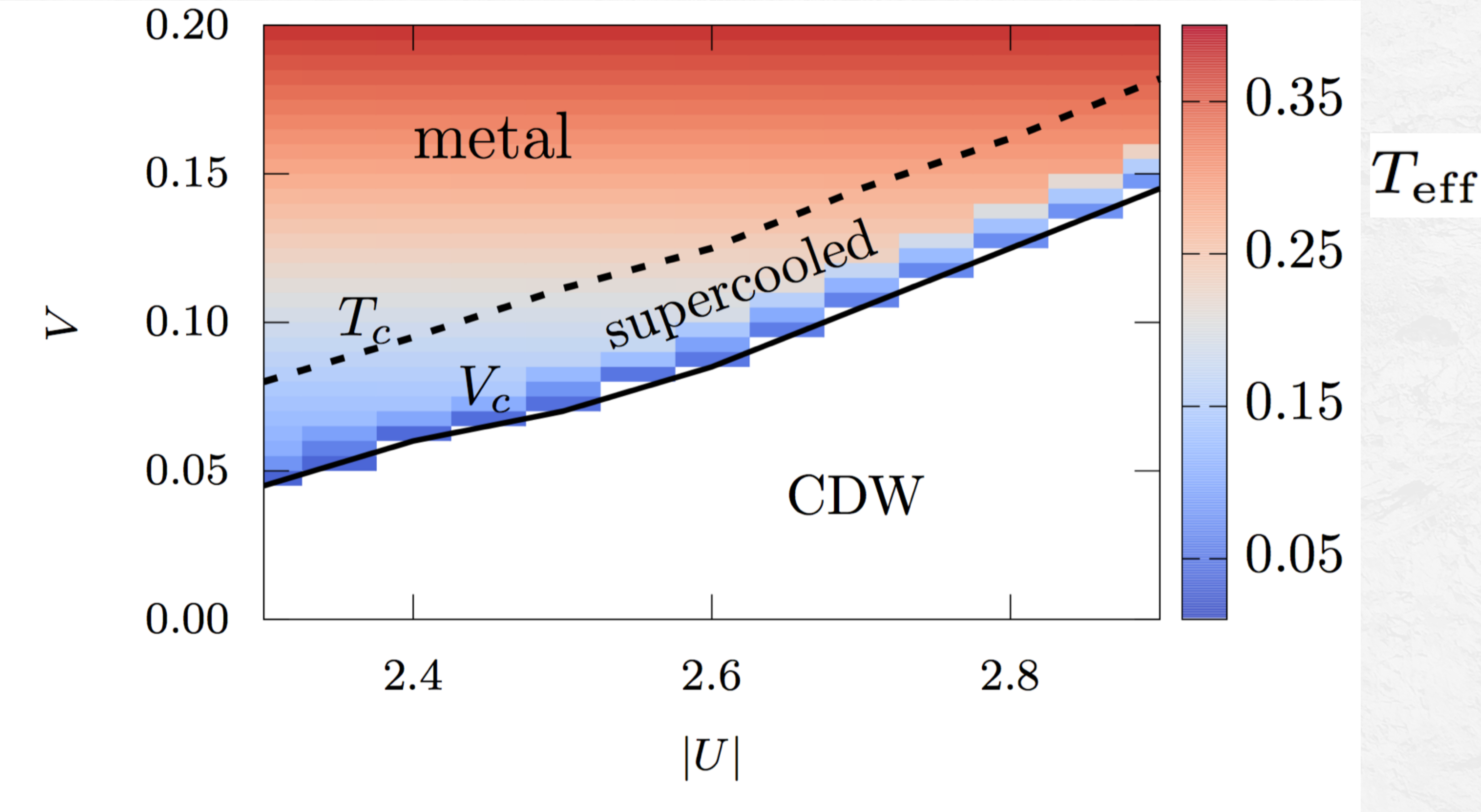


Non-equilibrium phase diagram

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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