Electronic structure of correlated materials out of equilibrium: non-equilibrium dynamical mean-field theory

Lecture 4: Photo-doping in Mott insulators from a theory perspective

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Strong-coupling expansion
& some words on CTQMC

Strong-coupling expansion

Expansion in the coupling to the bath?

$$\langle \mathcal{O}(t) \rangle = \frac{1}{Z} \mathrm{tr}\Big[T_{\mathcal{C}} e^{-i\int_{\mathcal{C}} dt' H_{loc}(t')} e^{-i\int_{\mathcal{C}} dt_1 dt_2 c^\dagger(t_1)\Delta(t_1,t_2)c(t_2)} \mathcal{O}(t) \Big]$$

0th order is interacting Hamiltonian H_{loc} $\Rightarrow$ very general starting point

But: no Wick's theorem → Resolvent expansions:

- Perturbative: "non-crossing approximation", first developed for the Kondo model

  Keiter & Kimball '71; Kuramoto '83; Grewe '83; Pruschke & Grewe 89
  Bickers, Cox & Wilkins '87; Coleman '83; Haule, Kirchner, Kroha & Wölfle '01

- Hybridization Quantum Monte Carlo: Werner *et al.*, 2006
  Stochastic resummation of perturbation series

Strong-coupling expansion

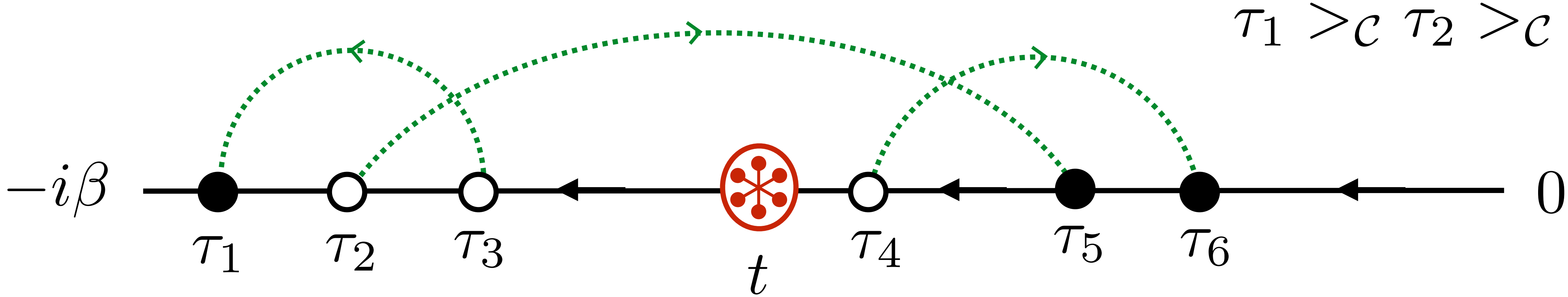
$$\langle \mathcal{O}(t)\rangle = \frac{1}{Z}\mathrm{tr}\big[T_{\mathcal{C}} e^{-i\int_{\mathcal{C}} dt' H_{loc}(t')} e^{-i\int_{\mathcal{C}} dt_1 dt_2 c^\dagger(t_1)\Delta(t_1,t_2)c(t_2)}\mathcal{O}(t)\big]$$

$$= \frac{1}{Z}\sum_{n=0}^{\infty}\frac{(-i)^n}{n!}\int_{\mathcal{C}} dt_1 dt_1' \cdots dt_n dt_n' \; \Delta(t_1,t_1')\cdots\Delta(t_n,t_n') \times$$

$$\times \; \mathrm{tr}\big[T_{\mathcal{C}} e^{-i\int_{\mathcal{C}} dt' H_{loc}(t')} c^\dagger(t_1)c(t_1')\cdots c^\dagger(t_n)c(t_n')\mathcal{O}(t)\big]$$

Sum of all possible contributions like this:

contour ordered $\tau_1 >_{\mathcal{C}} \tau_2 >_{\mathcal{C}} \ldots$



$= \mathcal{O}$ $\quad \bullet = c^\dagger \quad \circ = c \quad t' \dashrightarrow t = \Delta(t,t')$

$t' \longrightarrow t \; = g(t,t') = T_{\mathcal{C}} e^{-i\int_{t'}^{t} ds H_{loc}(s)}$

matrices in local many-body basis

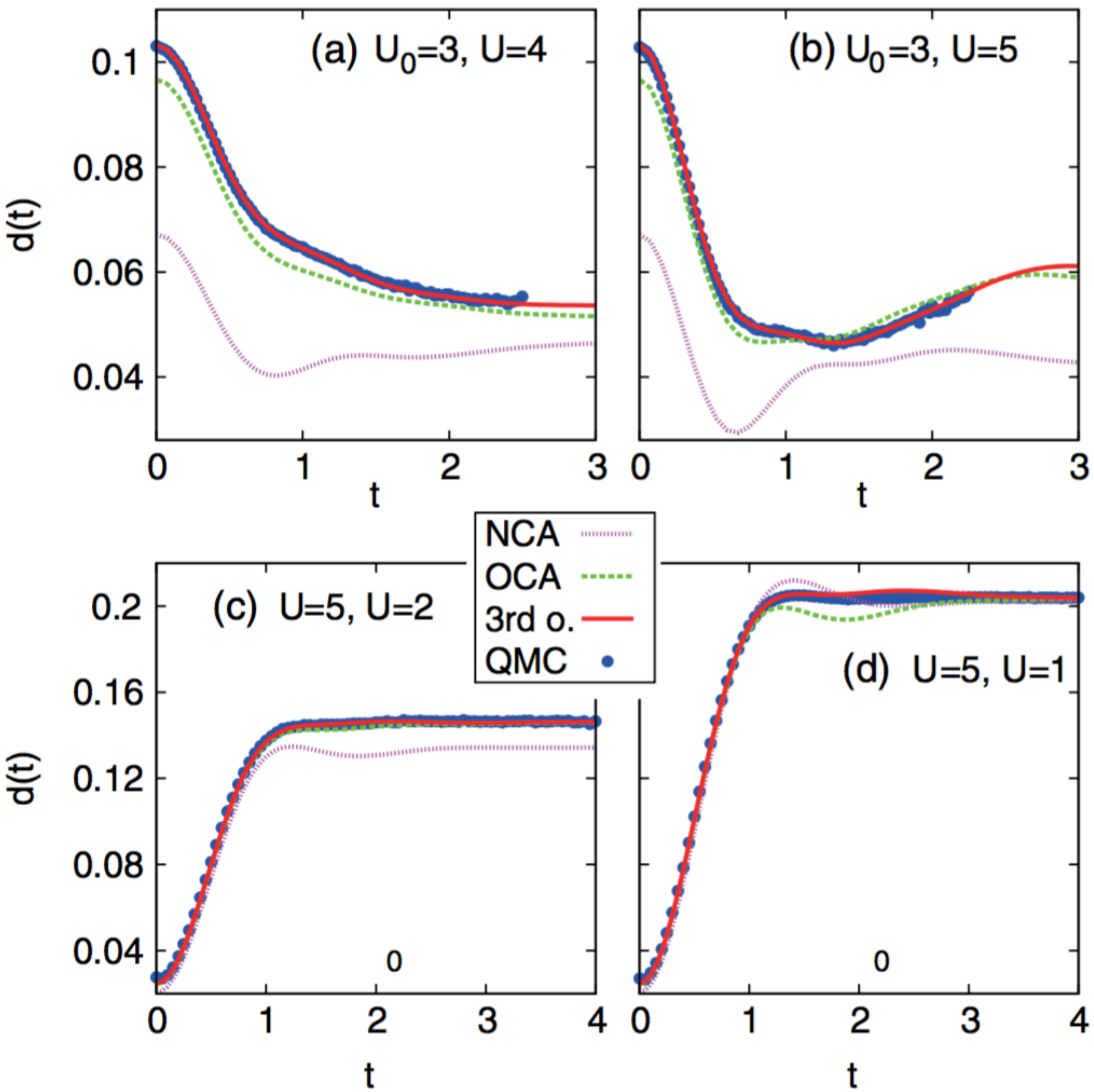
$\mathcal{O}_{nm} \equiv \langle n|\mathcal{O}|m\rangle$

etc.

Strong-coupling expansion

Example: Hubbard model, quench U_0 to U

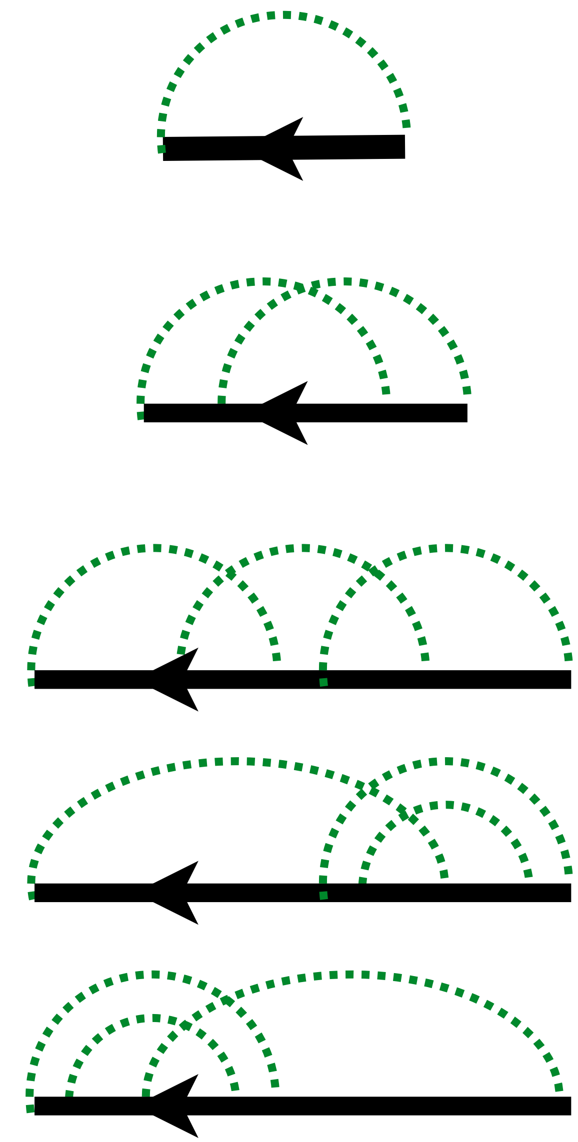
double occupancy $\langle n_{\uparrow} n_{i\downarrow} \rangle$:



NCA

OCA

3rd order

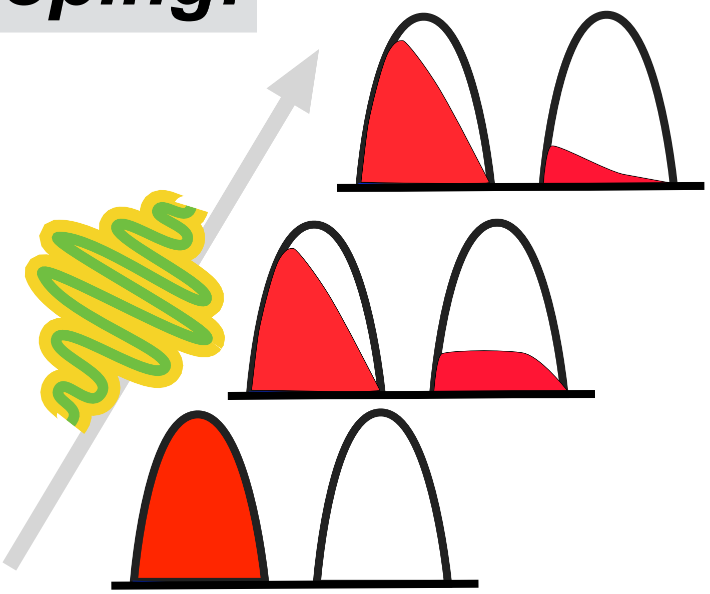


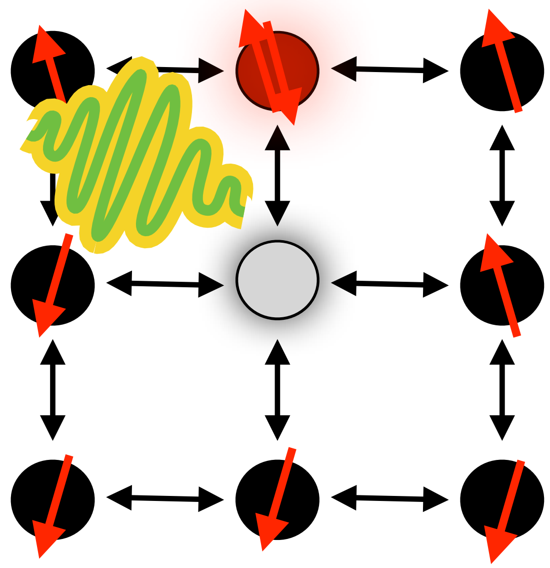
Eckstein & Werner, PRB 2010

Application: Photo-doping in Mott insulators

Photo-induced Mott transition

Photo-doping:





Mott insulators:

Spectrum depends on population

Closing of gap - screening?

Formation of electronic quasiparticles (good metal/bad metal)?

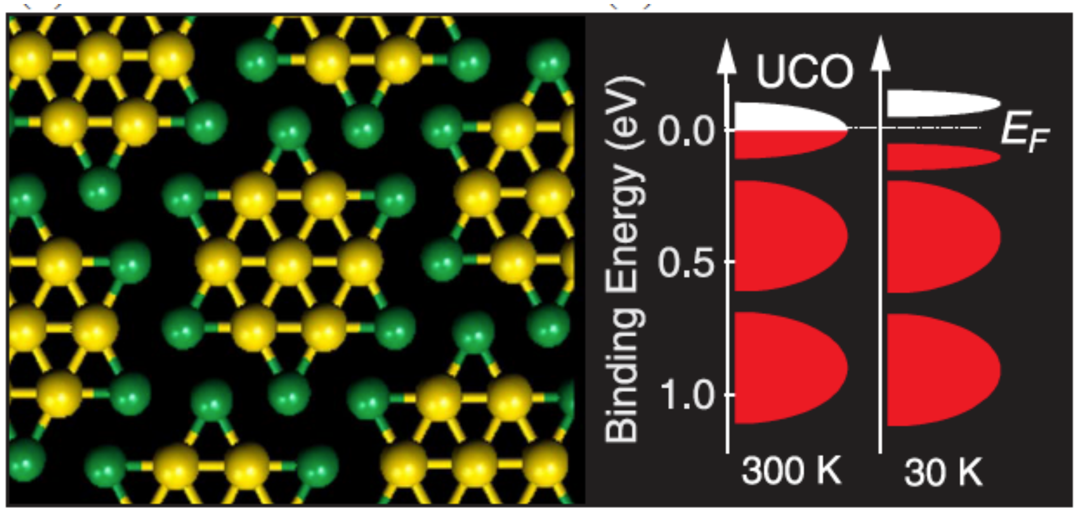
Active low energy degrees of freedom

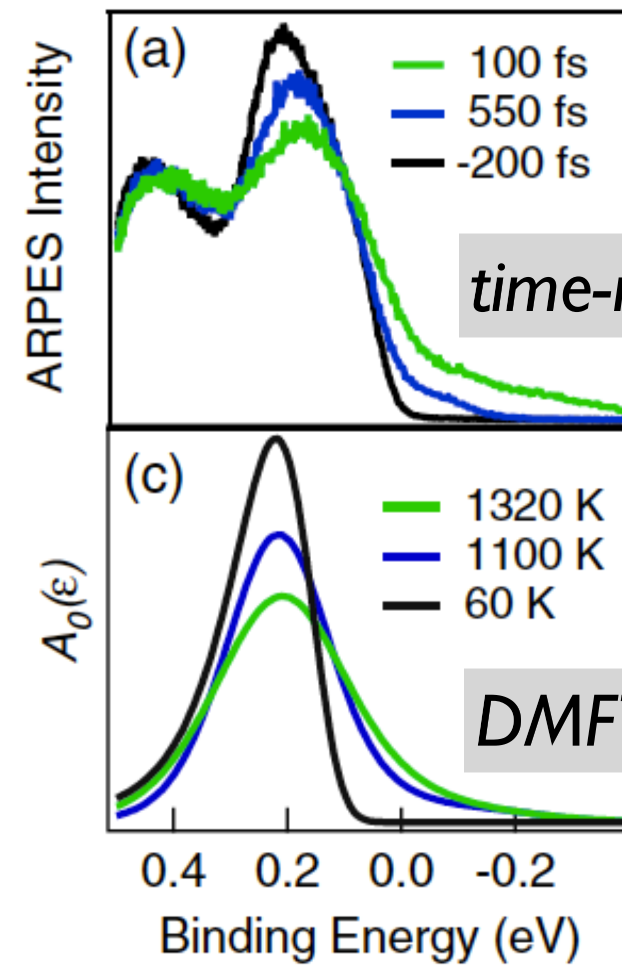
Unusually fast intra-band and inter-band dynamics

Modify spin/orbital/charge order by "photo-doping" ?

Photo-induced Mott transition in TaS_2

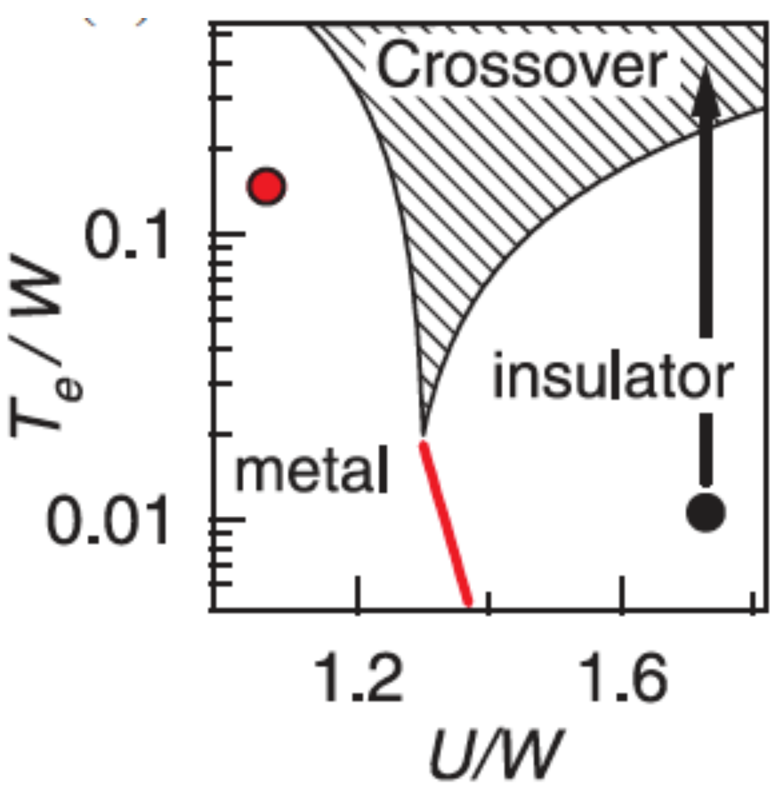
Perfetti et al., Phys. Rev. Lett. 97, 067402 (2007)





time-resolved ARPES

DMFT (high temperature)

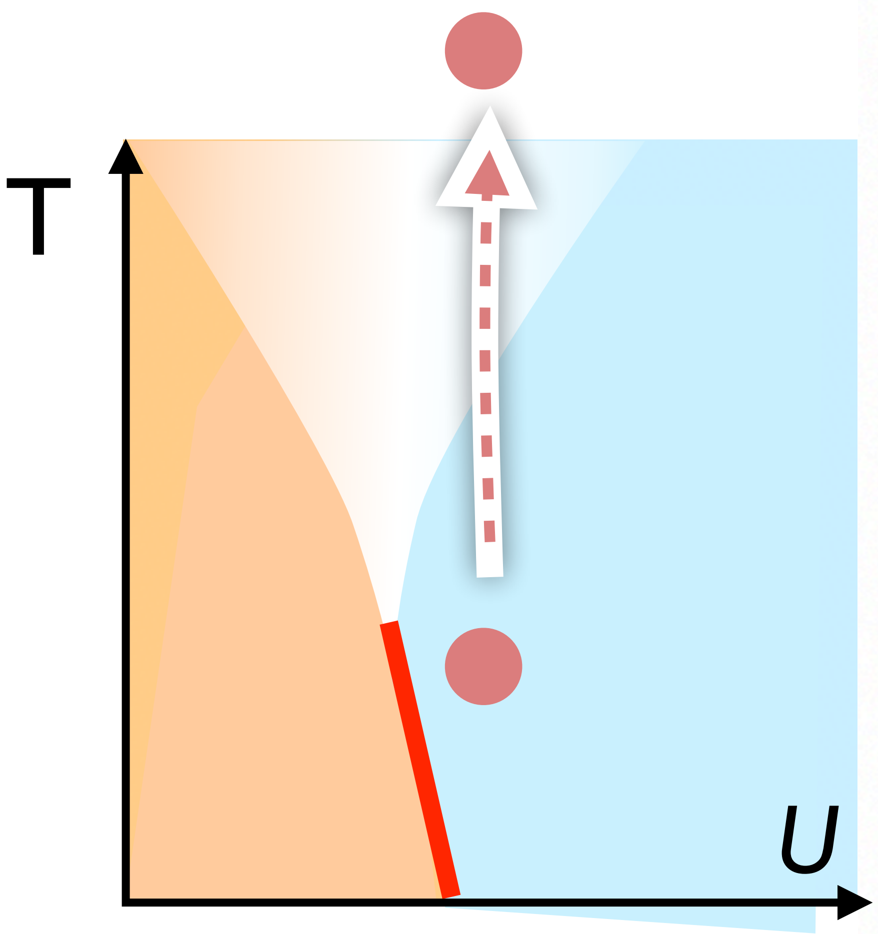


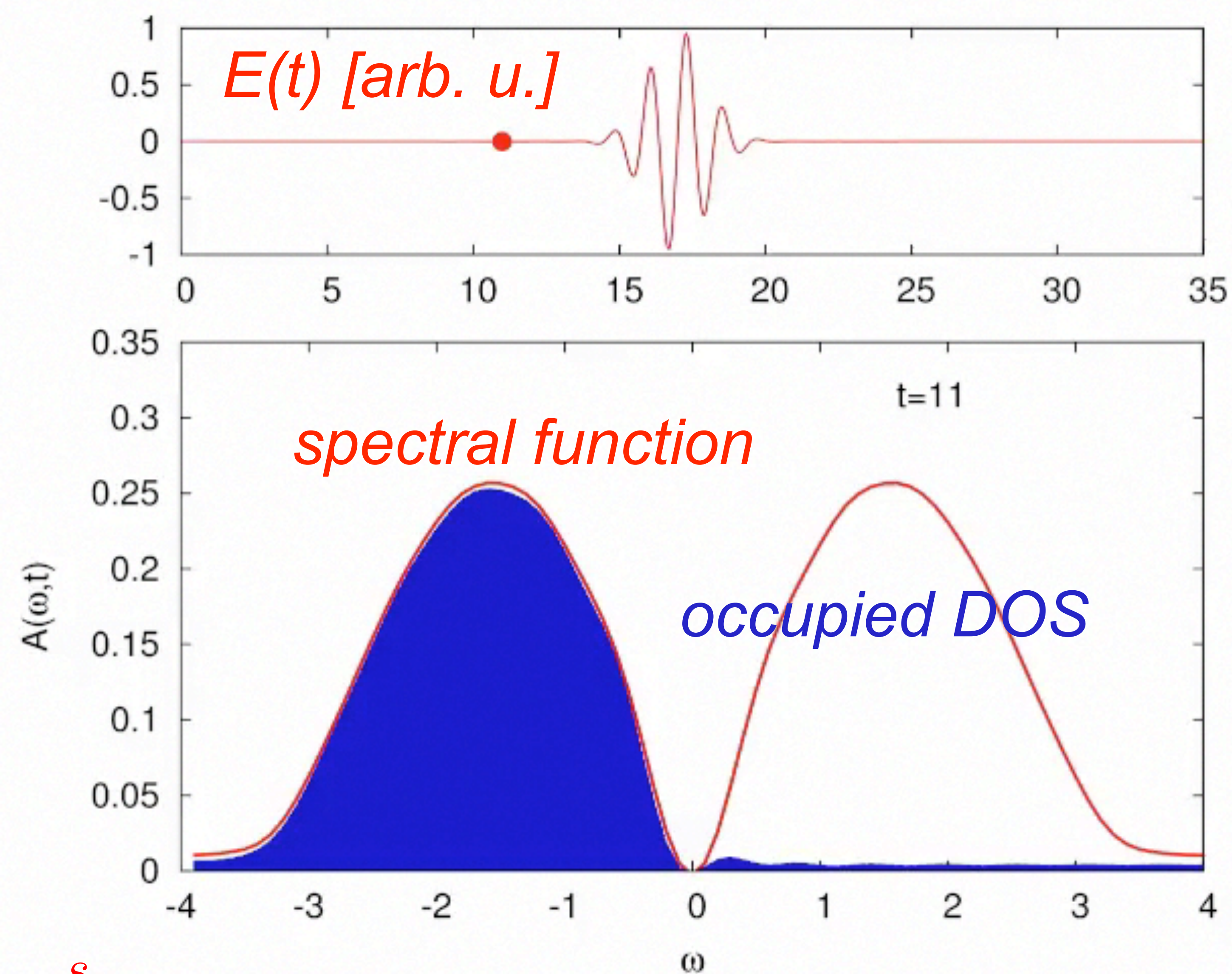
Crossover regime: Thermalization of electrons much faster than electron-lattice relaxation time
⇒ two temperature model?

Thermalization of the pump-excited Mott insulator

Thermalization of the pump-excited Mott insulator

(hypercubic lattice) ⇒ $U_c \approx 3$, closed system *(no bath)*





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

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

Eckstein & Werner Phys. Rev. B (2012)

Thermalization of the pump-excited Mott insulator

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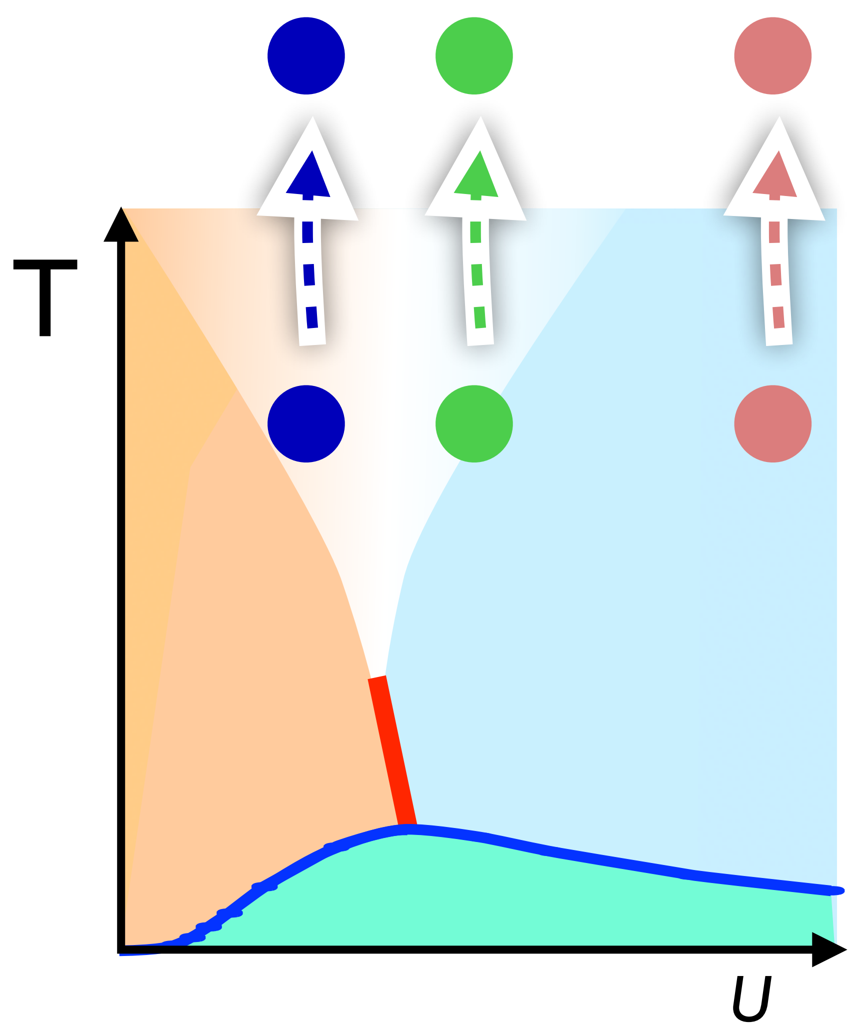
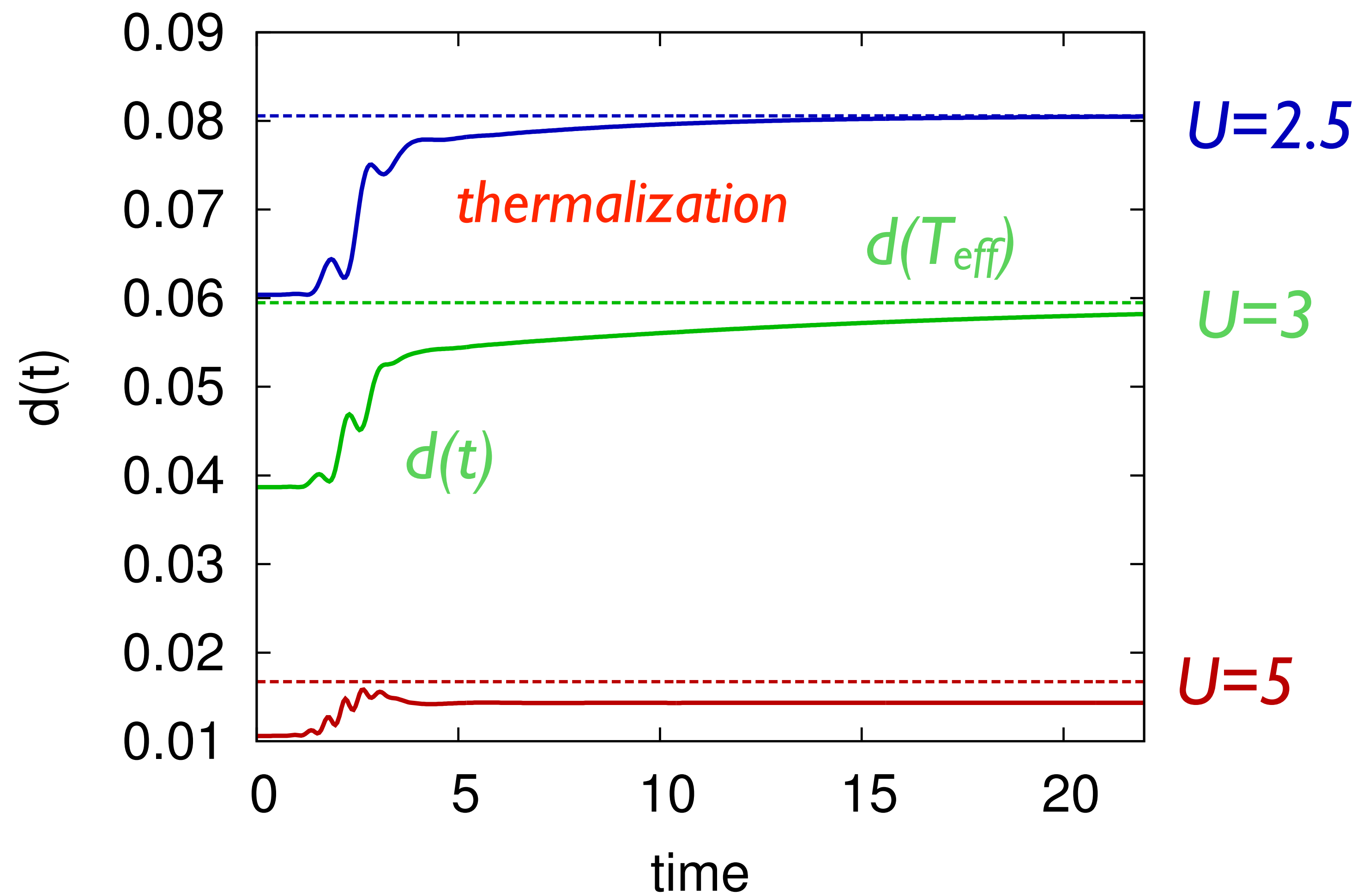


$$d(t) = \langle n_\uparrow(t) n_\downarrow(t) \rangle$$



Exitation:

initial temperature T=0.2 →

after excitation: energy ⇔ T_{eff} = 0.5

Eckstein & Werner Phys. Rev. B (2012)

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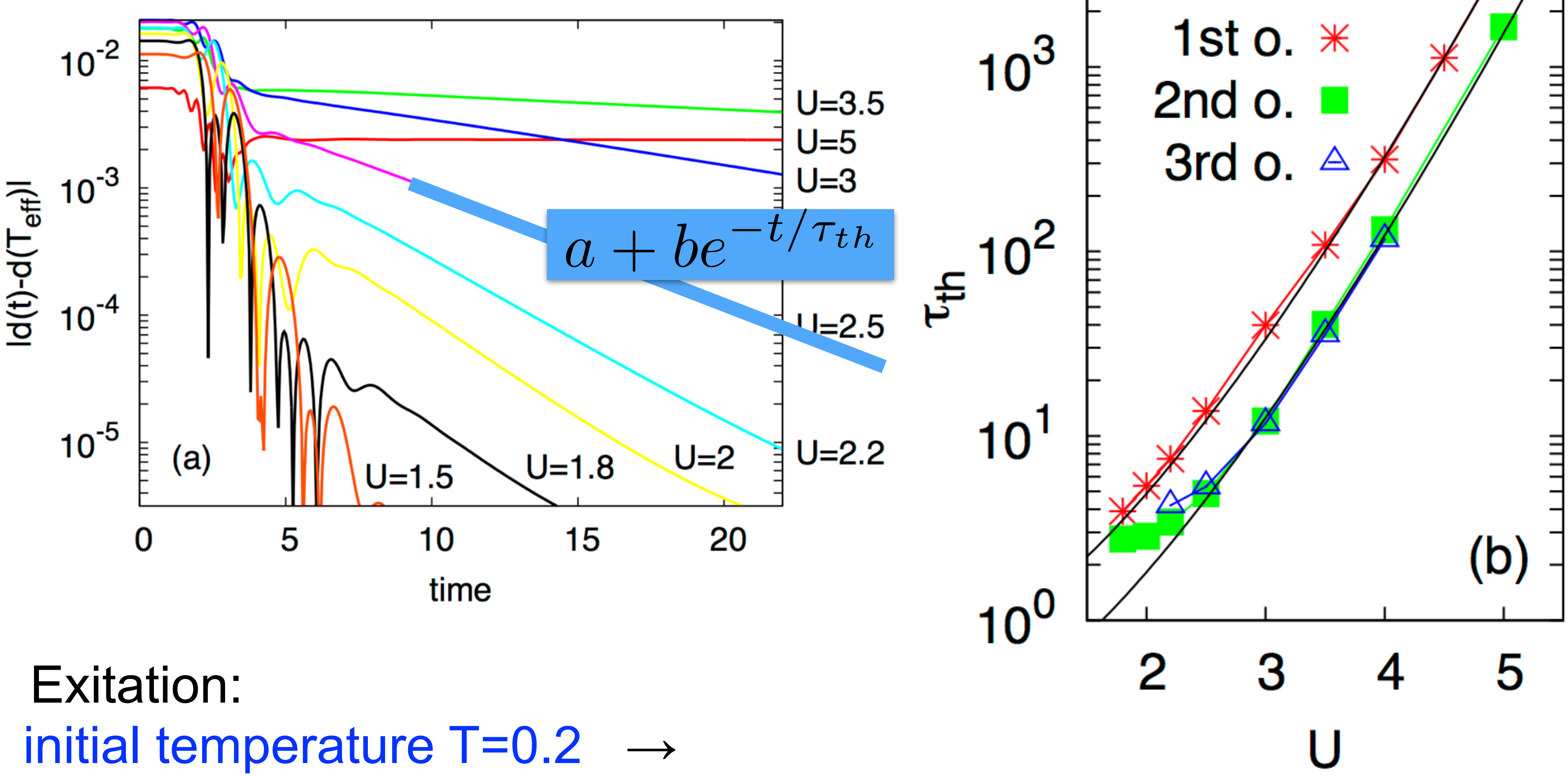

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Eckstein & Werner Phys. Rev. B (2012)

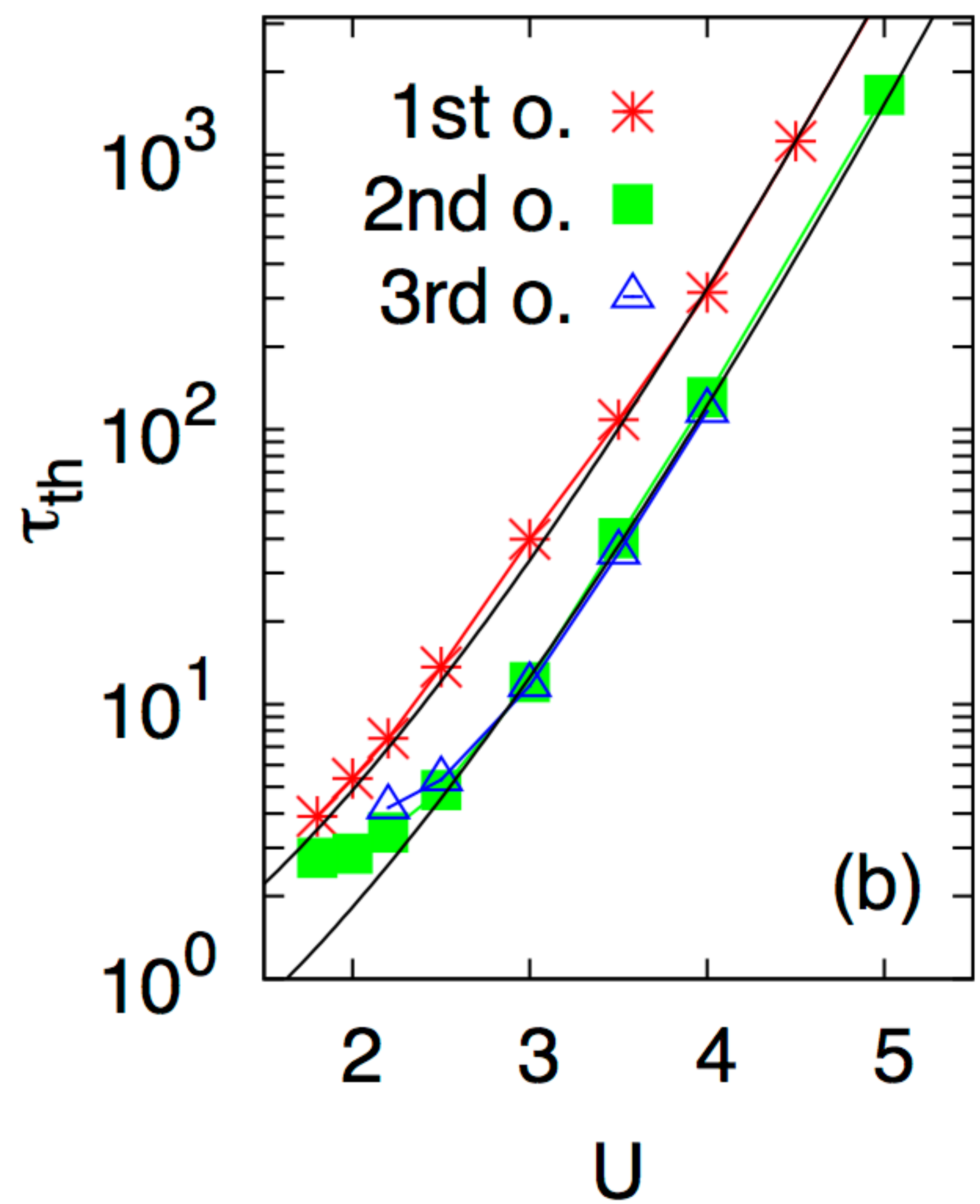
Thermalization of the pump-excited Mott insulator

(hypercubic lattice) ⇒ $U_c \approx 3$, closed system (no bath)

rapid thermalization in the small gap case (hopping time)

large U:

$$\tau_{th} \sim \exp[\alpha U/W \log(U/W)]$$



Eckstein & Werner Phys. Rev. B (2012)

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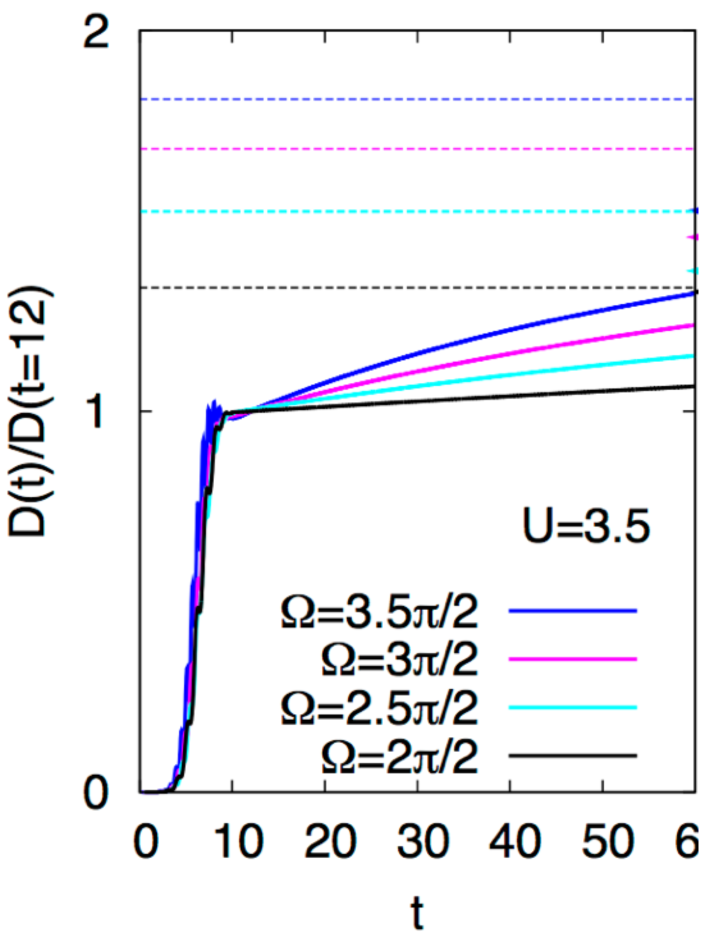
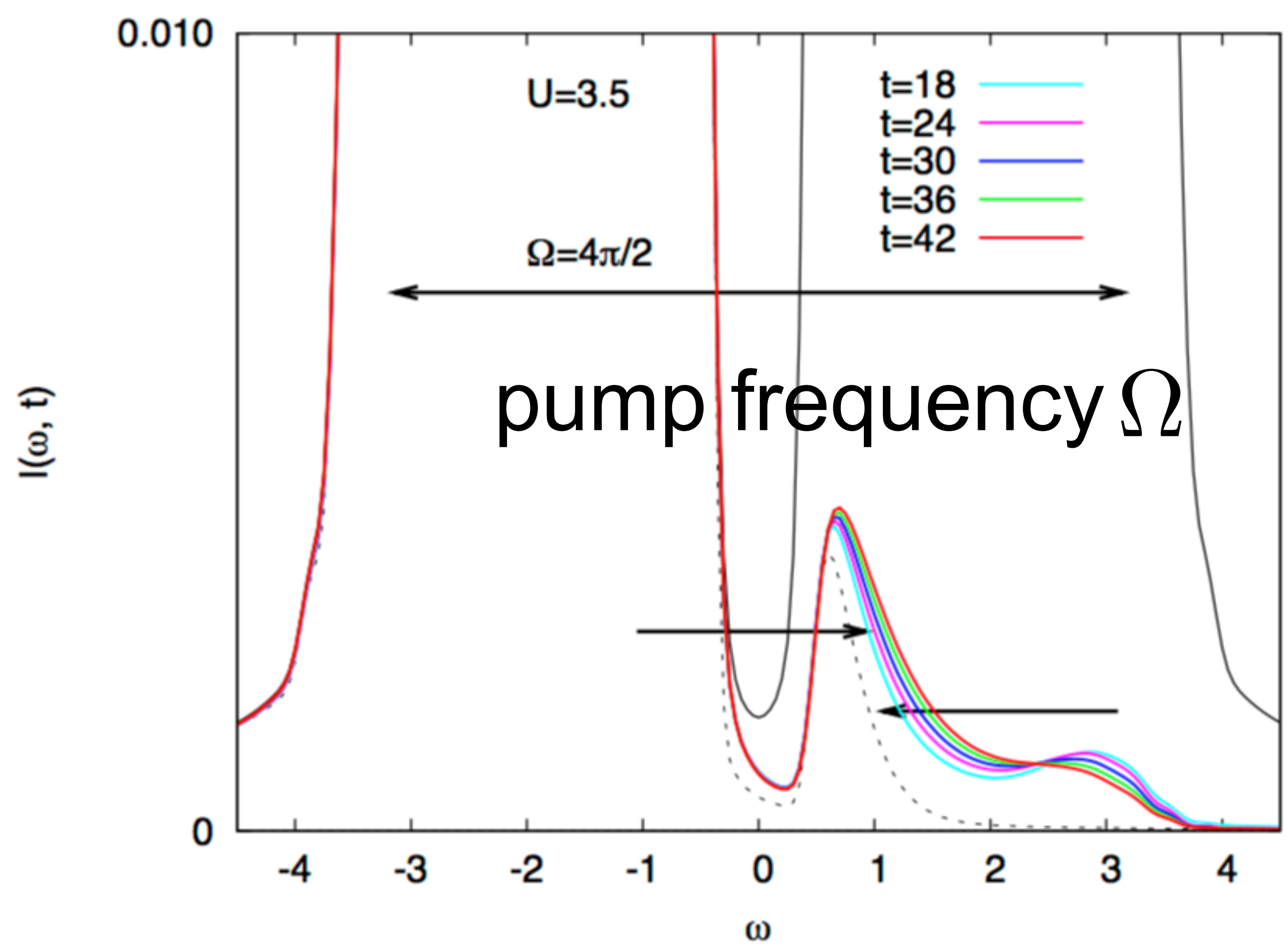




d(t) increases upon thermalization!

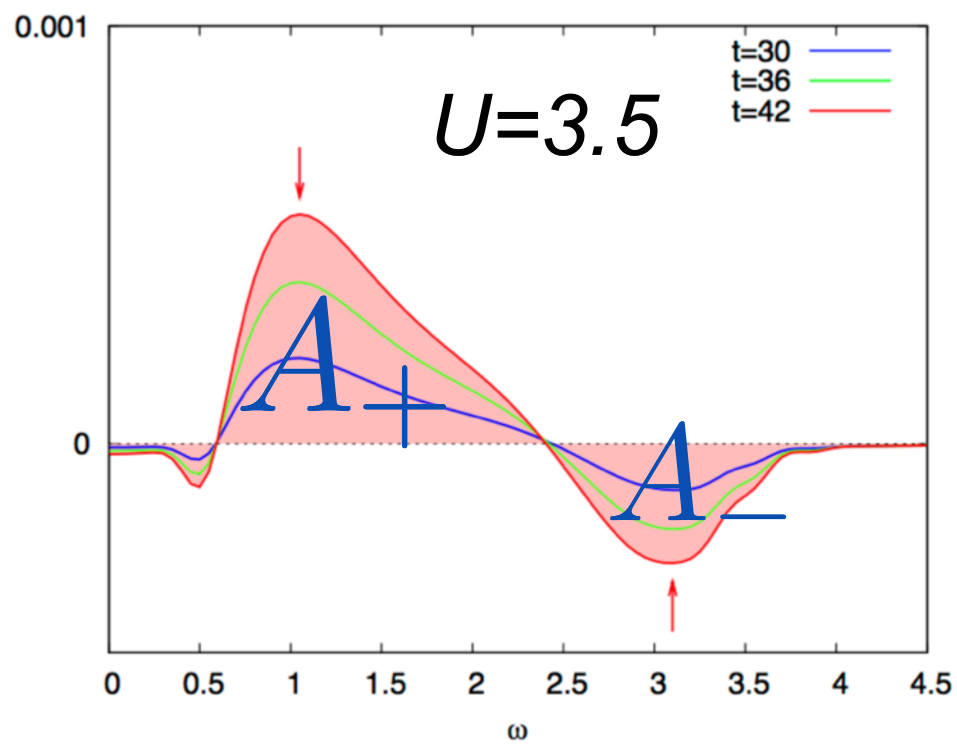
redistribution of occupied density of states

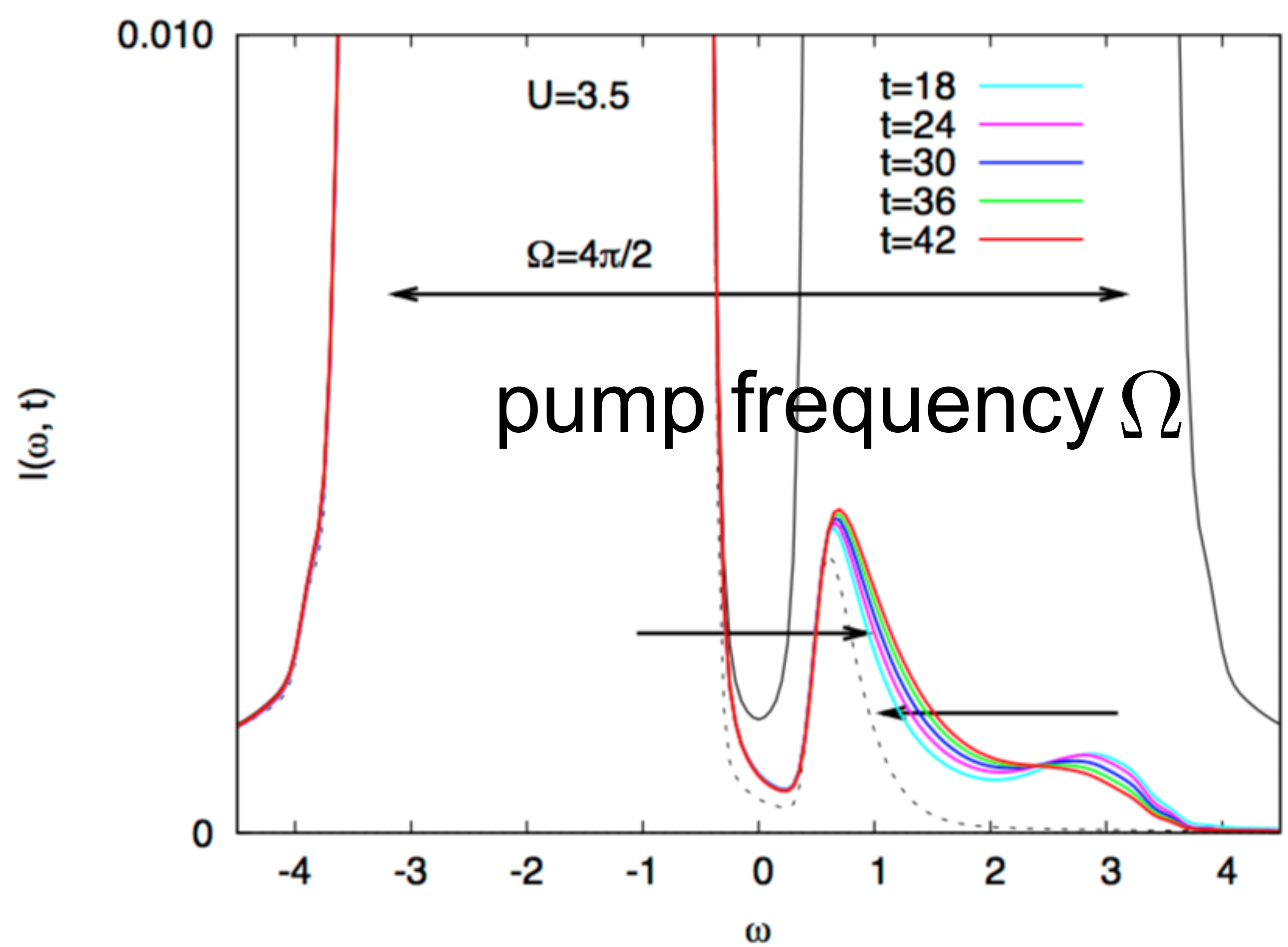
Werner, Held & Eckstein, (2014)

Thermalization of the pump-excited Mott insulator

(hypercubic lattice) ⇒ $U_c \approx 3$, *closed system* *(no bath)*

$A_+ = 2.3 \times A_-$





difference spectra

$I(\omega, t) - I(\omega, t = 24)$

"Carrier multiplication"

Perfect impact ionization $A_+ = 3 \times A_-$ expected

Werner, Held & Eckstein, (2014)

Thermalization of the pump-excited Mott insulator

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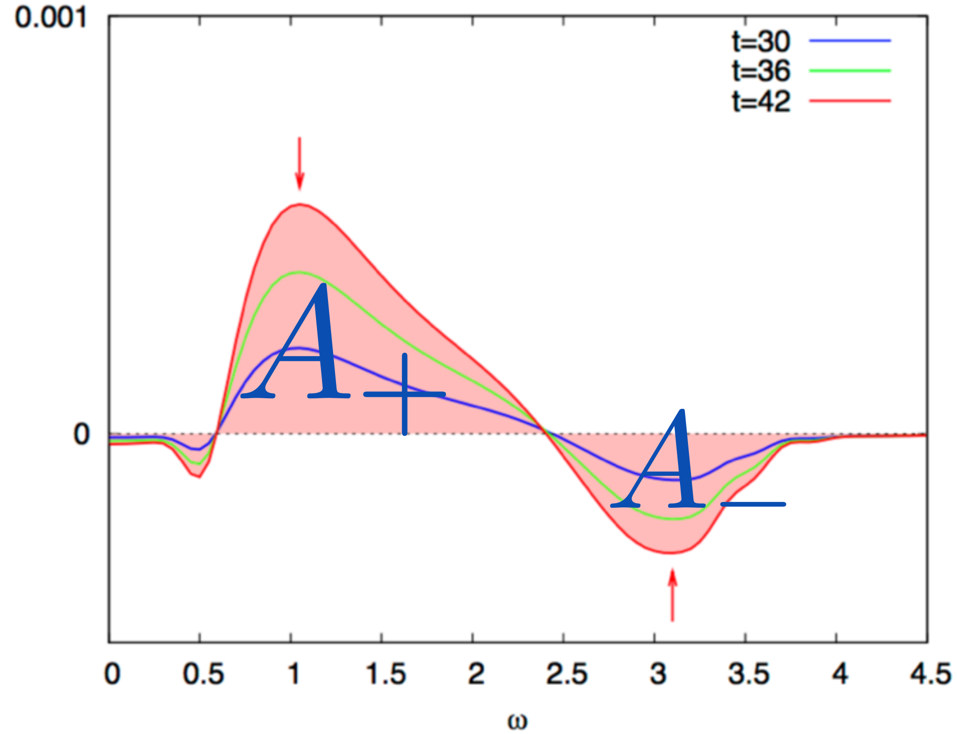
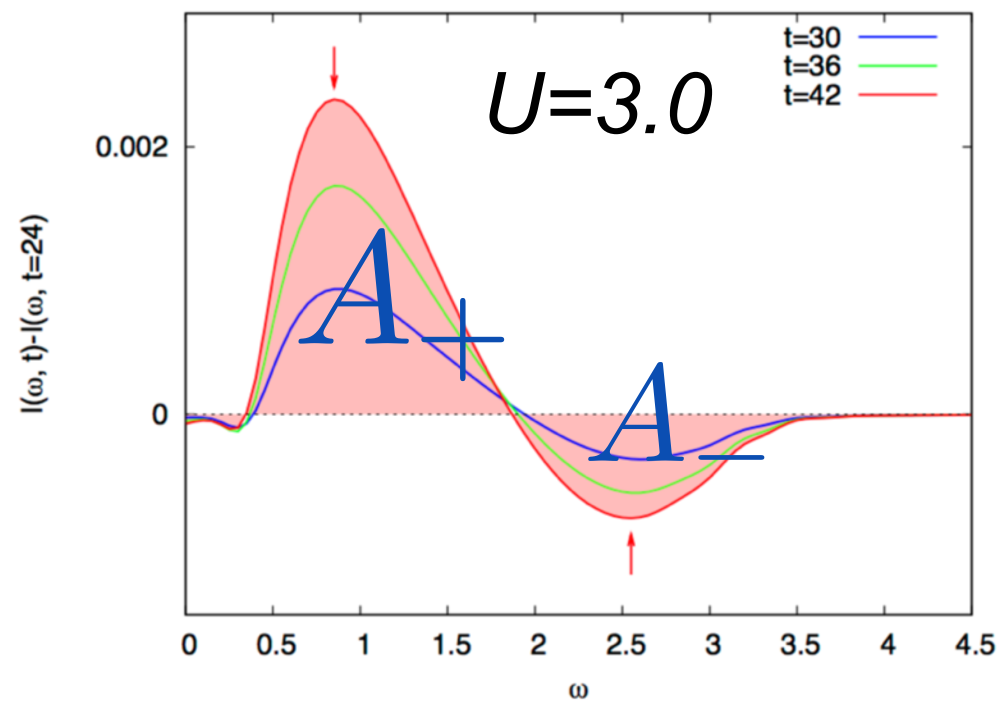


$$A_+ = 2.7 \times A_-$$



difference spectra

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Thermalization of the pump-excited Mott insulator

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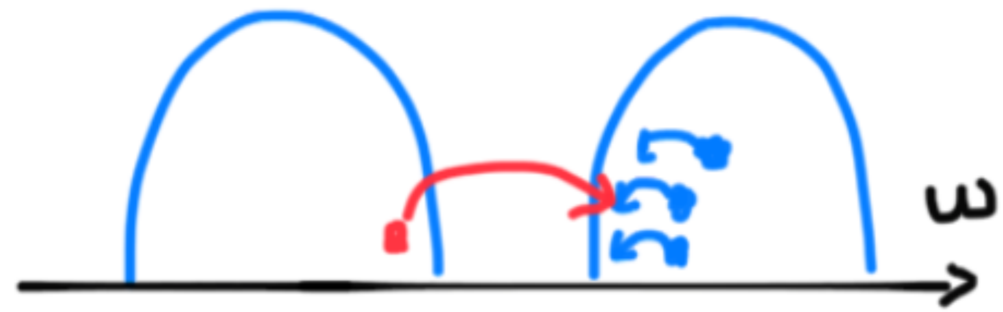
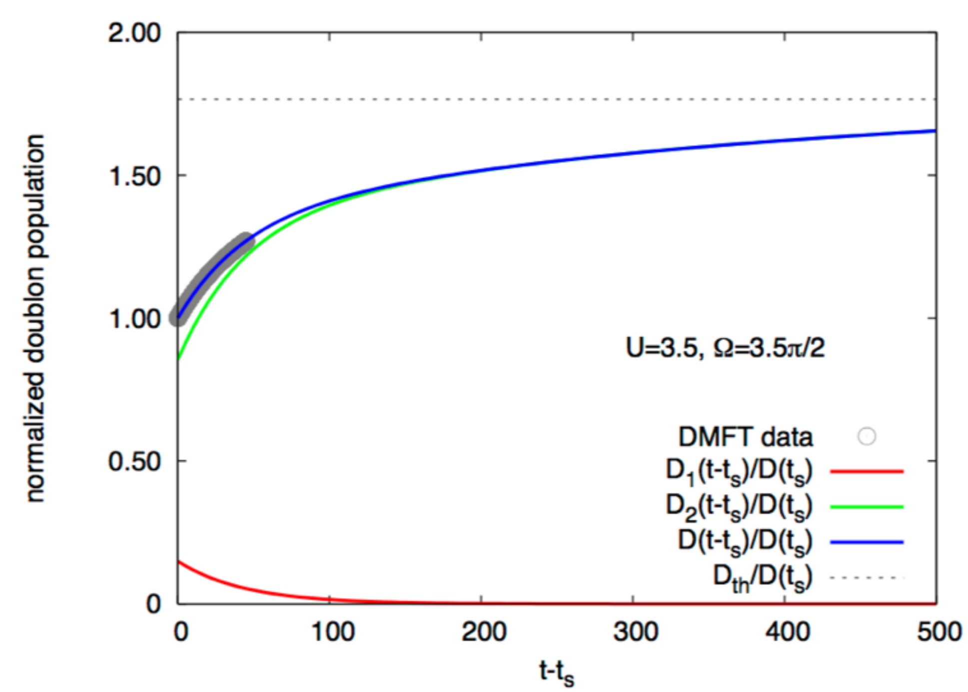
Difference: "high-order processes"



⇒ two-step thermalization?

D_1, D_2 : high/low energy occupation

$$\left(\frac{dD_1}{dt}\right)_{\text{imp}} = -\frac{1}{\gamma} D_1,$$
$$\left(\frac{dD_2}{dt}\right)_{\text{imp}} = -3\left(\frac{dD_1}{dt}\right)_{\text{imp}},$$
$$\left(\frac{d}{dt} D_2\right)_{\text{therm}} = \frac{1}{\tau}\left(D_{\text{th}} - D_2\right).$$



Werner, Held & Eckstein, (2014)

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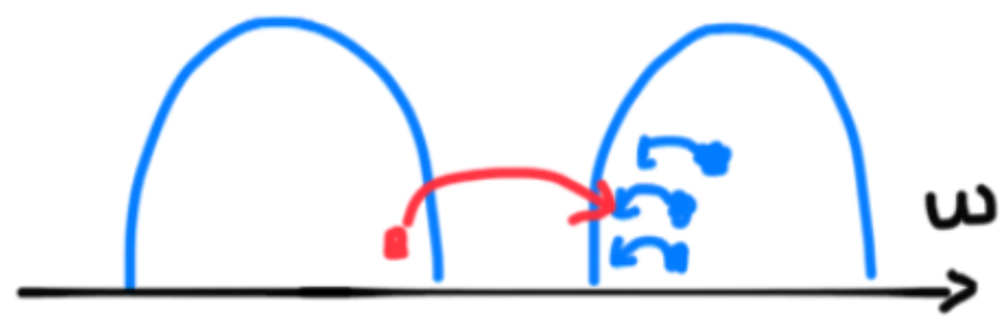

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$$\left(\frac{d}{dt}D_2\right)_{\text{therm}} = \frac{1}{\tau}\left(D_{\text{th}} - D_2\right).$$

femtosecond relaxation

U	Ω	γ	τ
2.5	$\frac{3\pi}{2}$	7.20	18.8
2.5	$\frac{2.5\pi}{2}$	7.75	19.0
2.5	$\frac{2\pi}{2}$	9.35	19.6
3	$\frac{3.5\pi}{2}$	13.4	60.3
3	$\frac{3\pi}{2}$	15.0	61.4
3	$\frac{2.5\pi}{2}$	16.5	64.9
3.5	$\frac{3.5\pi}{2}$	44.0	376
3.5	$\frac{3\pi}{2}$	48.4	257

Werner, Held & Eckstein, (2014)

Thermalization of the pump-excited Mott insulator

H*igh-order Fermi-Golden rule:*

Strohmaier et al. PRL (2010)

$$\frac{\tau_{\mathrm{D}}}{h/J} = C \exp\left(\alpha \frac{U}{6J}\right)$$

$$|N_d + 1, N_h + 1\rangle \longleftrightarrow |N_d, N_h, \ \text{excited}\rangle$$

$\Delta E = U = n6J$ Resonant coupling in nth order perturbation theory:

matrix element

$$\Gamma/J \propto M^2 \qquad M \sim \frac{J}{6J} \times \frac{J}{2{\times}6J} \times \cdots \times \frac{J}{n{\times}6J}$$

energy denominator

$$\ln(\Gamma/J) \sim -2\ln n! \sim -2n\ln n \ \sim const. \times (U/6J)\ln(U/6J)$$

Thermalization of the pump-excited Mott insulator

H*igh-order Fermi-Golden rule:*

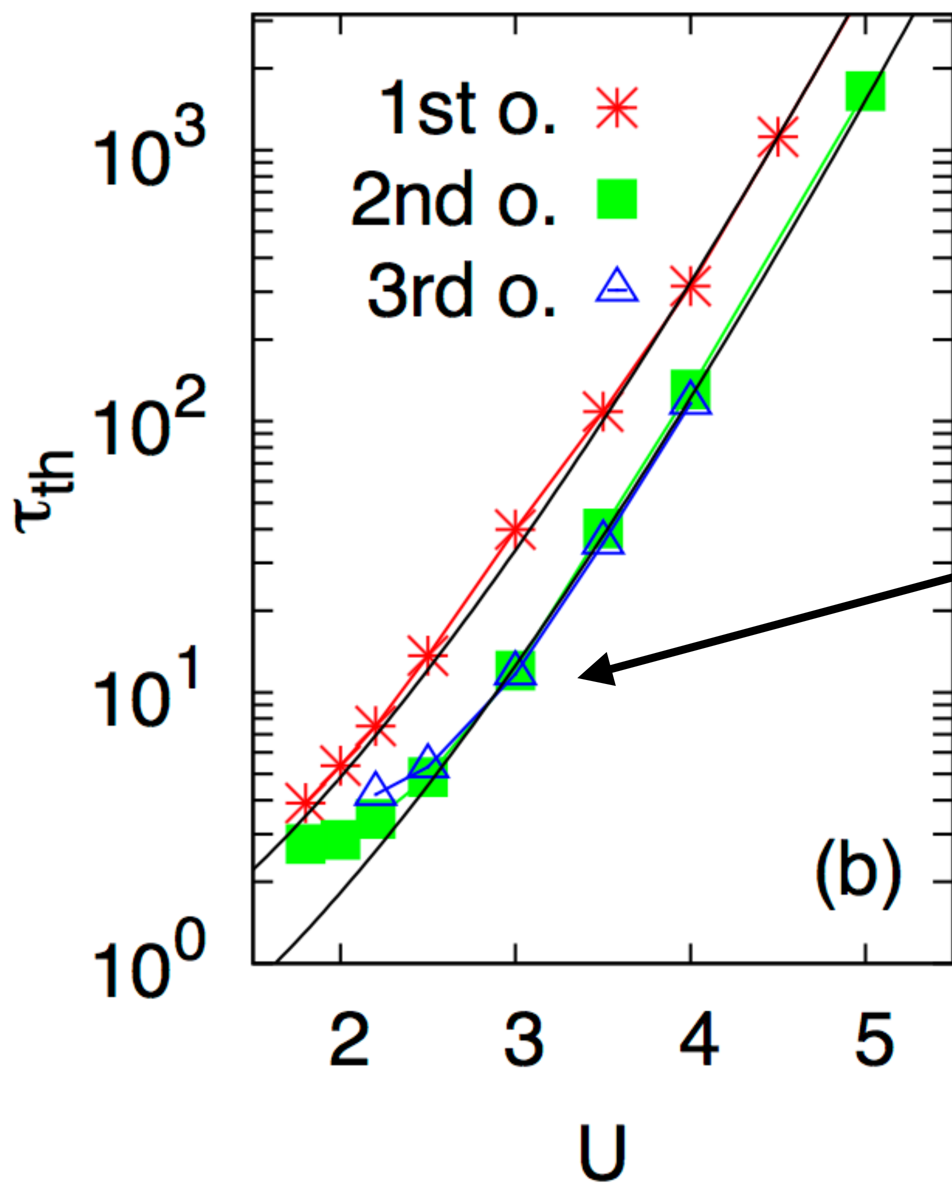


Photo-doped Mott - lines fit with:

$$\tau_{th} \sim \exp[\alpha U/W \log(U/W)]$$

Eckstein & Werner PRB (2012)

$$\ln(\Gamma/J) \sim -2\ln n! \sim -2n\ln n \sim const. \times (U/6J)\ln(U/6J)$$

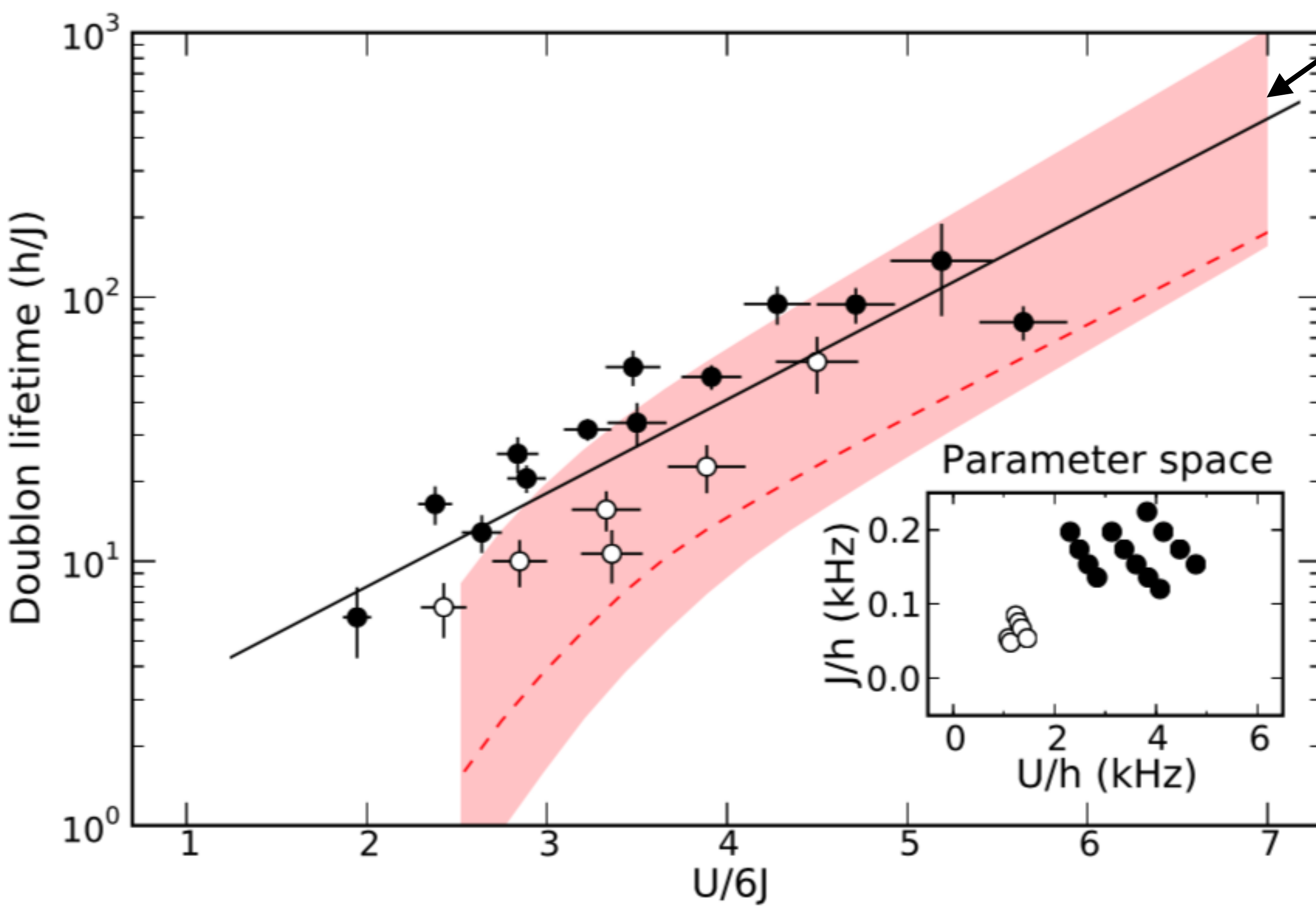
Strohmaier et al. PRL (2010)

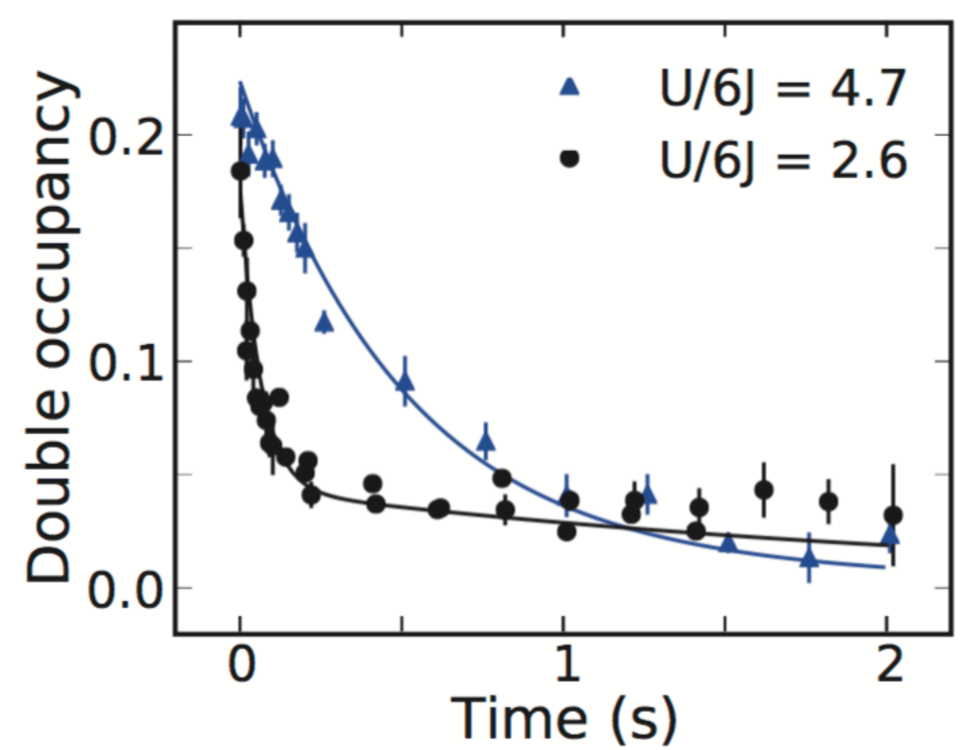
Thermalization of the pump-excited Mott insulator

large U: only high-order processes possible
⇒ slow thermalization rate

Decay of dynamically generated doublons in ultra-cold atoms (3d Hubbard model)

$$\frac{\tau_{\mathrm{D}}}{h/J} = C \exp\left(\alpha \frac{U}{6J}\right)$$





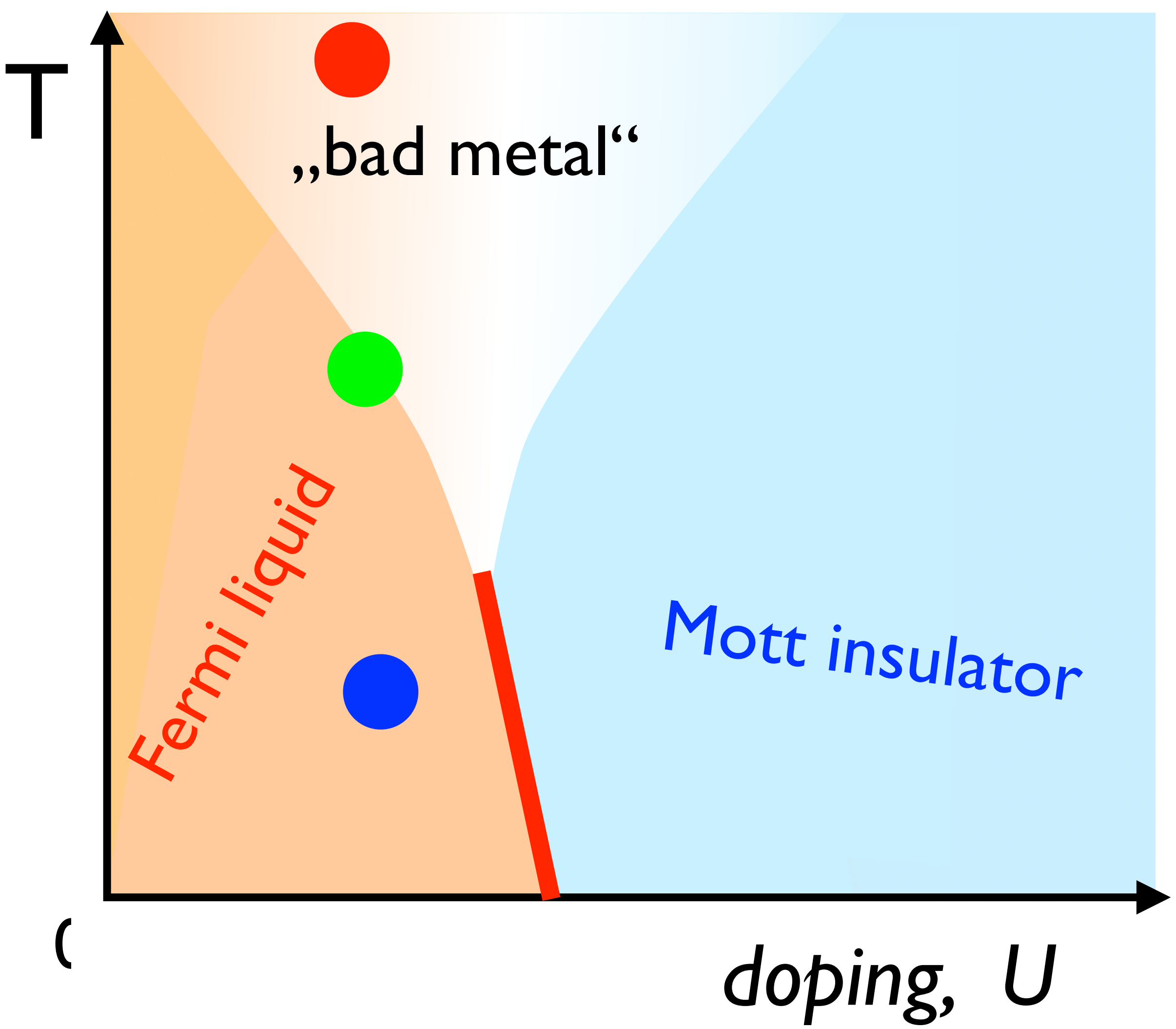
Strohmaier et al. PRL (2010)

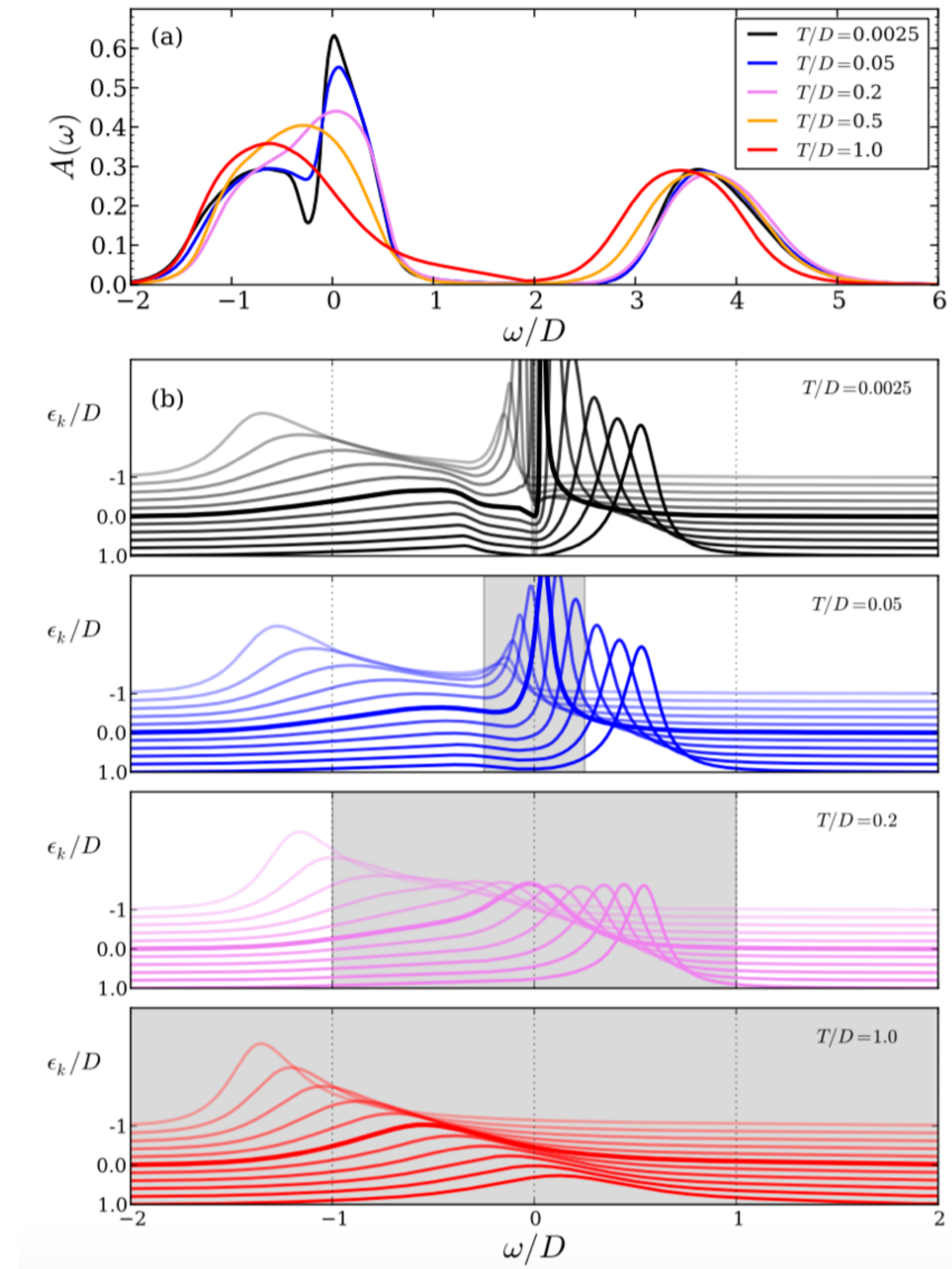
Quasi-particle formation at the Mott transition

Quasi-particle formation at the Mott transition

DMFT, Bethe lattice (bandwith=4)

schematic phase diagram,
single-site DMFT



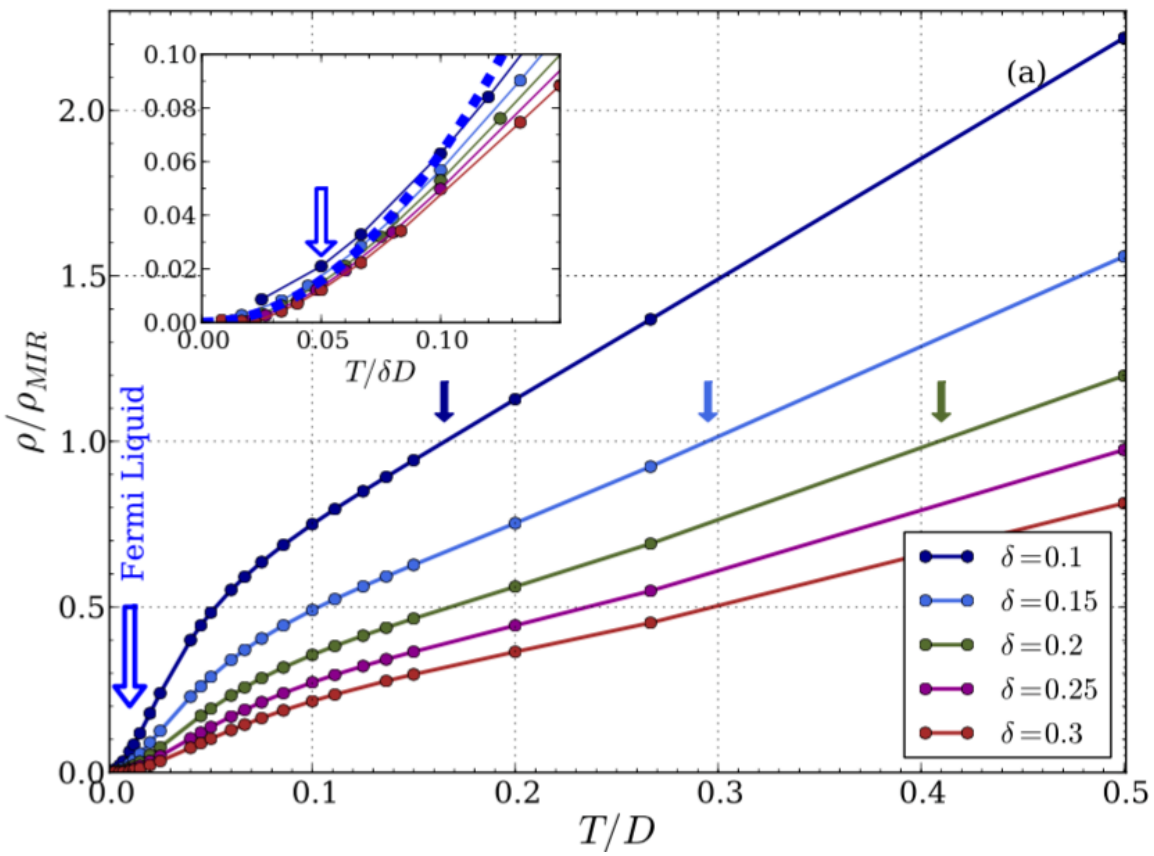


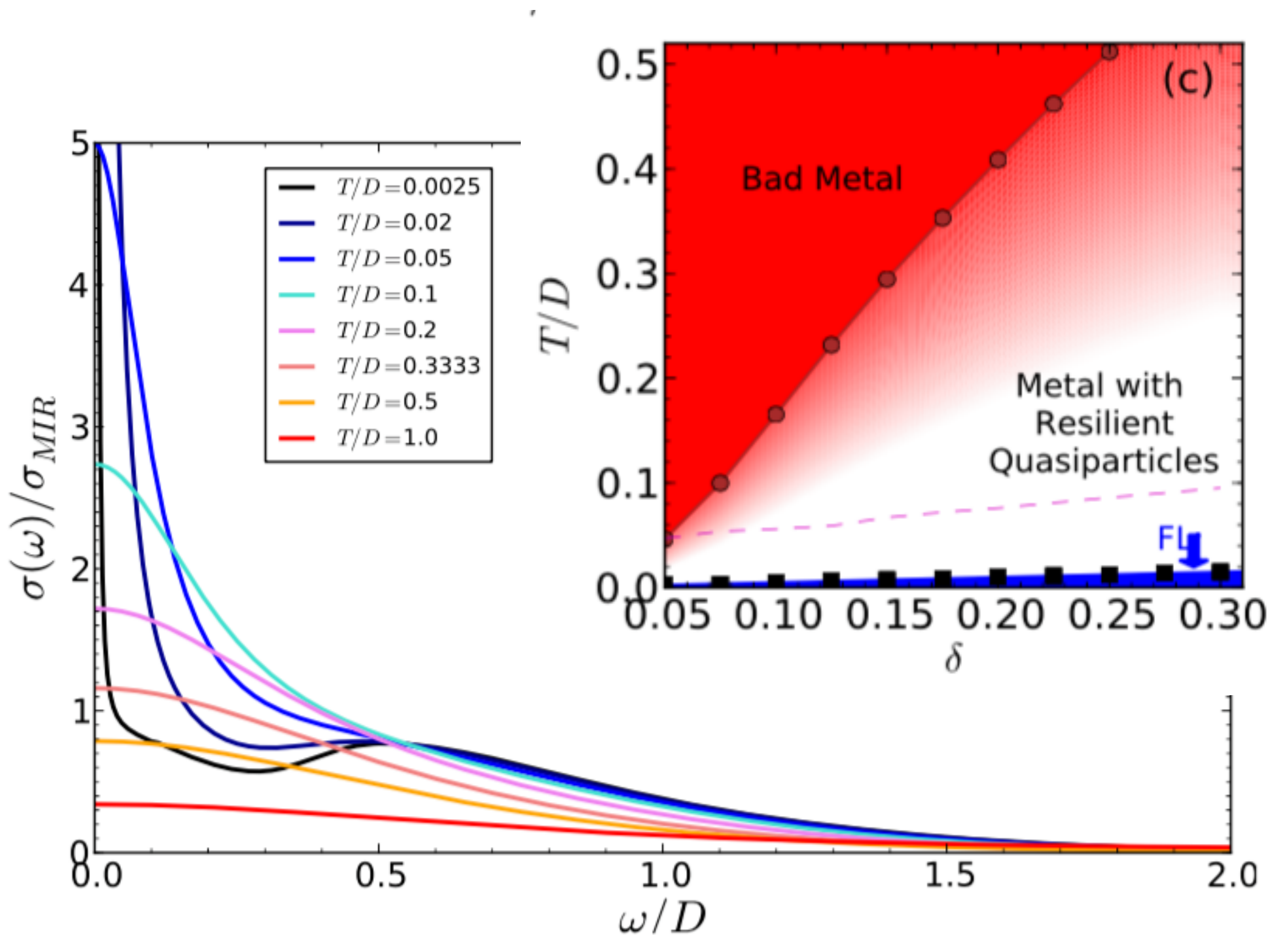
from Deng *et al.*, PRL 2013

Quasi-particle formation at the Mott transition

DMFT, Bethe lattice (bandwidth=4)

conductivity $\sigma(\omega)$

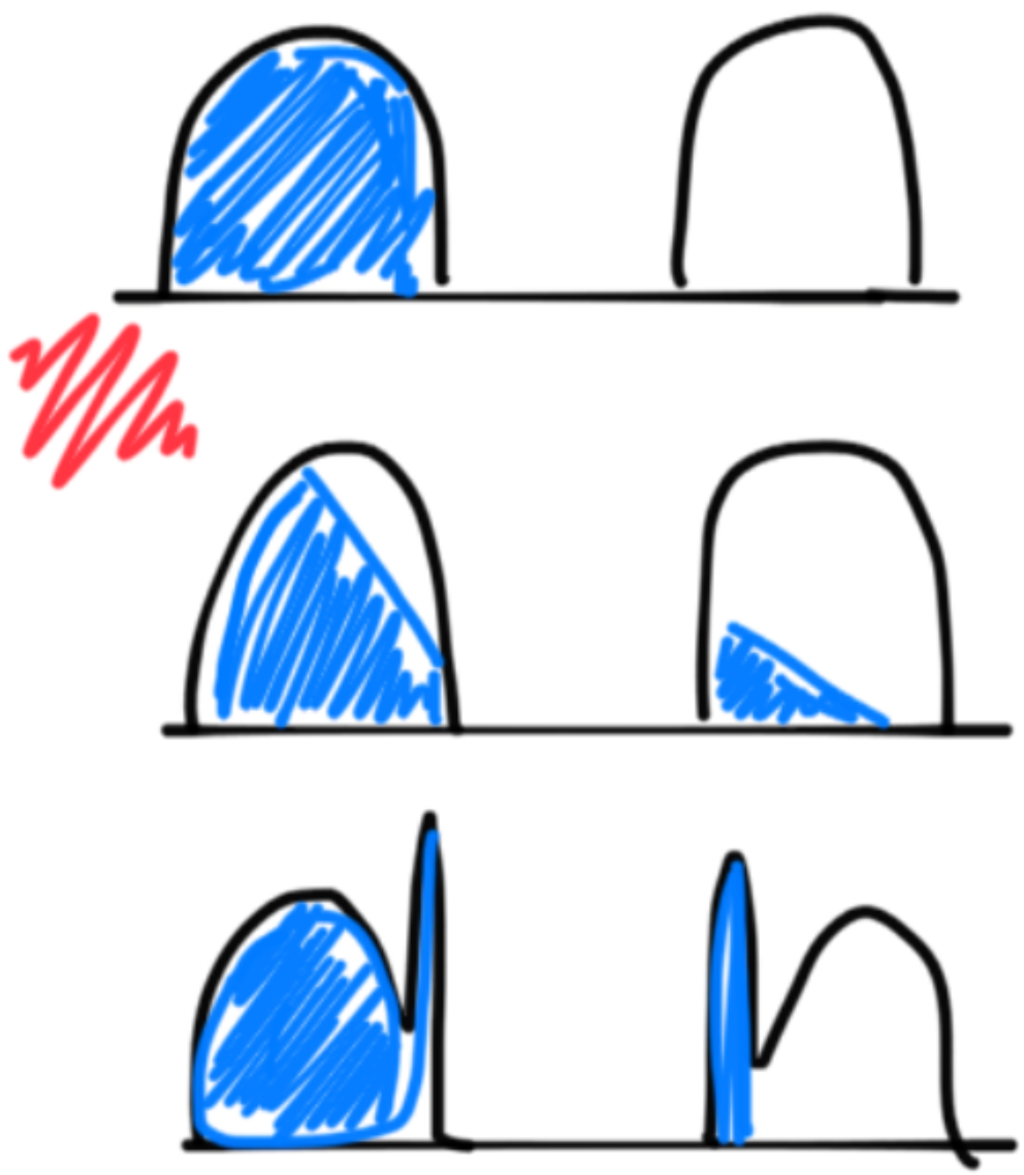




Mott-Ioffe-Regel limit for quasiparticle description of transport: $k_F \ell < 1$

from Deng *et al.*, PRL 2013

Quasi-particle formation at the Mott transition



$\approx >$ bath

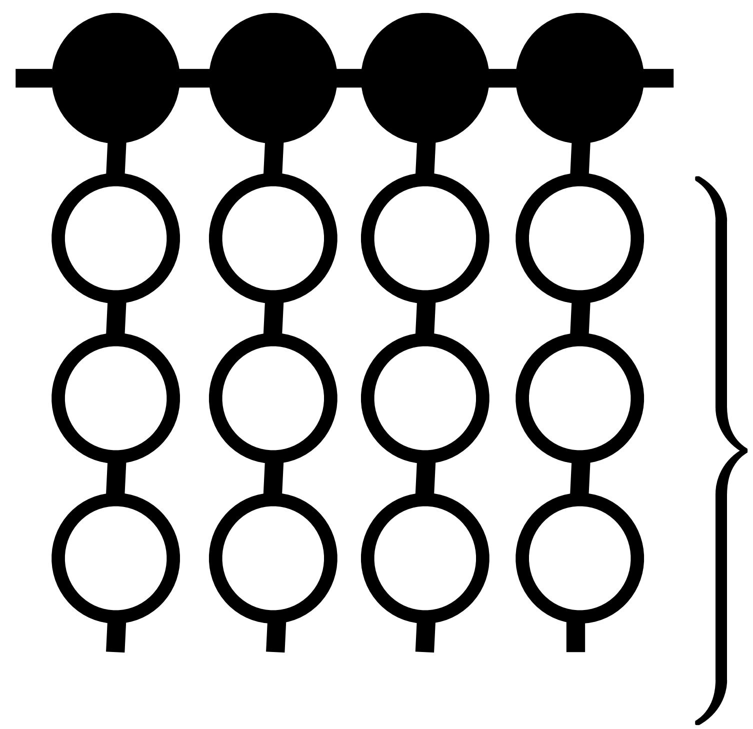
Quasi-particle formation at the Mott transition

Hubbard model plus bath in DMFT:

Fermion bath:

$$\Delta \rightarrow \Delta[G] + \Delta_{bath}$$

self-consistent part



additional sites

$\rightsquigarrow \ \Delta_{bath}$

Quasi-particle formation at the Mott transition

Hubbard model plus bath in DMFT:

<u>Bosonic bath:</u>

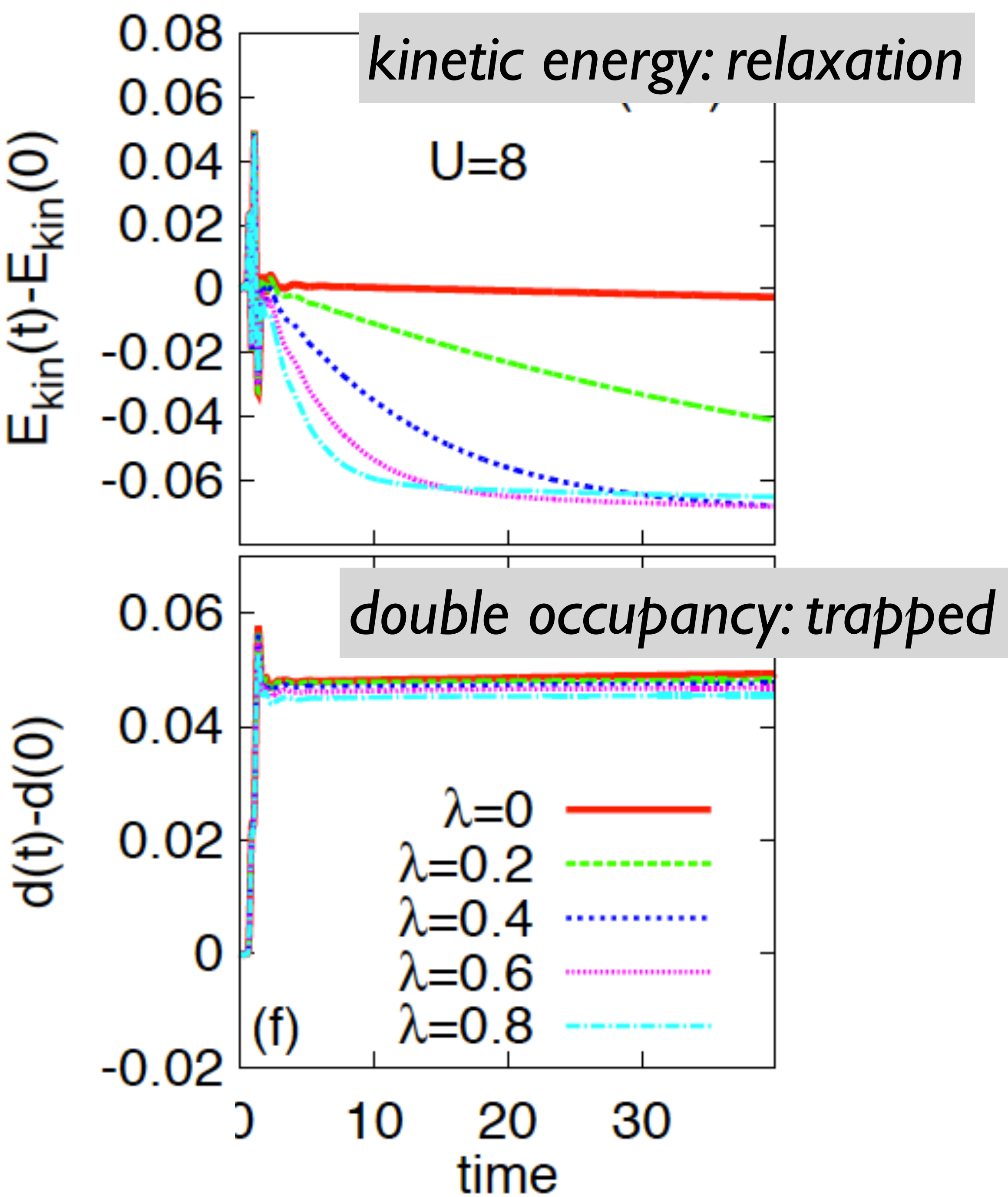
$$\Sigma = \Sigma_U[G] + \Sigma_{bath}$$

summed by auxiliary impurity model

$$\Sigma_{bath}(t,t') = \lambda G(t,t') \underbrace{D_{bath}(t,t')}_{\text{equilibrium propagator of bosons}}$$

Quasi-particle formation at the Mott transition

Hubbard model plus bath (1d density of states, W=4)



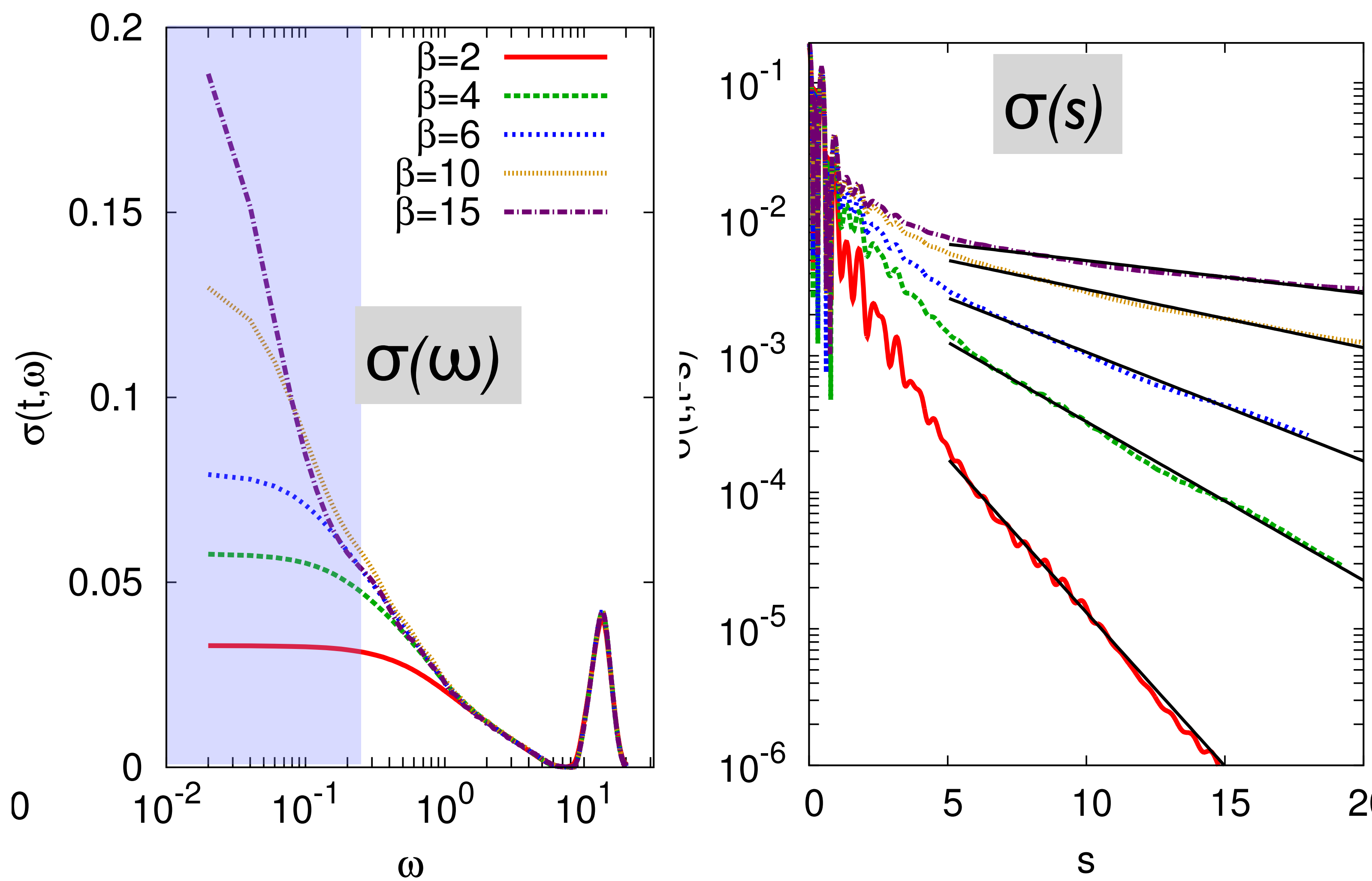
⇒ only "intra-band relaxation"

Eckstein and Werner, PRL 2013

Quasi-particle formation at the Mott transition

Conductivity: $\sigma(\omega)$

$$j(t) = \int_{-\infty}^{t} ds\, \sigma(t,s) E(s) \quad , \quad \sigma(\omega) = \int_{-\infty}^{t} ds\, \sigma(t-s) e^{i\omega s}$$



Equilibrium, doped Mott insulator U=8

Eckstein and Werner, PRL 2013

Quasi-particle formation at the Mott transition

How fast can a Drude peak of with 0 form in time?

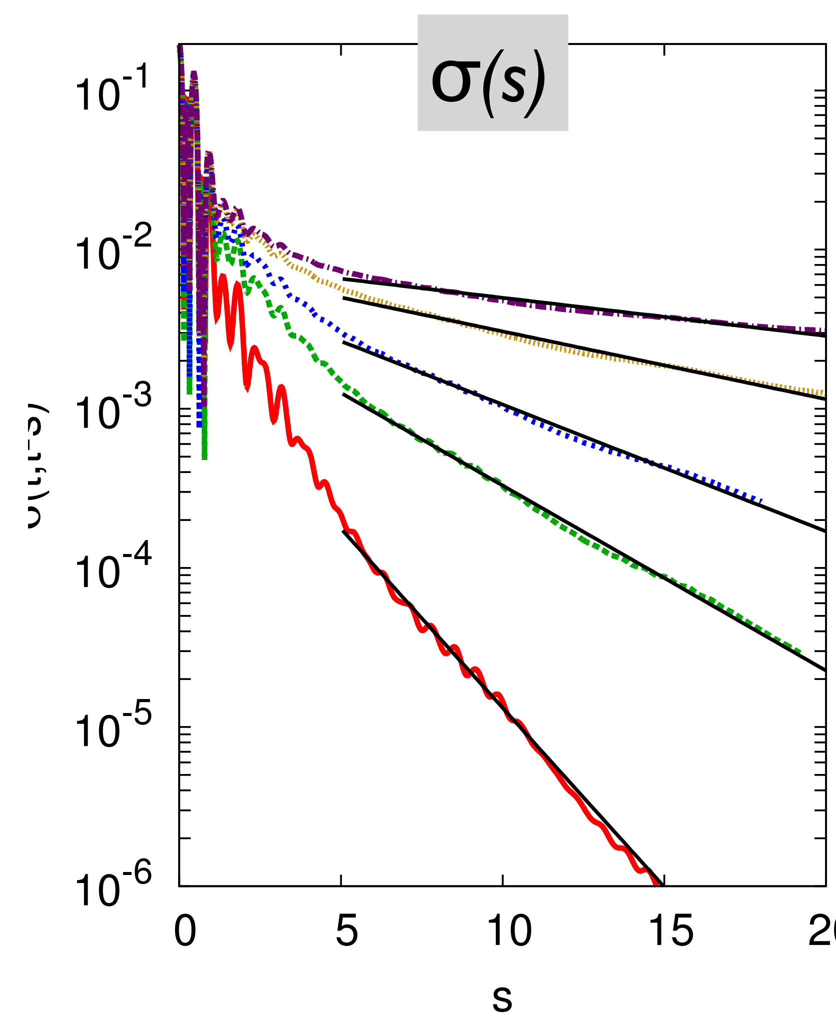
How fast can the scattering rate be changed in time?

no limit, e.g.,

$$\sigma(t, t-s) = const. \text{ for } t-s > 0$$

would imply nor scattering for t>0

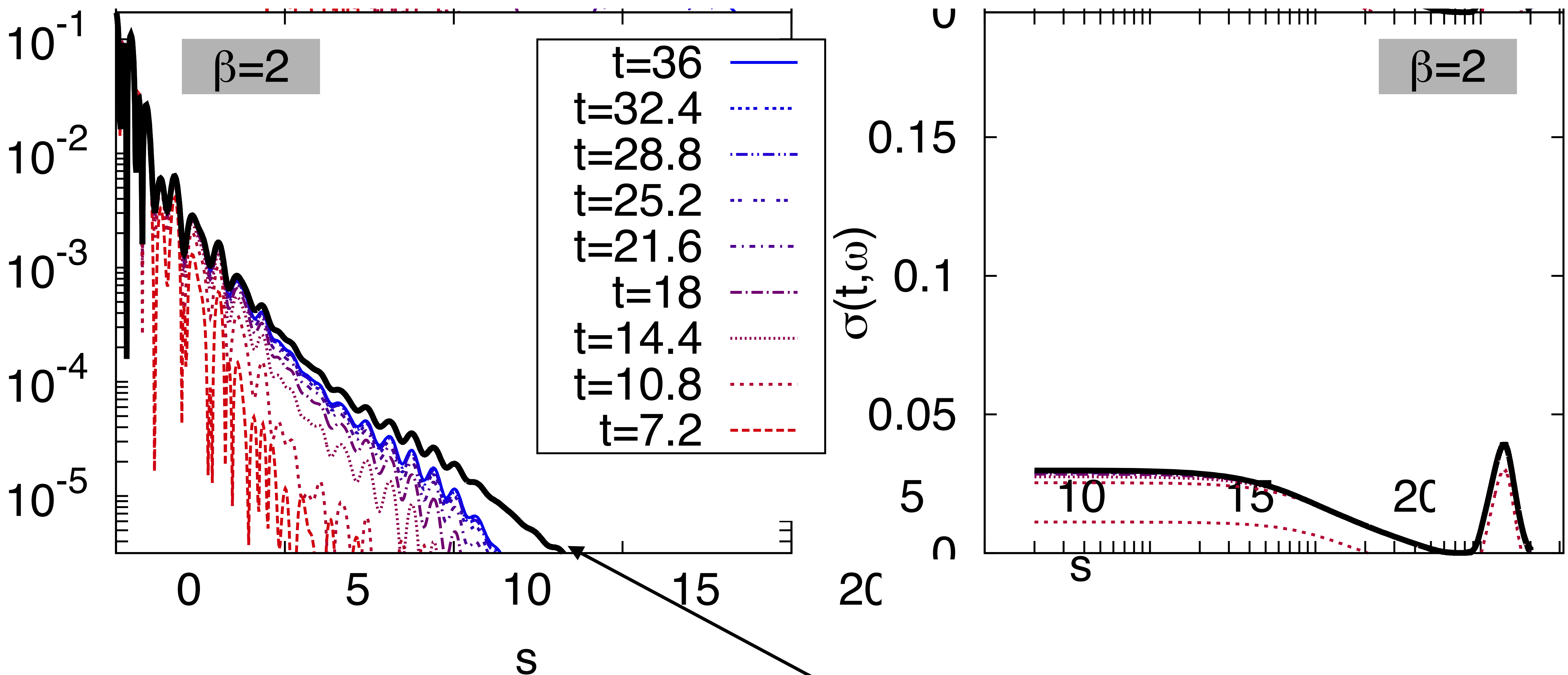
$\Rightarrow$ look at $\sigma(t, t-s)$ to analyze real-time data.



Eckstein and Werner, PRL 2013

Quasi-particle formation at the Mott transition

Hubbard model plus bath (1d density of states, W=4)



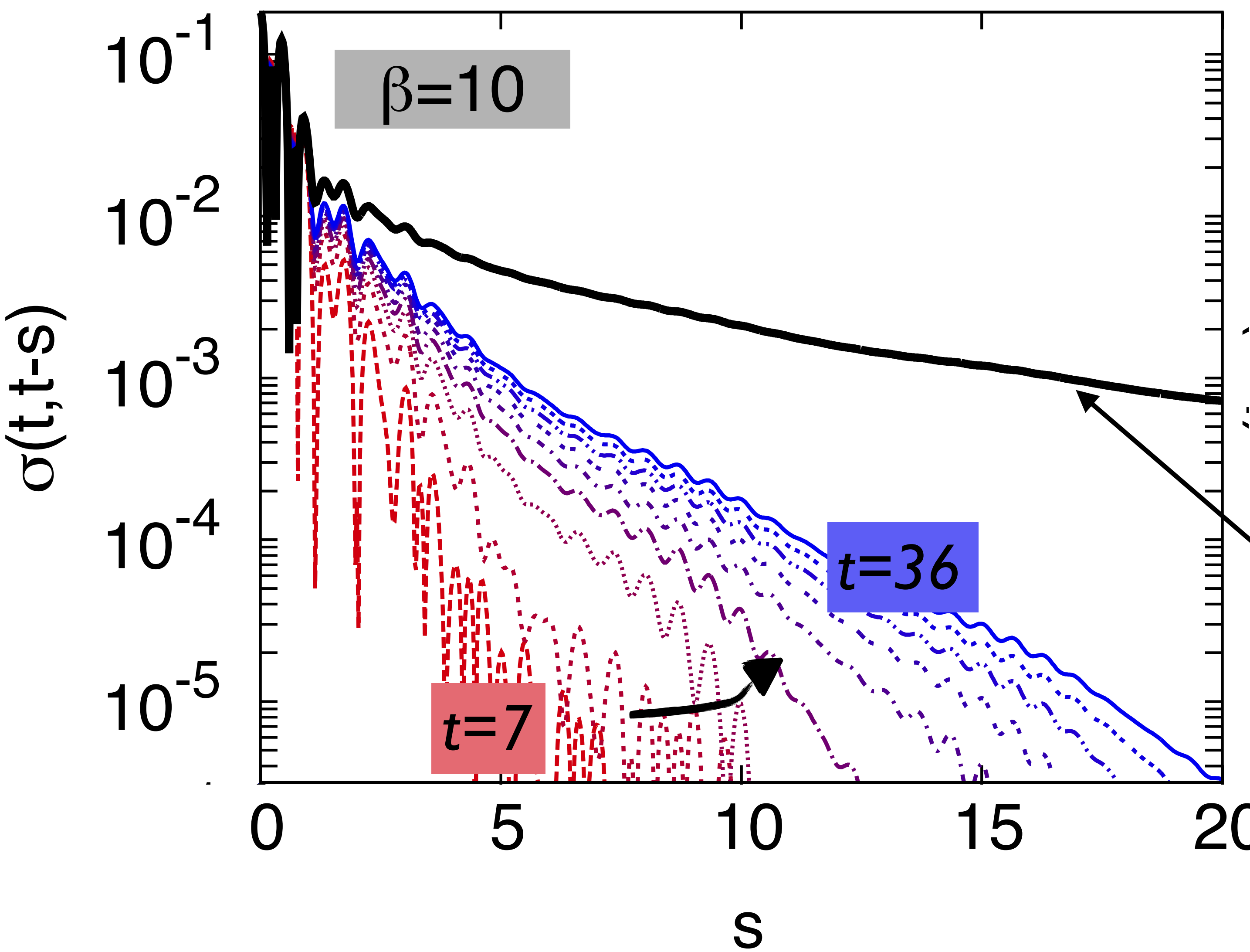
high bath temperature:
transport like in a bad metal
at same carrier density

doped Mott at $T=T_{bath}$:
# of holes = # holes + # doublo
in photo-doped case

Eckstein and Werner, PRL 2013

Quasi-particle formation at the Mott transition

Hubbard model plus bath (1d density of states, W=4)



doped Mott at $T=T_{bath}$:
# of holes = # holes + # doublons
in photo-doped case

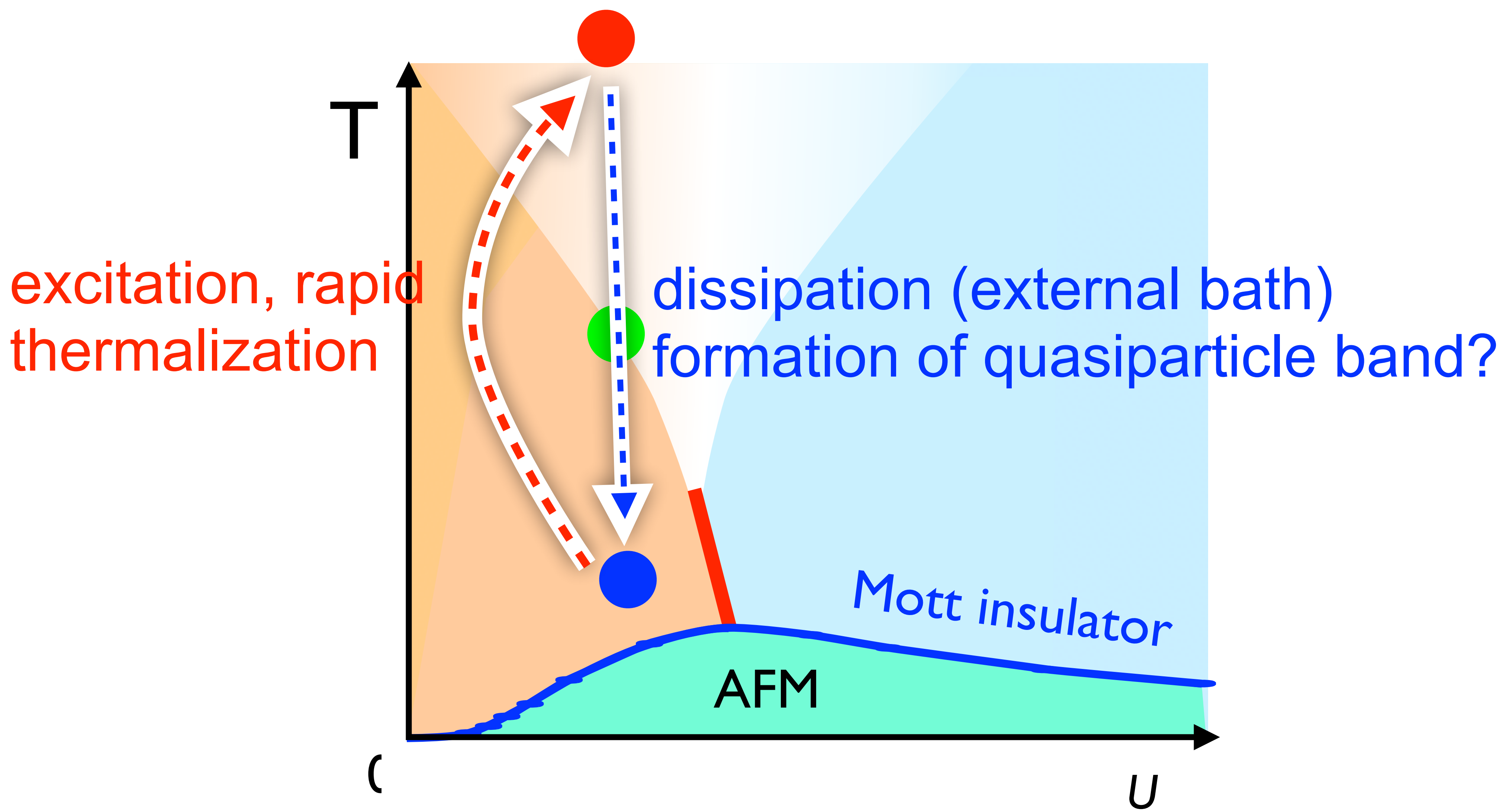
⇒ *Formation time of coherent metal much longer than scattering time of quasiparticles in metal*

Eckstein and Werner, PRL 2013

Quasi-particle formation at the Mott transition

Analogous setup close to the Mott transition:

Sayyad and Eckstein, PRL 2016

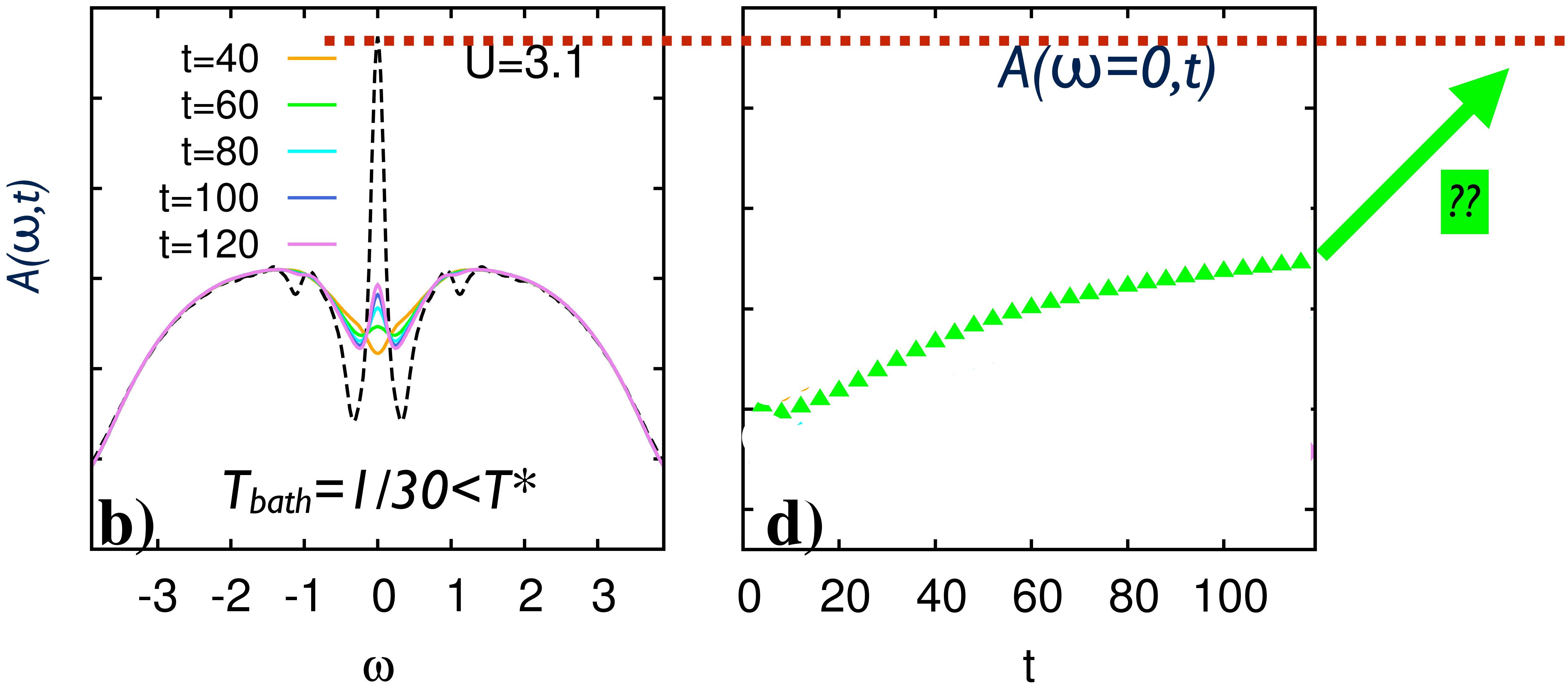


- Hubbard model plus bath $\Sigma_{bath}(t,t') = \lambda G(t,t') D_{bath}(t,t')$

initial state: atomic limit/rapid switch-on of the hopping (ramp time 2.5)

Quasi-particle formation at the Mott transition

- *slow formation of quasiparticle peak*
  (slow-down towards transition)



⇒ *intrinsic "bottleneck" of dynamics?*

failure of kinetic relaxation picture

units: *bandwidth=4 (Bethe lattice)* ⇒ *U_c=4.69 (in NCA: U_c ~ 3.4)*

Quasi-particle formation at the Mott transition

Identify "hidden slow degrees of freedom"? … Slave Rotor decoupling

electron = constrained composite particle

$$|0\rangle_c = |0\rangle_f |-1\rangle_\theta$$
$$|\uparrow\rangle_c = |\uparrow\rangle_f |0\rangle_\theta$$
$$|\downarrow\rangle_c = |\downarrow\rangle_f |0\rangle_\theta$$
$$|\uparrow\downarrow\rangle_c = |\uparrow\downarrow\rangle_f |1\rangle_\theta$$

quantum rotor $\theta \in [0, 2\pi)$

angular momentum = charge

$$\Rightarrow \quad c^\dagger_\sigma = \underbrace{e^{i\theta}}_{L_+} f^\dagger_\sigma$$

constraint: $L = \sum_\sigma f^\dagger_\sigma f_\sigma - 1$

$$U(n_\uparrow - \frac{1}{2})(n_\downarrow - \frac{1}{2}) + \epsilon_0 \sum_\sigma c^\dagger_\sigma c_\sigma = \frac{U}{2} L^2 + \epsilon_0 \sum_\sigma f^\dagger_\sigma f_\sigma$$

Florens & Georges Phys. Rev. B 66, 165111 (2002)

Quasi-particle formation at the Mott transition

$$H_{imp} = U(n_\uparrow - \frac{1}{2})(n_\downarrow - \frac{1}{2}) + \sum_{p\sigma} V_p c_\sigma^\dagger a_{p\sigma} + h.c. + \sum_{p\sigma} \epsilon_p a_\sigma^\dagger a_{p\sigma}$$

$$= \frac{U}{2} L^2 + \sum_{p\sigma} V_p e^{i\theta} f_\sigma^\dagger a_{p\sigma} + h.c. + \sum_{p\sigma} \epsilon_p a_\sigma^\dagger a_{p\sigma}$$

constrained complex field (like particle in 2d)

$$X = e^{i\theta} \quad |X|^2 = 1$$

Florens & Georges Phys. Rev. B 66, 165111 (2002)

Quasi-particle formation at the Mott transition

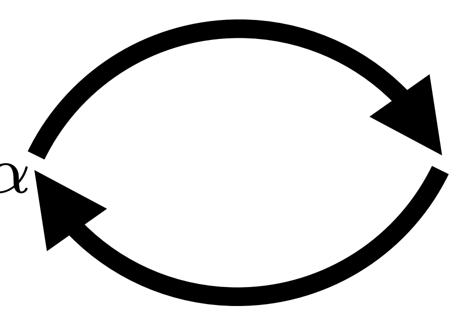
$$\longrightarrow \quad G_f(t,t') = -i\langle T_C f_\sigma(t) f^*_\sigma(t')\rangle$$

$$\dashrightarrow \quad G_X(t,t') = -i\langle T_C X(t) X^*(t')\rangle$$

Gx related to charge correlation function:

$$\chi_c(\tau) \equiv \left\langle \sum_\sigma \left(n_\sigma(\tau) - \frac{1}{2}\right) \sum_{\sigma'} \left(n_{\sigma'}(0) - \frac{1}{2}\right) \right\rangle = \left\langle \hat{L}(\tau)\hat{L}(0) \right\rangle$$

$$\vec{S} = \sum_{\sigma\sigma'} f^\dagger_\sigma \vec{\tau}_{\sigma\sigma'} f_{\sigma'} \qquad \langle S_\alpha(t) S^{(}_{\beta}0)\rangle = \tau_\alpha$$



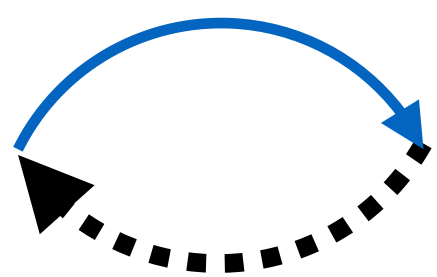
$$\tau_\beta + \ldots$$

Florens & Georges Phys. Rev. B 66, 165111 (2002)

Quasi-particle formation at the Mott transition

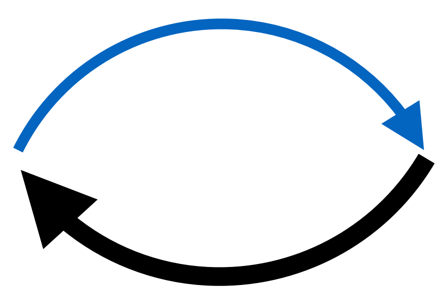
treat constraints on average, lowest order bold perturbation expansion …

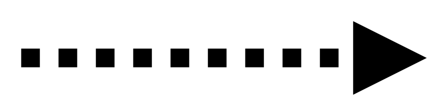
$$H_{int} = \sum_{p\sigma} V_p c_\sigma^\dagger a_{p\sigma} + h.c. = \sum_{p\sigma} V_p X^* f_\sigma^\dagger a_{p\sigma} + h.c.$$

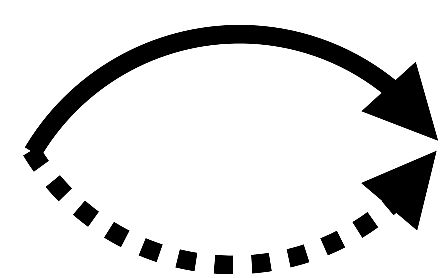
$\Sigma_f =$ 

 $= \Delta(t,t')$

 $G_f(t,t') = -i\langle T_C f_\sigma(t) f_\sigma^*(t')\rangle$

$\Sigma_X =$ 

 $G_X(t,t') = -i\langle T_C X(t) X^*(t')\rangle$

$G = -i\langle T_C c(t) c^\dagger(t')\rangle =$ 

+ constraints

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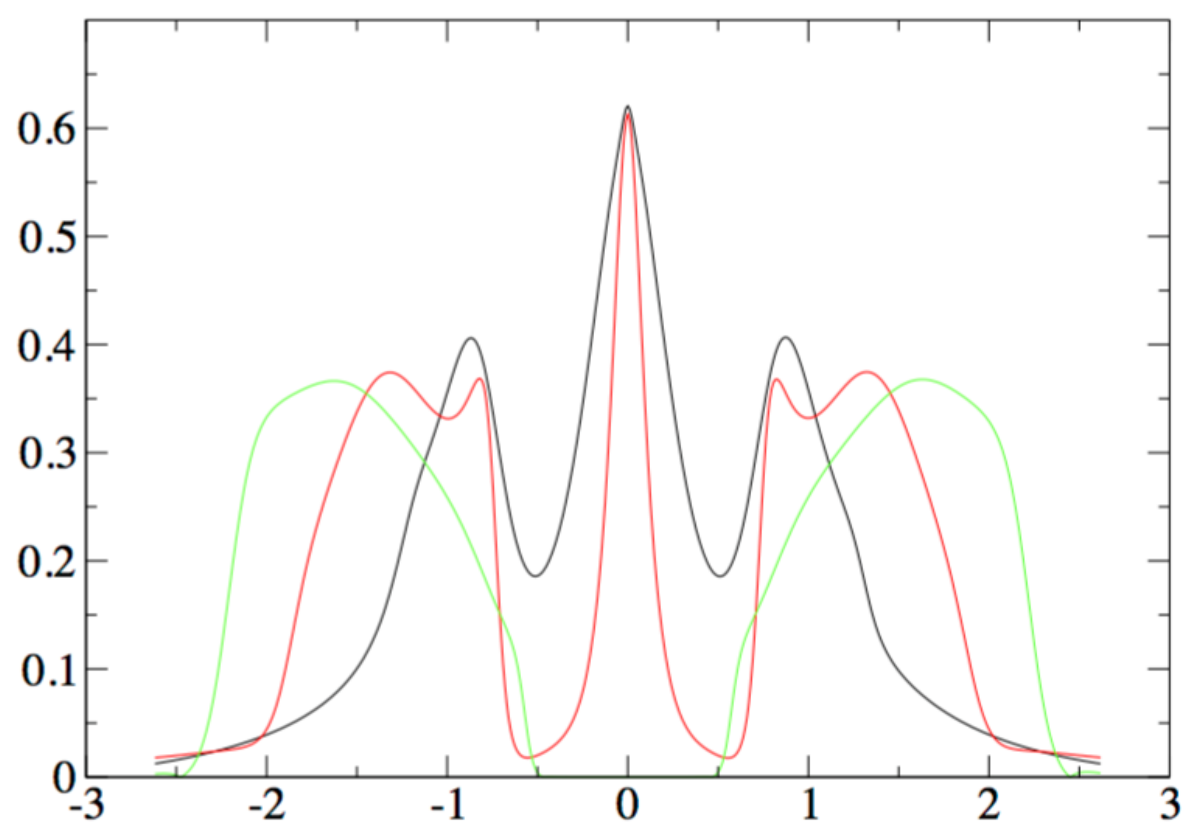
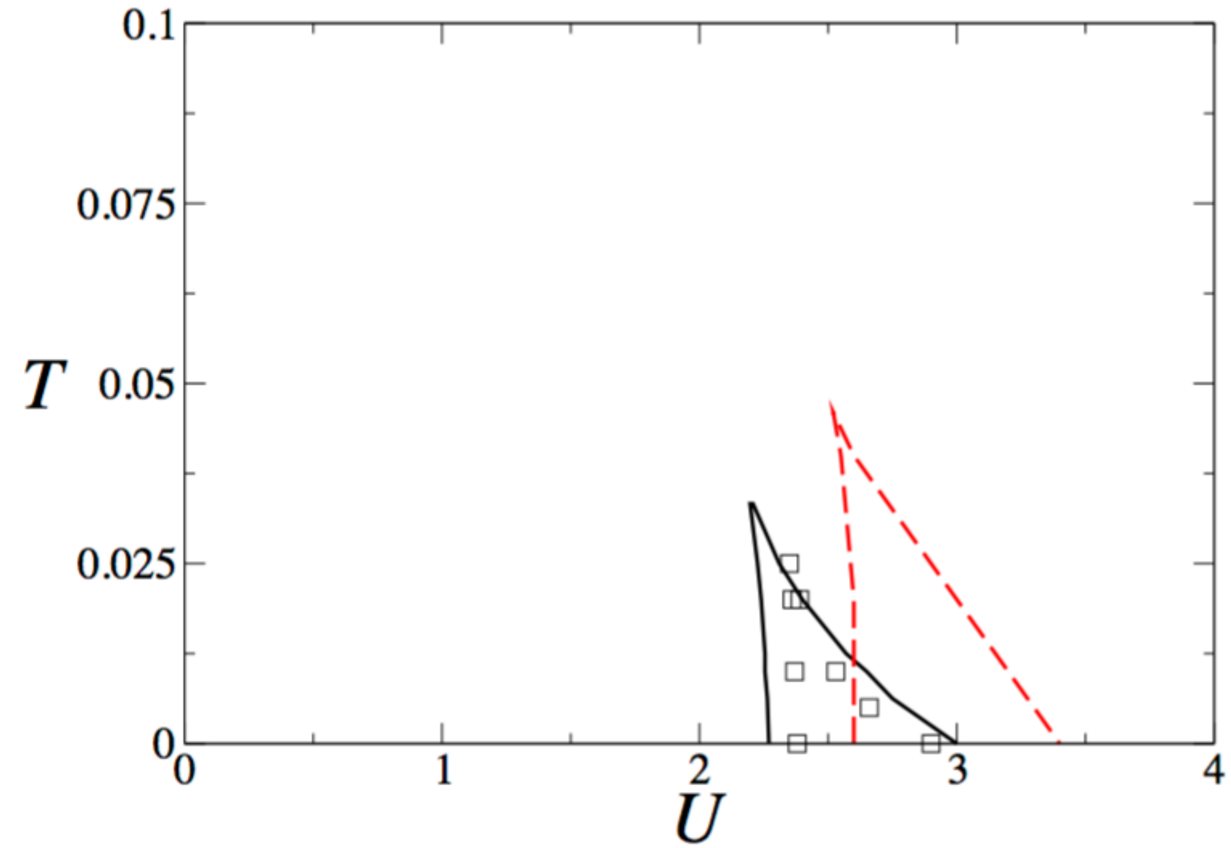
Quasi-particle formation at the Mott transition

U=1,2,3, bandwidth=2





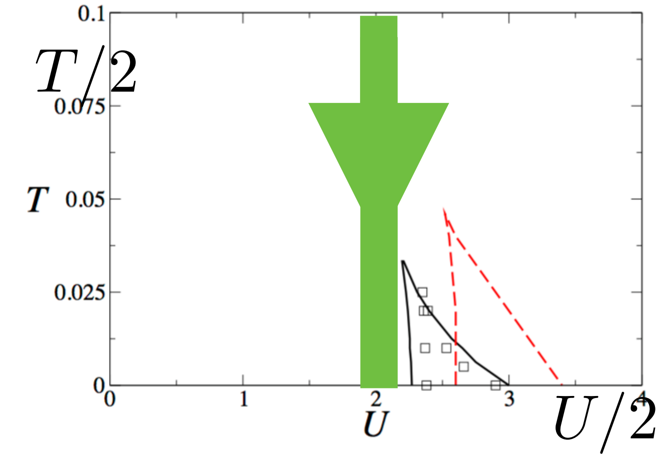
Qualitative picture of the Mott transition
(quasiparticle peak and Hubbard bands)

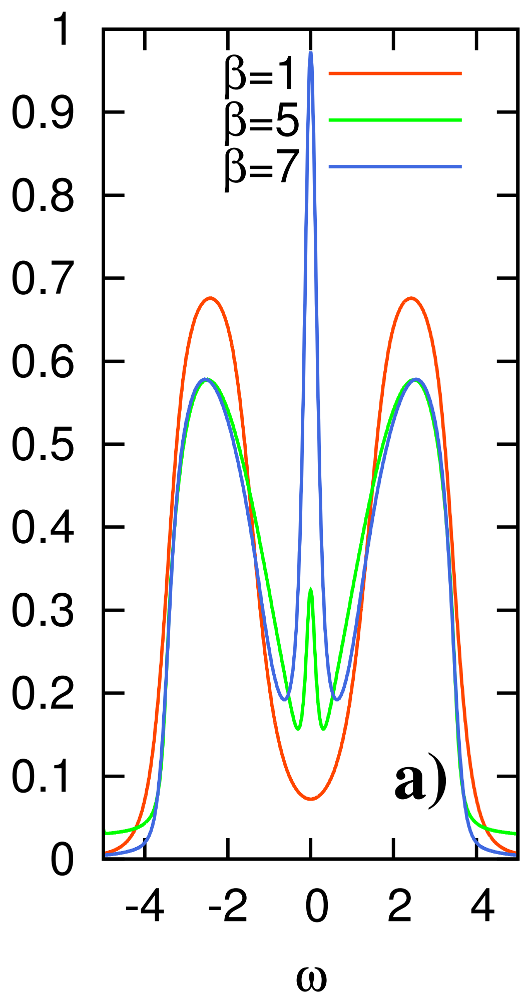
Florens & Georges Phys. Rev. B 66, 165111 (2002)

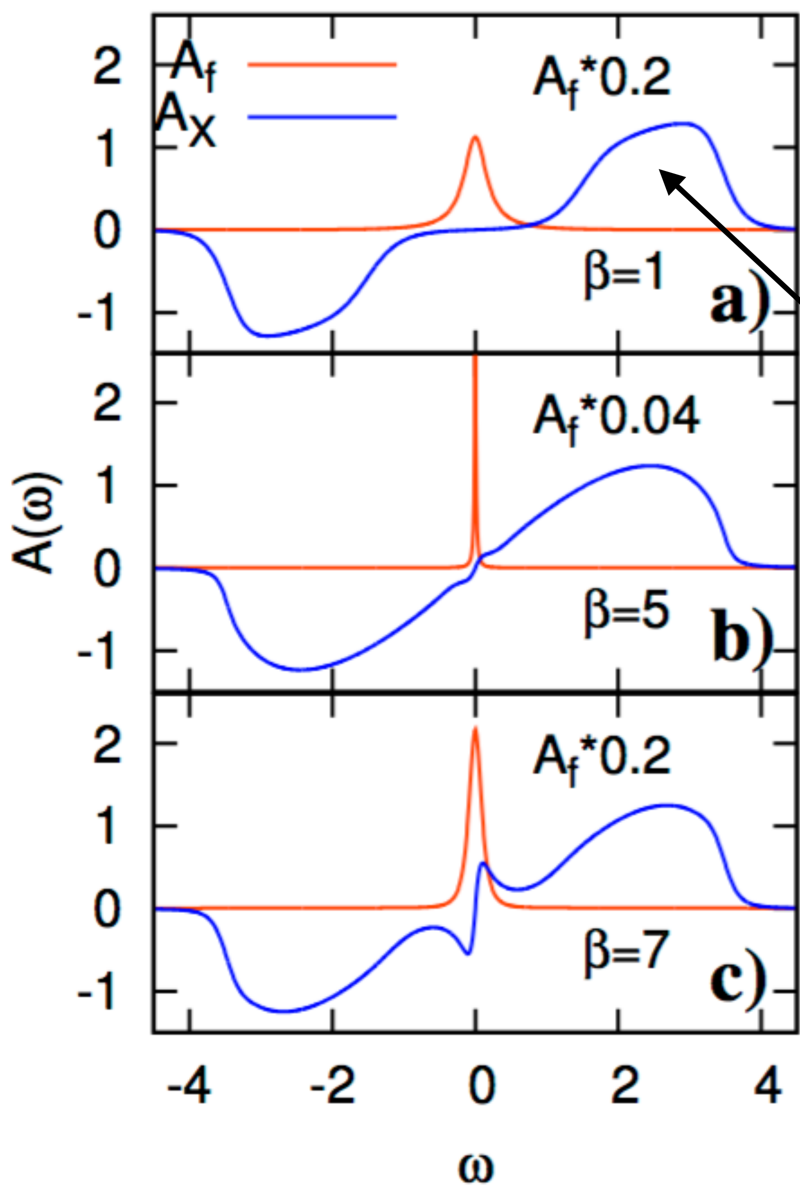
Quasi-particle formation at the Mott transition

Crossover regime:

$$A_{f,X}(\omega) = -\frac{1}{\pi}\mathrm{Im}G^R_{f,X}(\omega + i0)$$

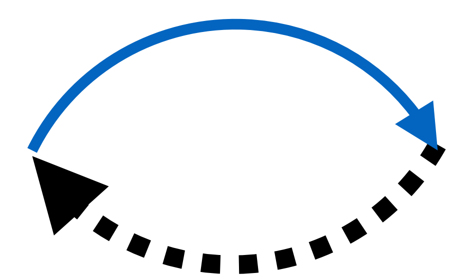






High-temperature:
spinon couples to thermal charge fluctuations

$\Sigma_f =$

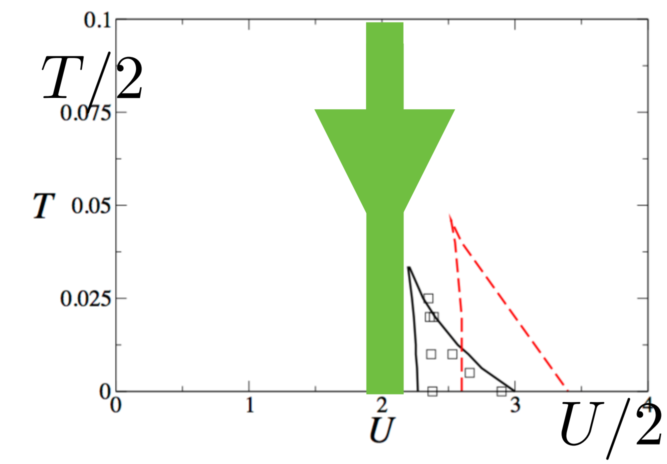


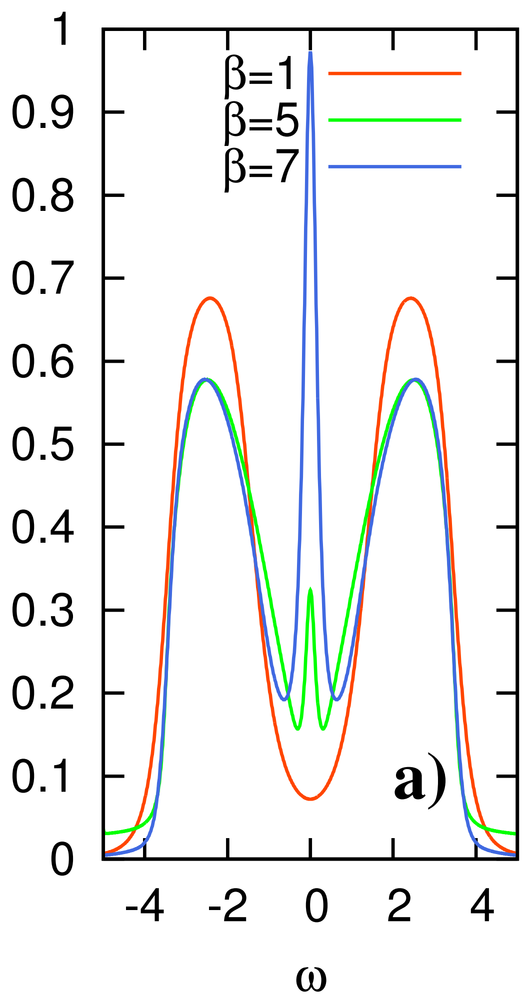
$$-\mathrm{Im}\Sigma_(\omega = 0) \propto \int d\omega \frac{\Delta(\omega)A_X(\omega)}{\cosh(\omega/2T)}$$

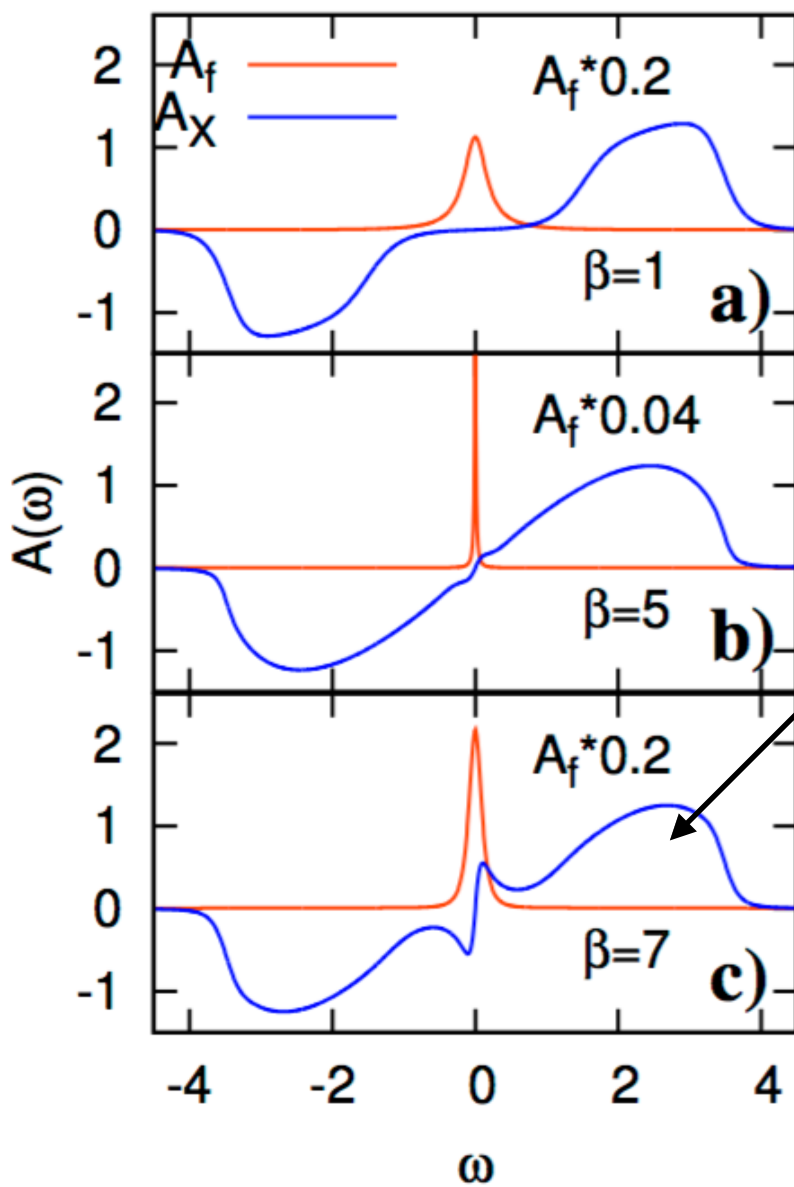
Quasi-particle formation at the Mott transition

Crossover regime:

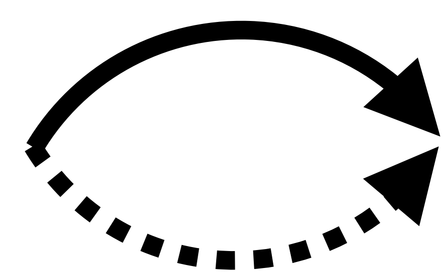
$$A_{f,X}(\omega) = -\frac{1}{\pi}\mathrm{Im}G^R_{f,X}(\omega + i0)$$







Rotor and spinon develop low energy spectral weight

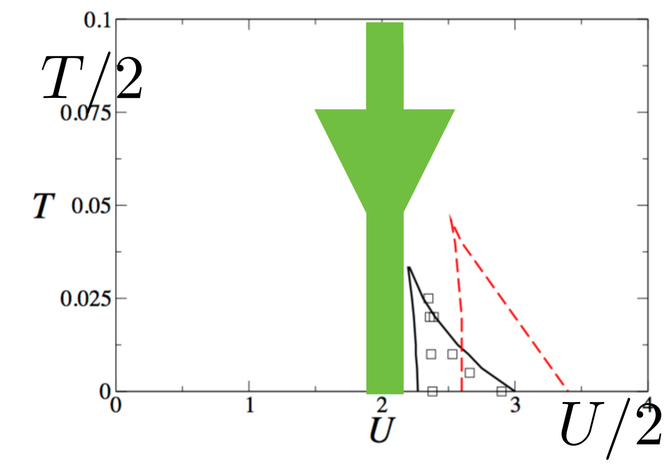
$G =$ 

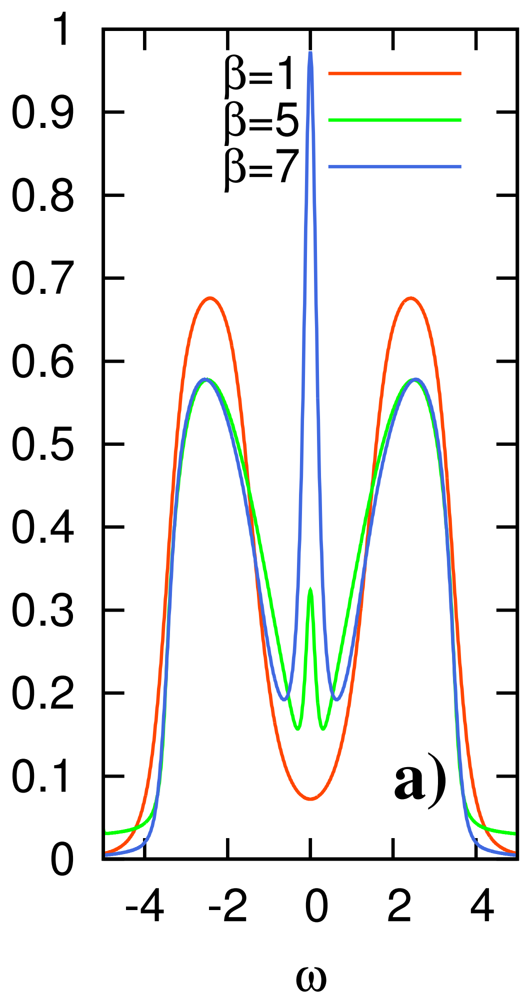
$$A(\omega) \propto \int d\omega' \frac{A_f(\omega')A_X(\omega-\omega')}{\cosh((\omega-\omega')/2T)}$$

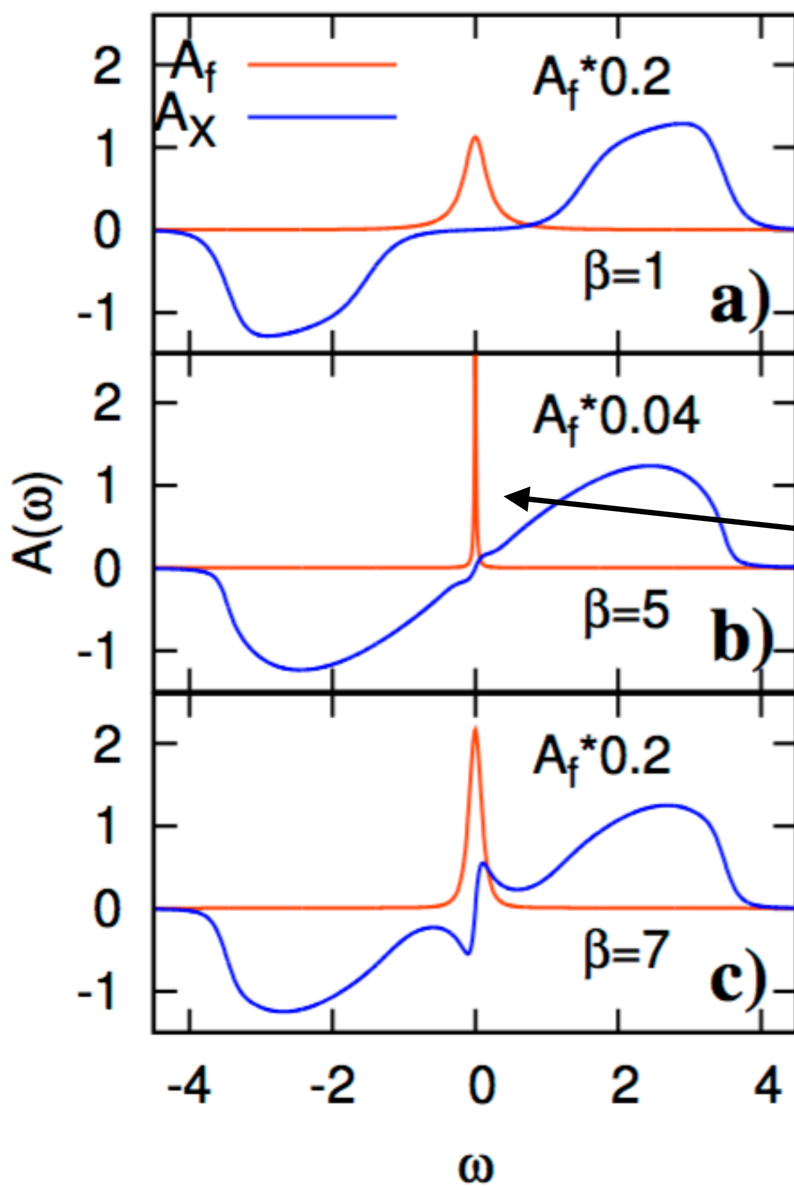
Quasi-particle formation at the Mott transition

Crossover regime:

$$A_{f,X}(\omega) = -\frac{1}{\pi}\mathrm{Im}G^R_{f,X}(\omega + i0)$$

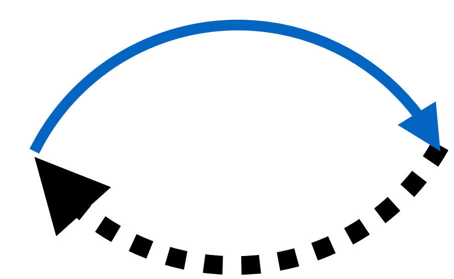






"Energetic decoupling" of spinon and rotor

$$\Sigma_f =$$

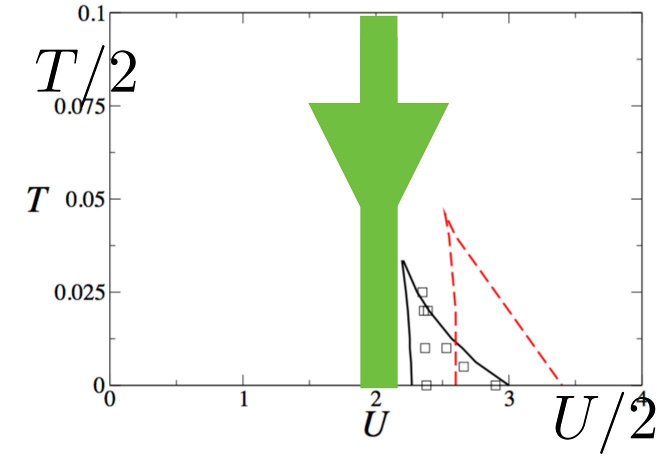


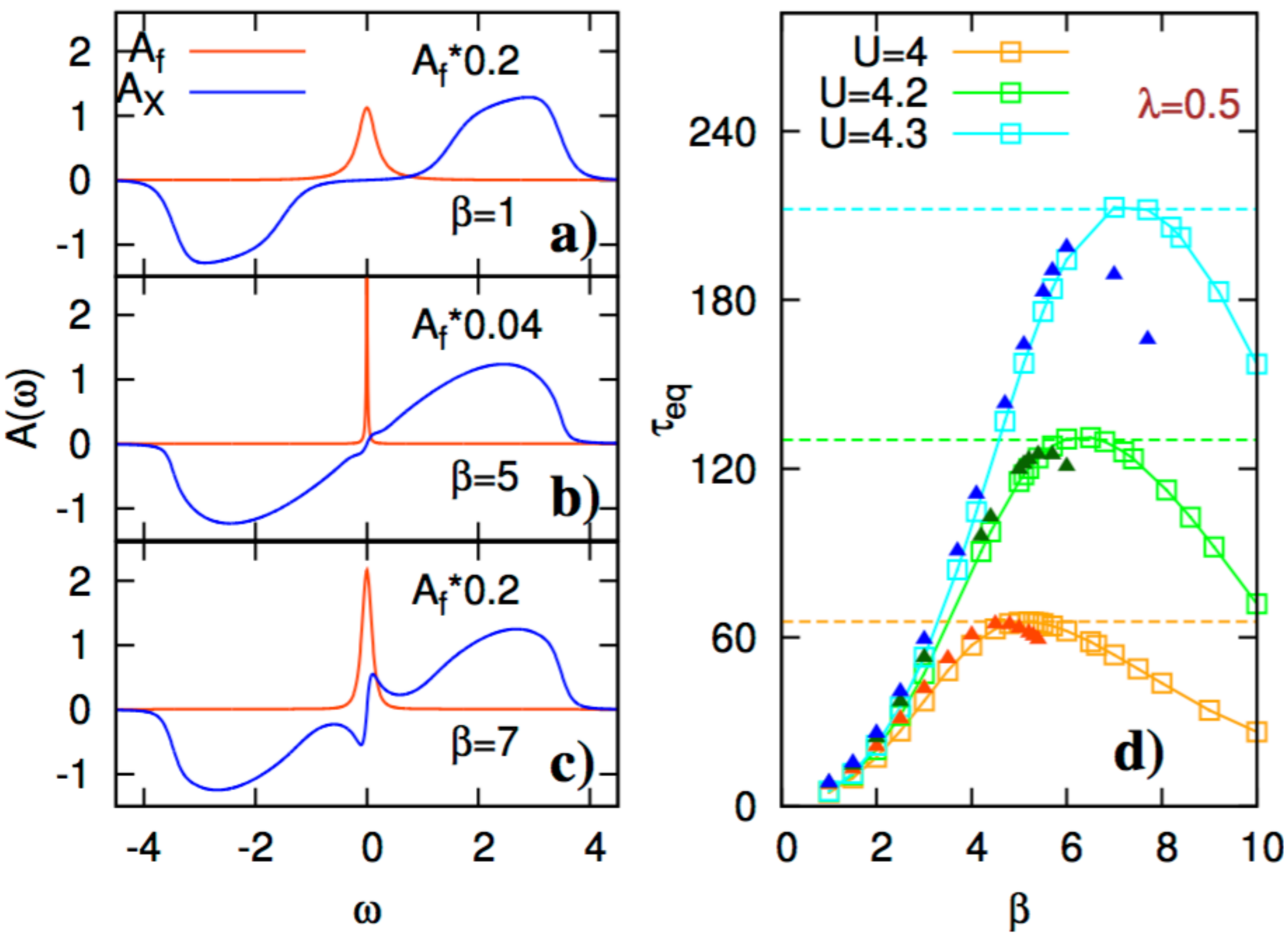
$$-\mathrm{Im}\Sigma_(\omega = 0) \propto \int d\omega \frac{\Delta(\omega) A_X(\omega)}{\cosh(\omega/2T)}$$

Quasi-particle formation at the Mott transition

Crossover regime:

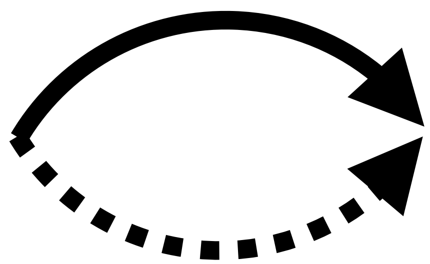
$$A_{f,X}(\omega) = -\frac{1}{\pi}\mathrm{Im}G^{R}_{f,X}(\omega + i0)$$





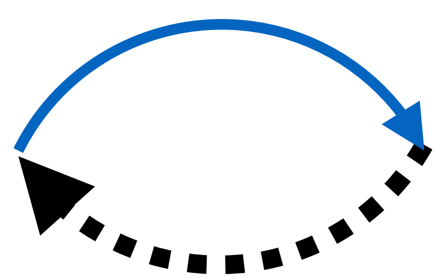
non-monotonous evolution of spinon lifetime as a function of temperature trough crossover

Quasi-particle formation at the Mott transition

$G =$ 

$$A(\omega) \propto \int d\omega' \frac{A_f(\omega')A_X(\omega-\omega')}{\cosh((\omega-\omega')/2T)} \quad \approx \frac{A_X(\omega)}{\cosh(\omega/2T)}$$

small spinon lifetime: spectrum dominated by charge

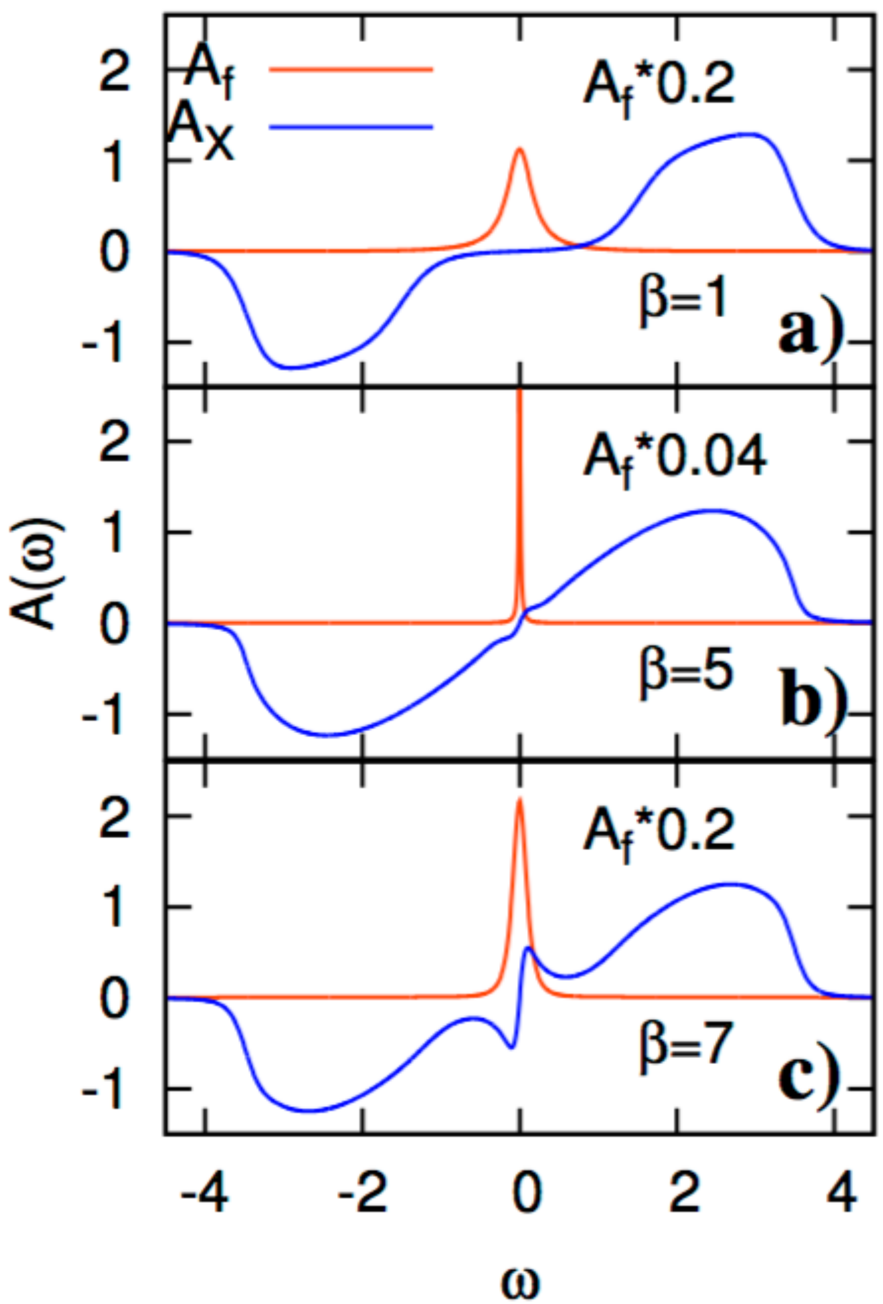
$\Sigma_f =$ 

$$-\mathrm{Im}\Sigma_{(}\omega = 0) \propto \int d\omega \frac{\Delta(\omega)A_X(\omega)}{\cosh(\omega/2T)} \approx \int d\omega \frac{\Delta(\omega)A(\omega)}{\cosh^2((\omega)/2T)}$$

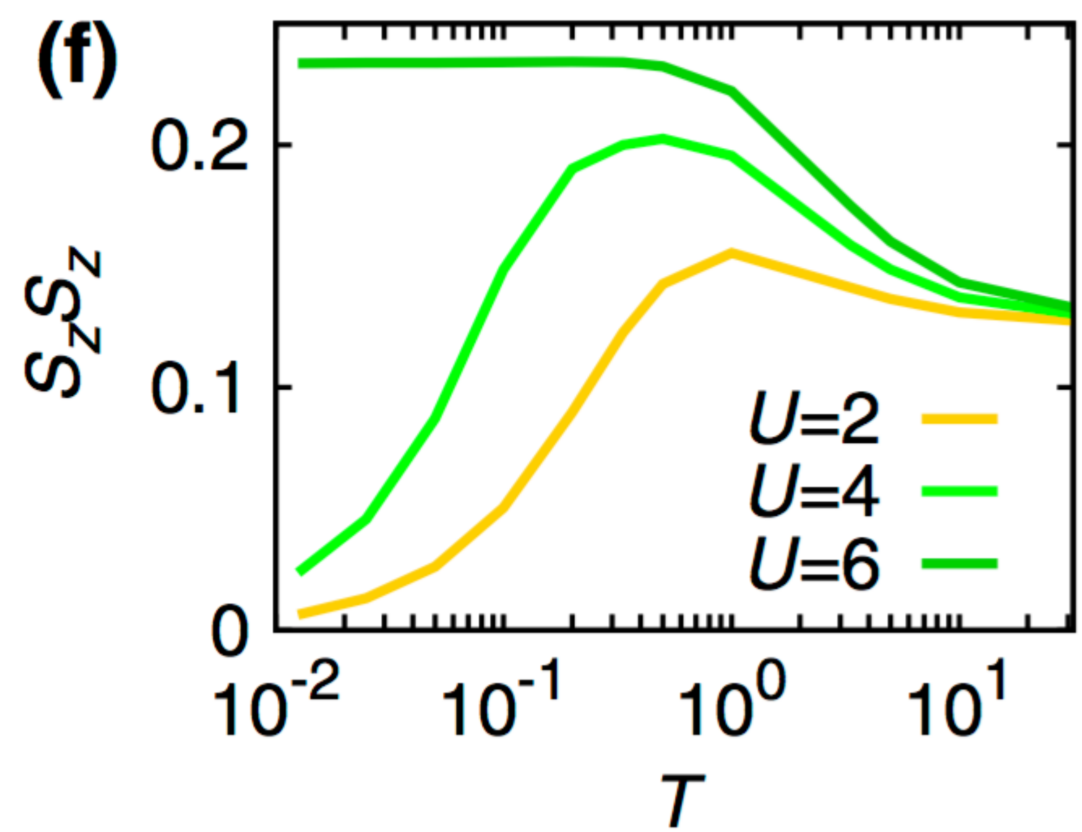
Quasi-particle formation at the Mott transition

Crossover regime:

$$A_{f,X}(\omega) = -\frac{1}{\pi}\mathrm{Im}G^R_{f,X}(\omega + i0)$$



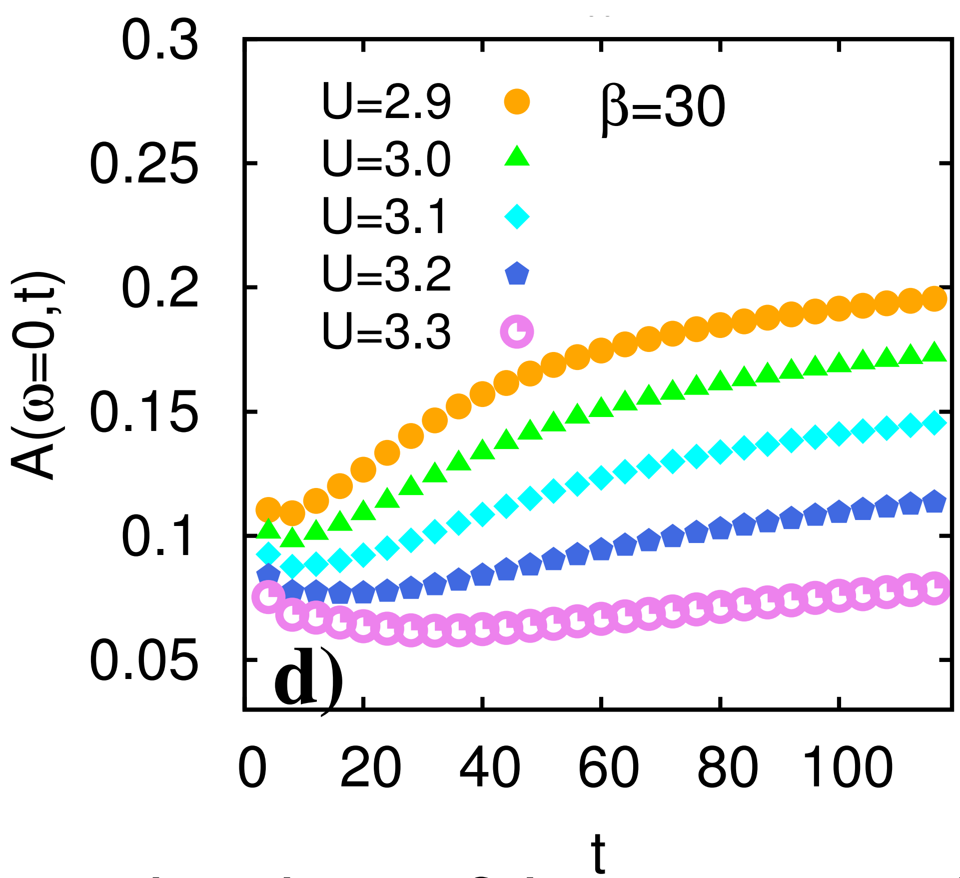
For comparison: nonmonotonous behavior of spin correlation function (from CTQMC)

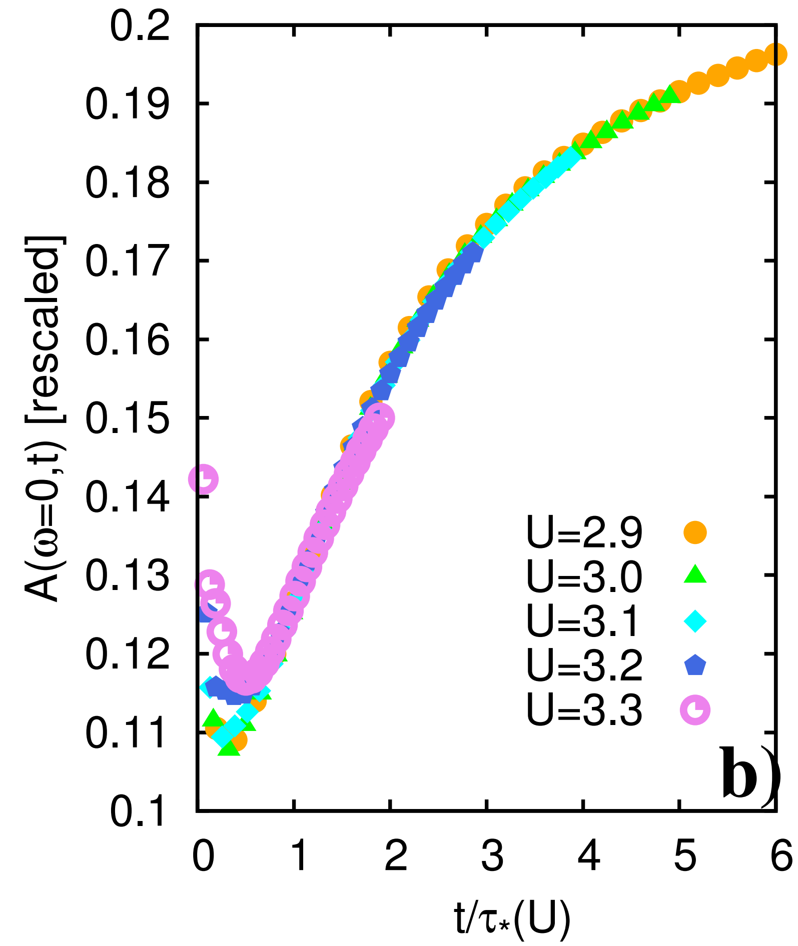


$$(1/\beta)\int_0^\beta d\tau\langle S_z(\tau)S_z(0)\rangle$$

Formation of well-defined moments

Quasi-particle formation at the Mott transition





slow-down of dynamics towards metal-insulator transition

$\Rightarrow$ *rescaling* $A(\omega = 0, t) = a_U f(t/\tau_*(U))$ $\tau_{\mathrm{eq}}^{-1}(T) = -\int d\omega \, \frac{\Delta''(\omega) A(\omega)}{\cosh(\omega/2T)^2}$

slave rotor dynamics in Keldysh $\Rightarrow$ bottleneck because charge and spinon do not talk

"schizophrenic electrons"