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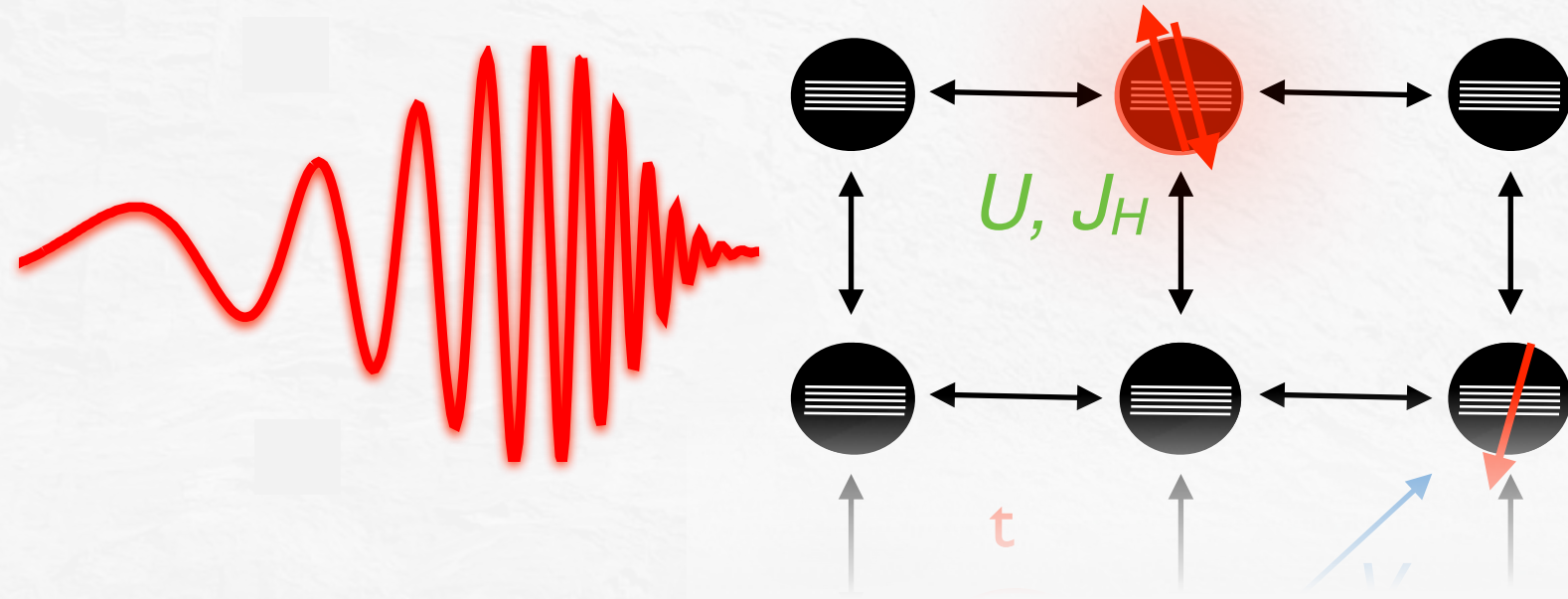


Non-equilibrium Superconductivity in strongly correlated systems: Insights from DMFT

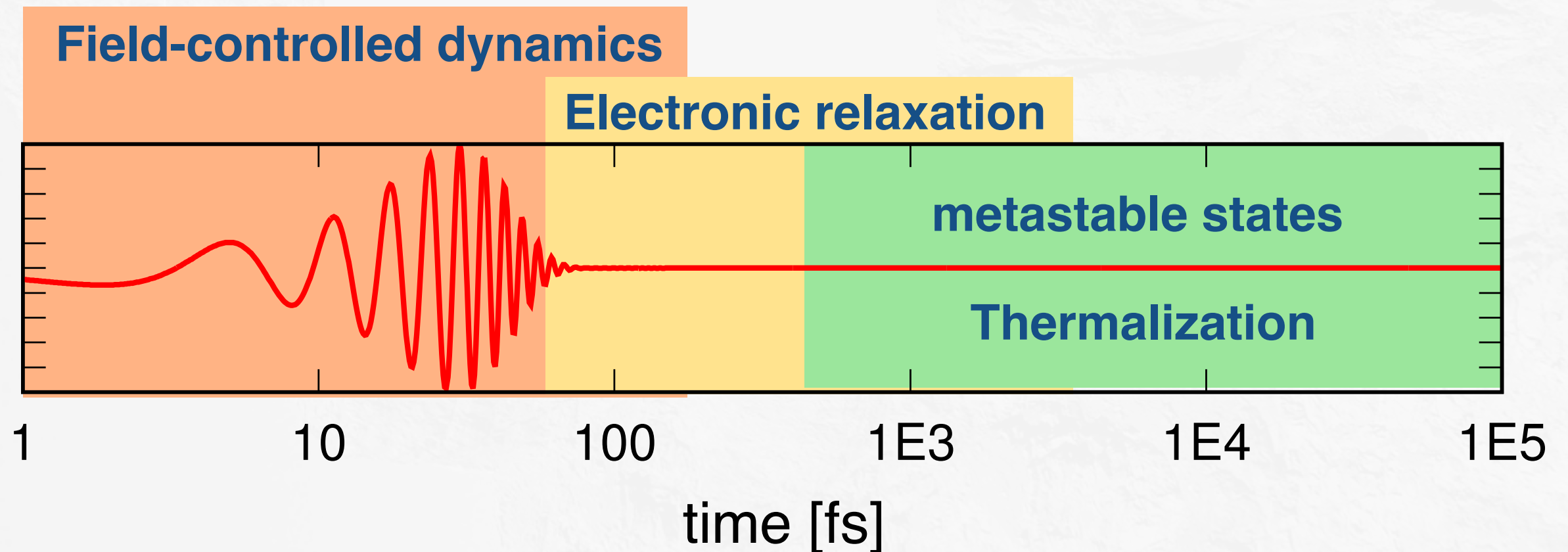
Martin Eckstein

Flatiron Institute, New York, January 13, 2020

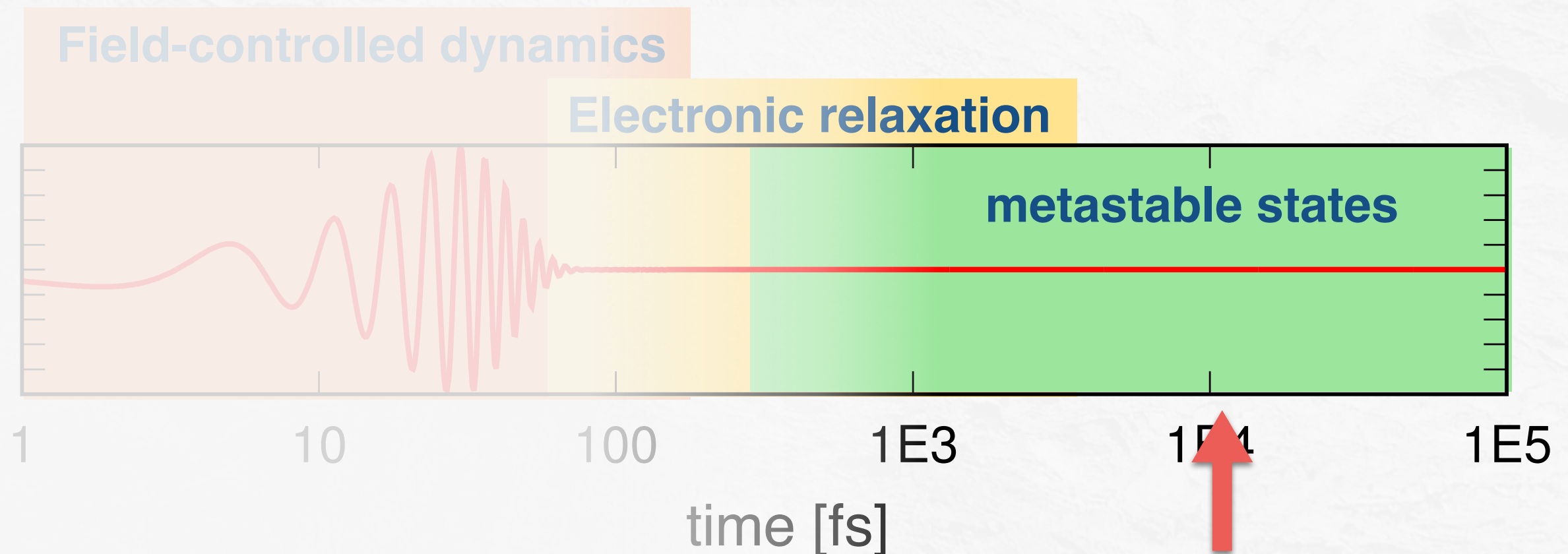
Outline



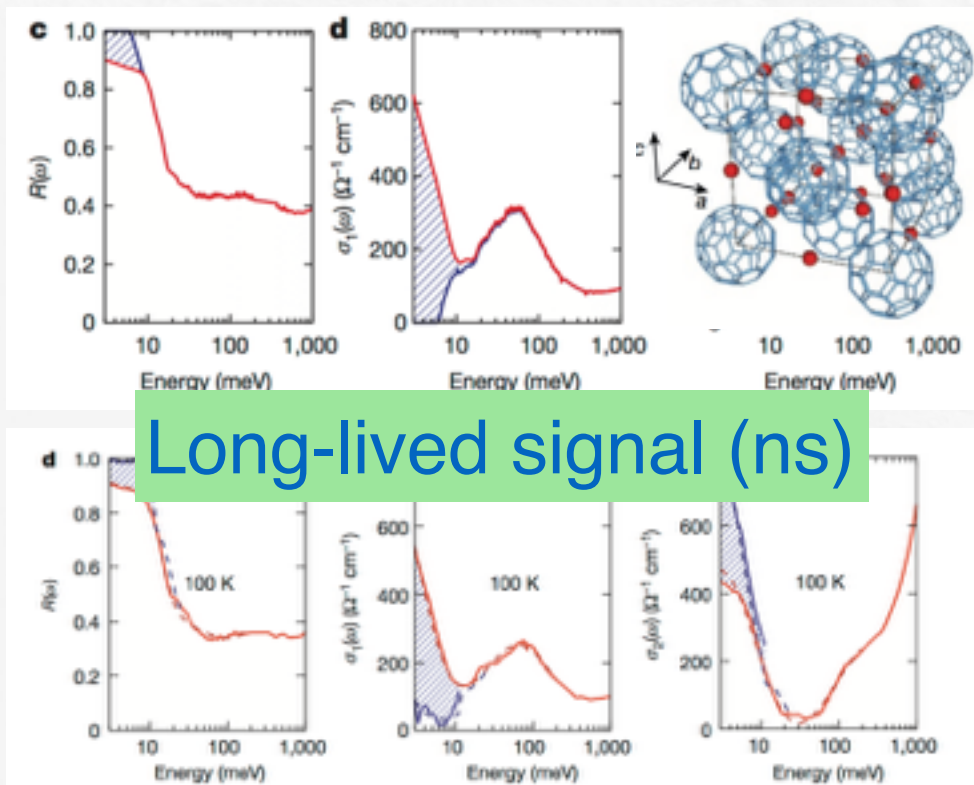
Nonequilibrium phases
in light-driven correlated
electrons?



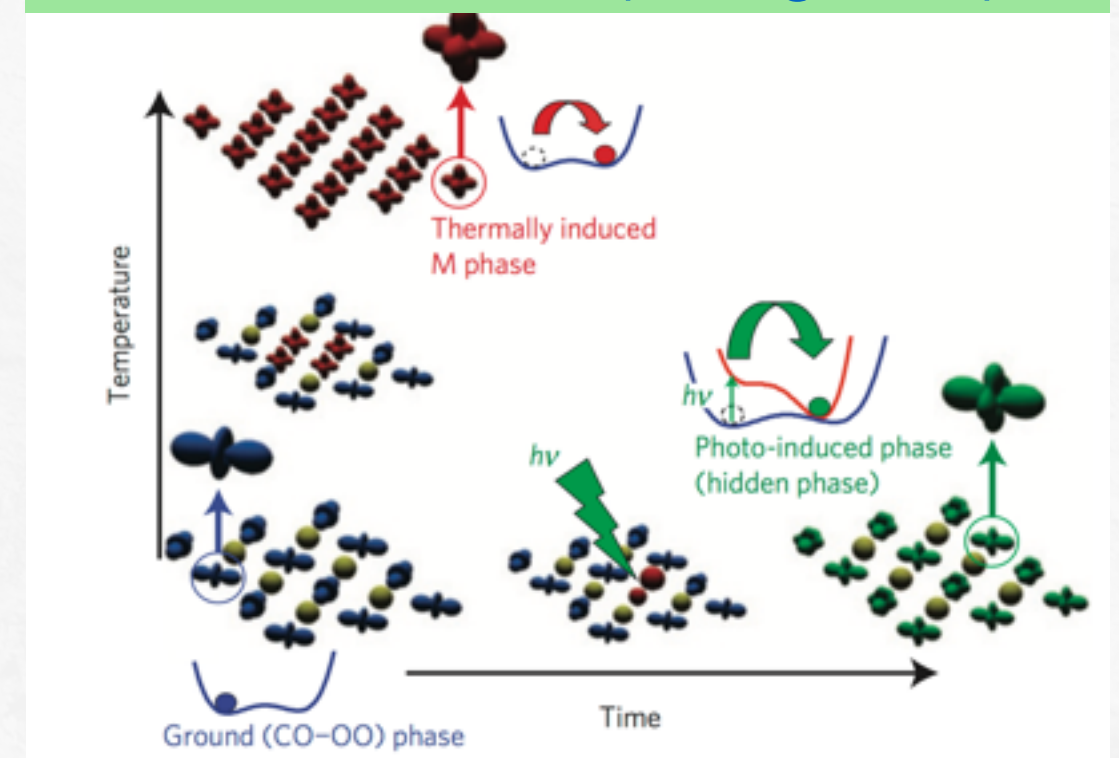
Outline



Light-induced SC, Fullerides



Hidden states (manganite)



Mitrano, et al. et Cavalleri, Nature (2016)

Ichikawa et al. Nat. Mat. (2011)

Photo-doped states in the Hubbard model

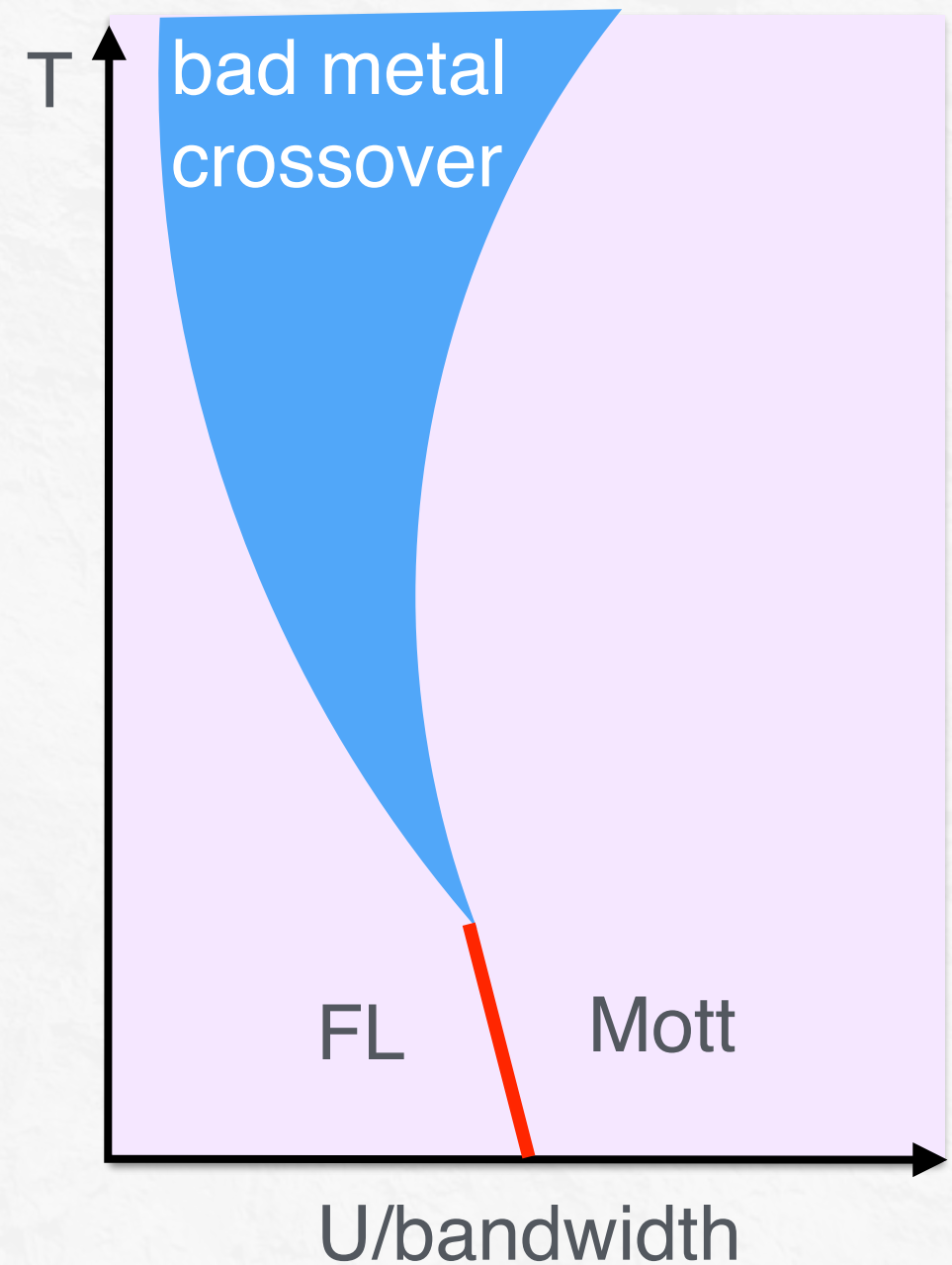
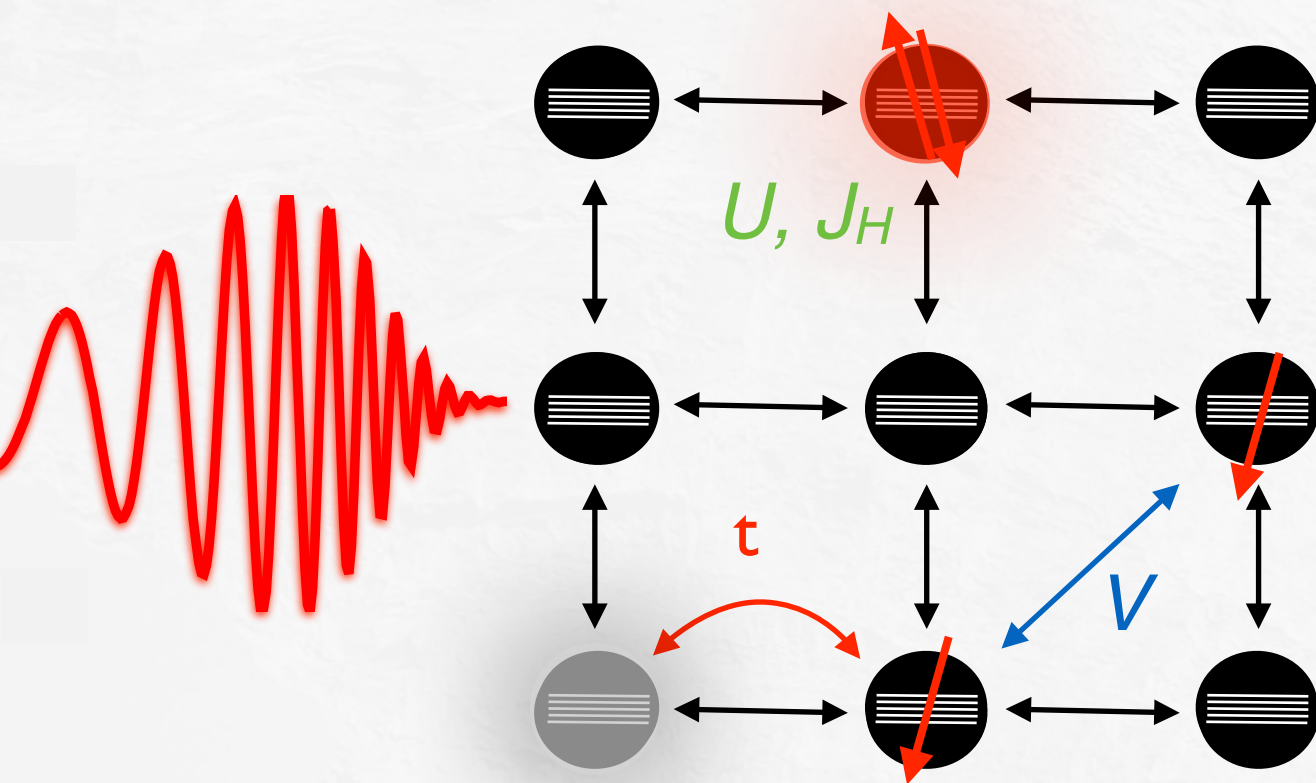
η -superconductivity in the photo-doped Hubbard model

Photo-induced evaporative cooling

Comments on driven systems

Methods

$$H = \sum_{ij} \sum_{\alpha\beta} t_{ij}^{\alpha\beta} c_{i\alpha}^\dagger c_{j\beta} e^{iq\vec{A}(t) \cdot (\vec{R}_i - \vec{R}_j)} + \sum_i U_{\alpha,\alpha'\beta\beta'} c_{i\alpha}^\dagger c_{i\alpha'}^\dagger c_{i\beta'} c_{i\beta}$$



Local (and non-local) interactions

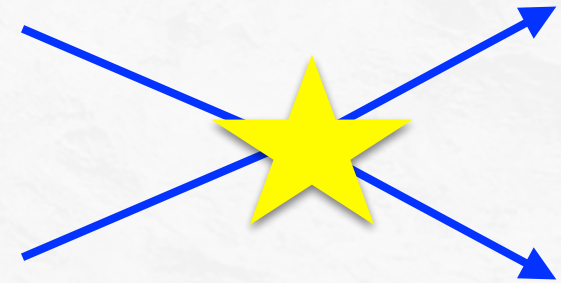
External electromagnetic fields

Phonons, dissipative environment,

Methods

Kinetic equations:

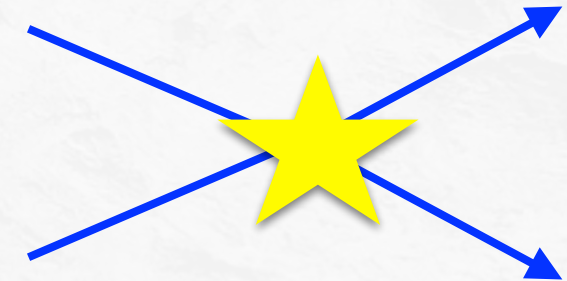
well-defined quasiparticles
& scattering interactions



Methods

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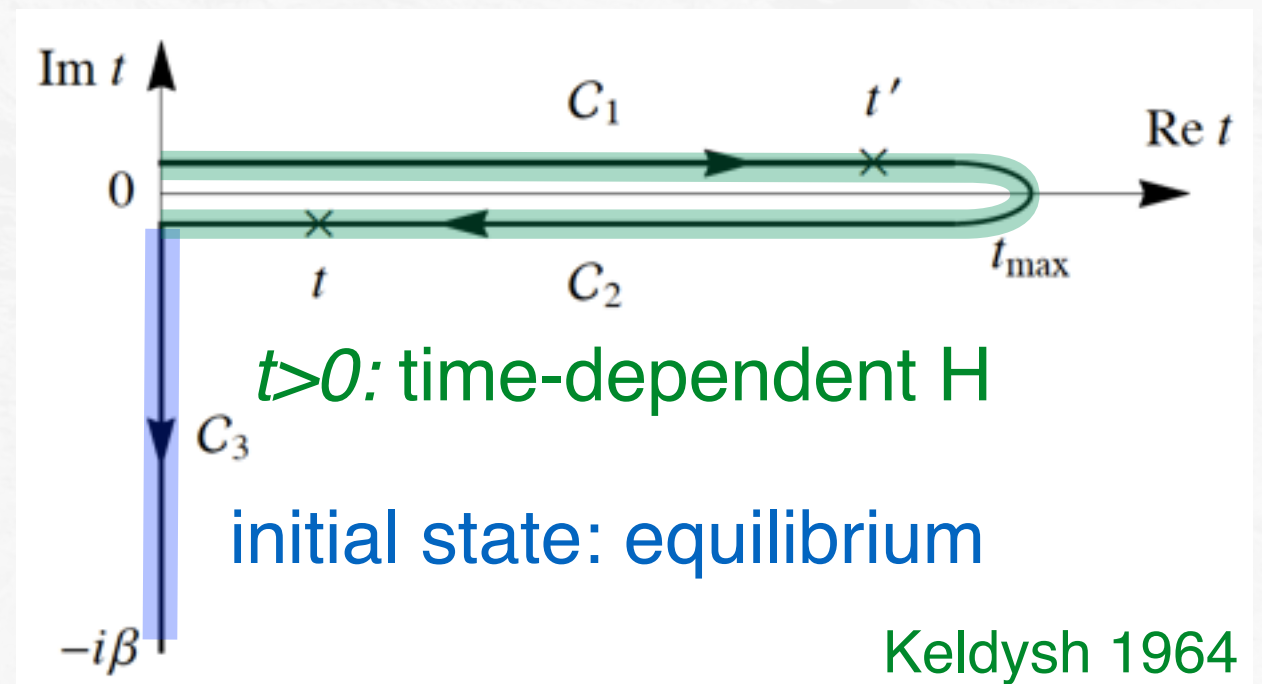
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non-equilibrium Green's functions:

(Quantum kinetic theory without
quasiparticle or relaxation time
approximations)

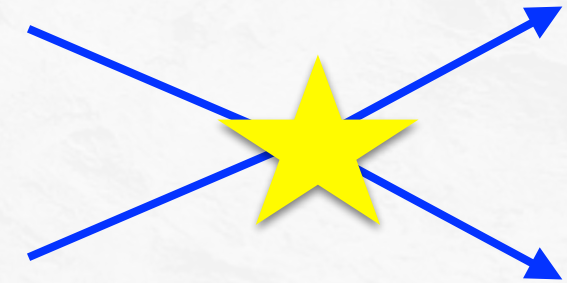
intertwined evolution of electronic
structure & interactions



Methods

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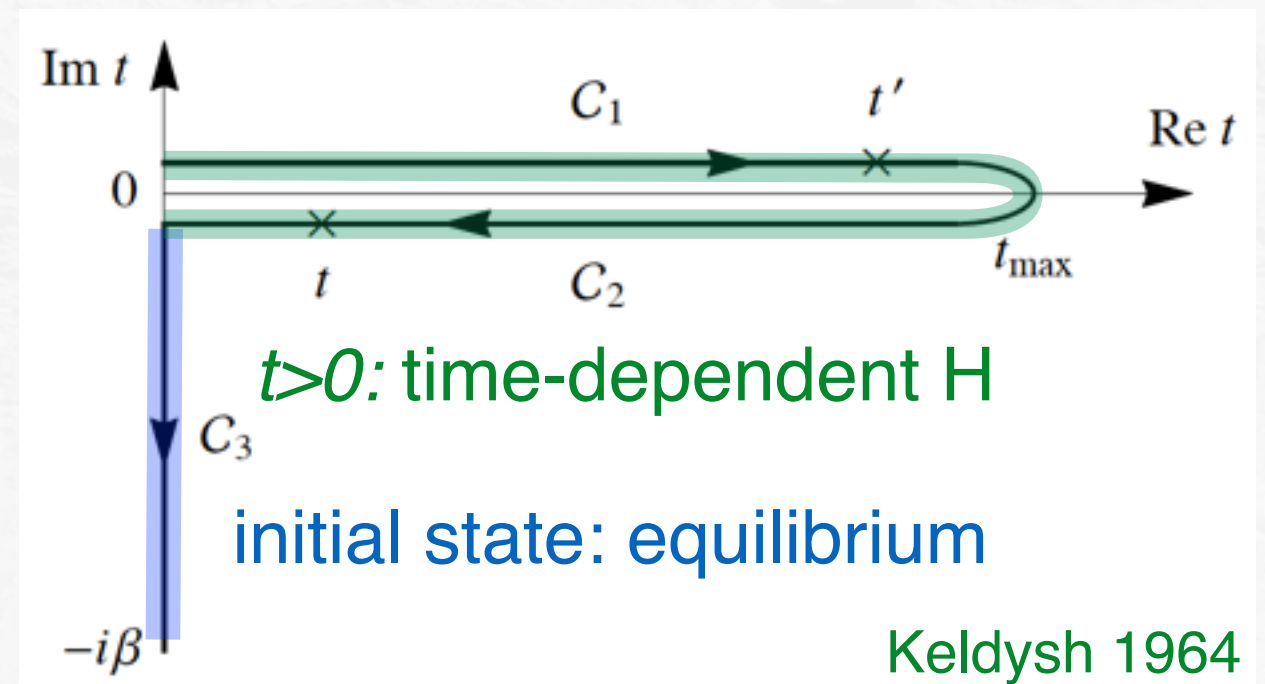
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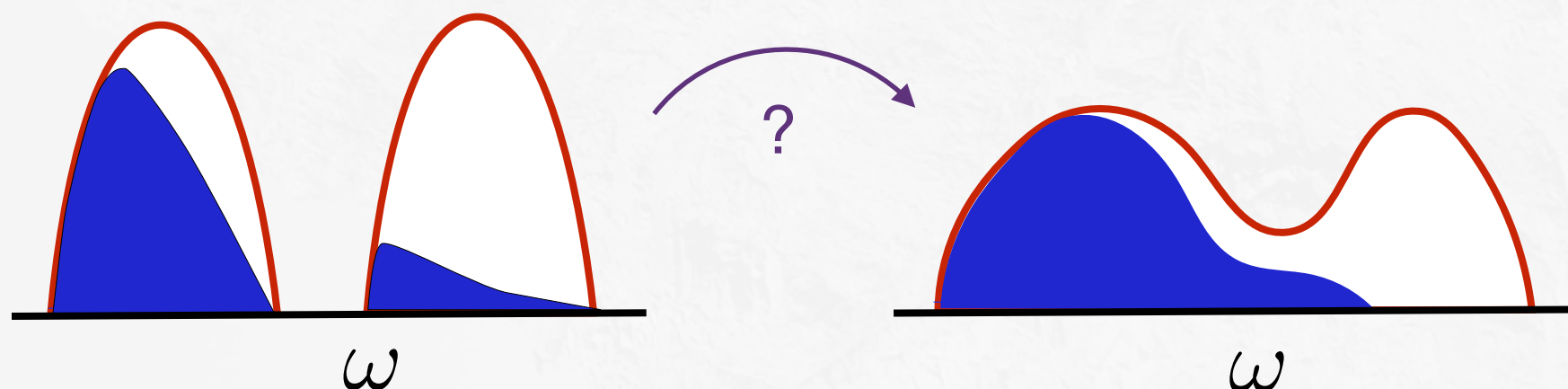
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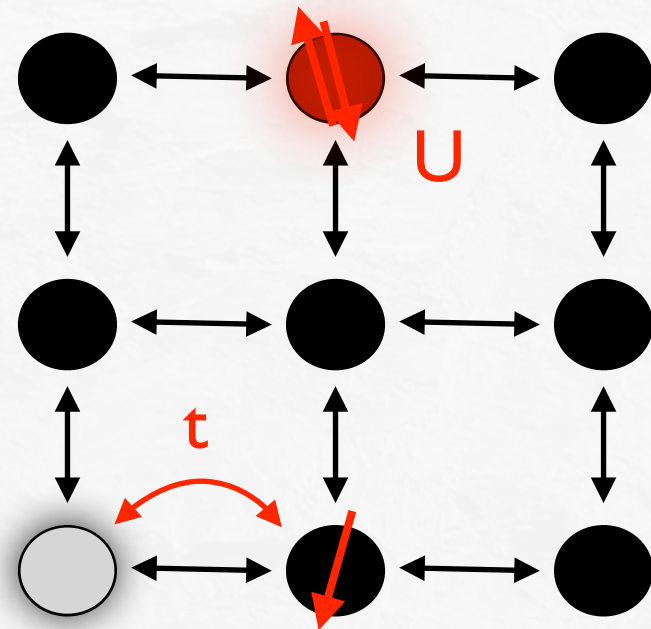
intertwined evolution of electronic
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spectrum $\Leftrightarrow$ occupied density of states (trPES)



non-equilibrium DMFT ++



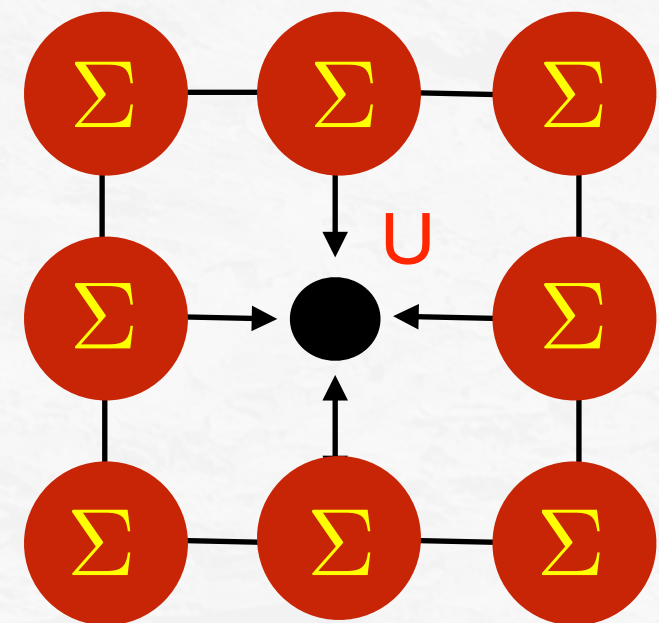
Classical diagrammatic perturbation theory

$$\overline{G} = \overline{G_0} + \overline{G_0} \Sigma \overline{G_0}$$

strong correlations: NEGF embedding

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

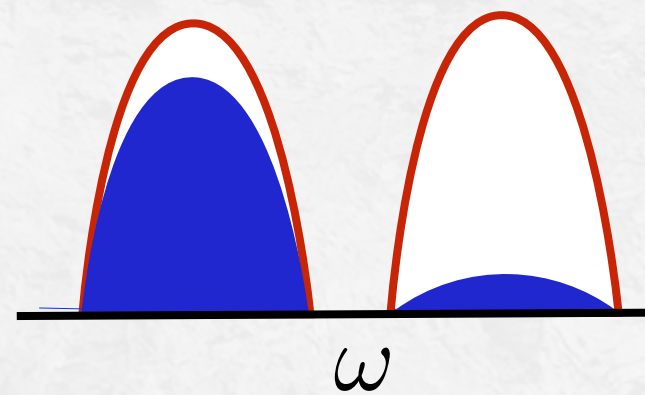
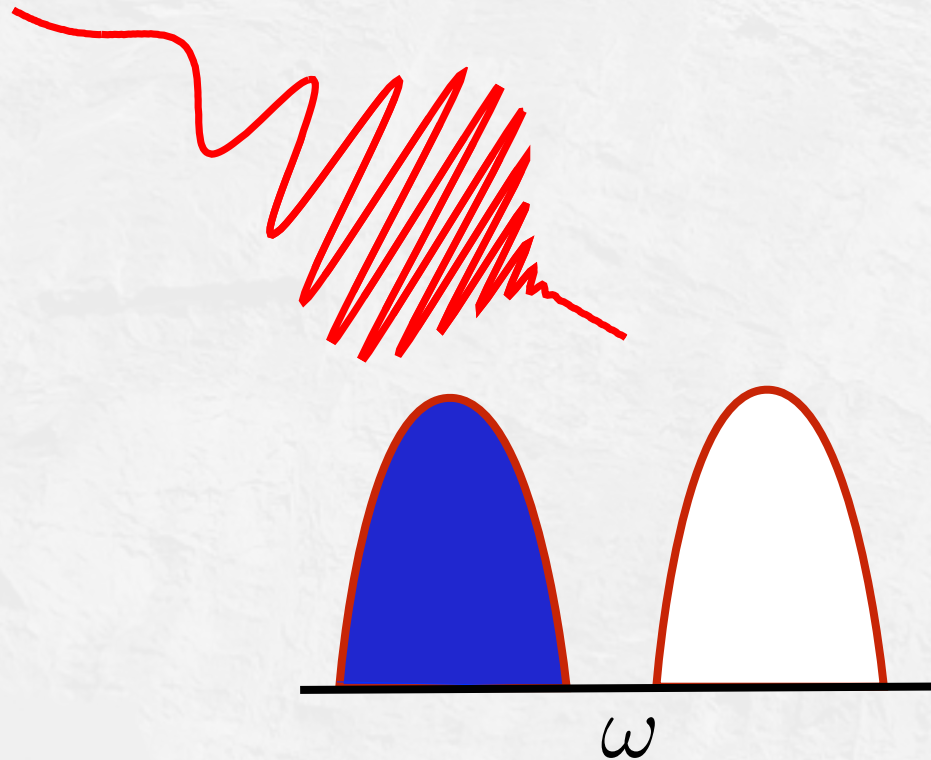
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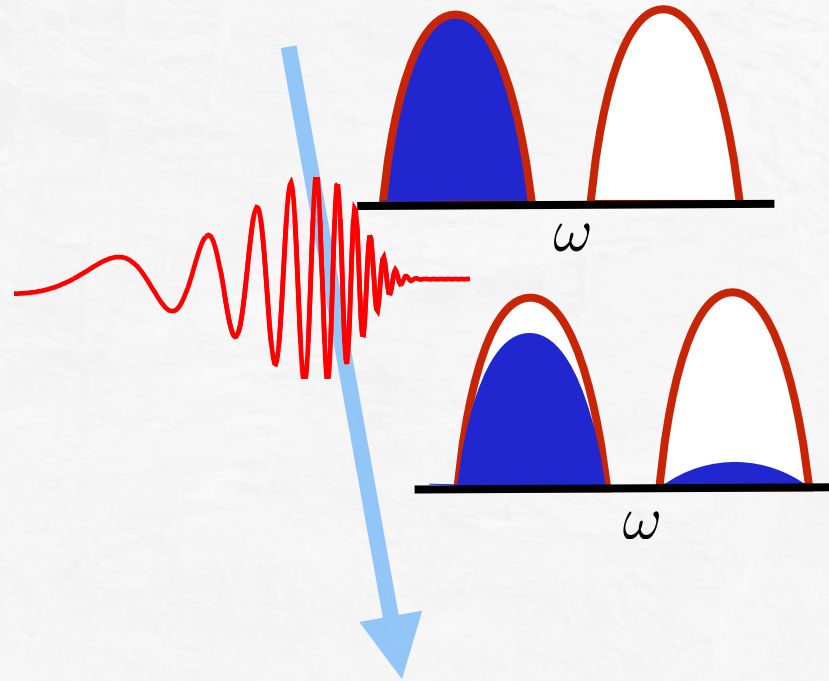
Quantum impurity solver

$$\mathcal{S} = \int dt dt' c(t)^* \Delta(t, t') c(t) + \mathcal{S}_{int}$$

Cold photo-doped states



Lifetime of photo-doped states

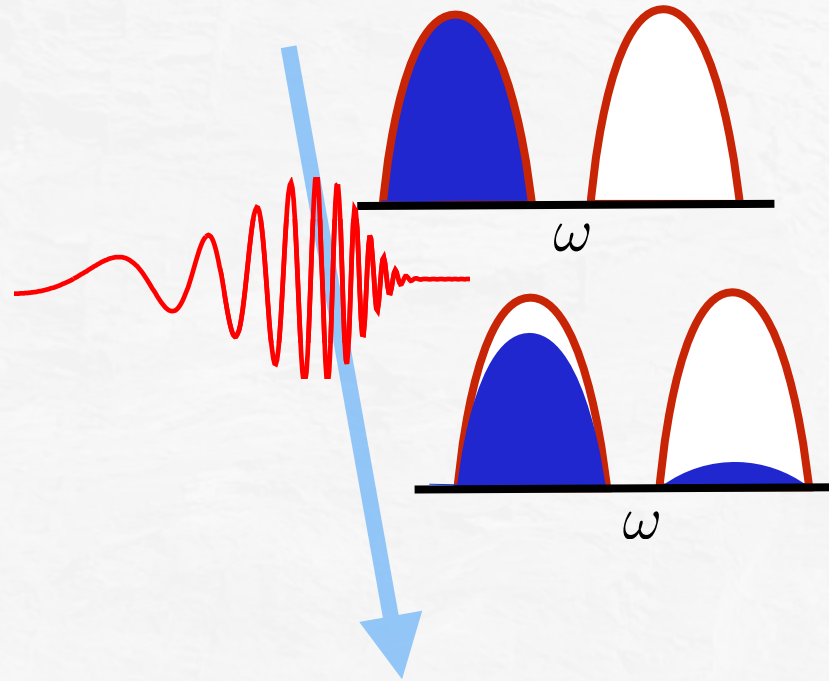


Thermalization of the double occupancy
after (pretty much any kind of) excitation:

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

T_{eff} $\uparrow$ determined by total energy

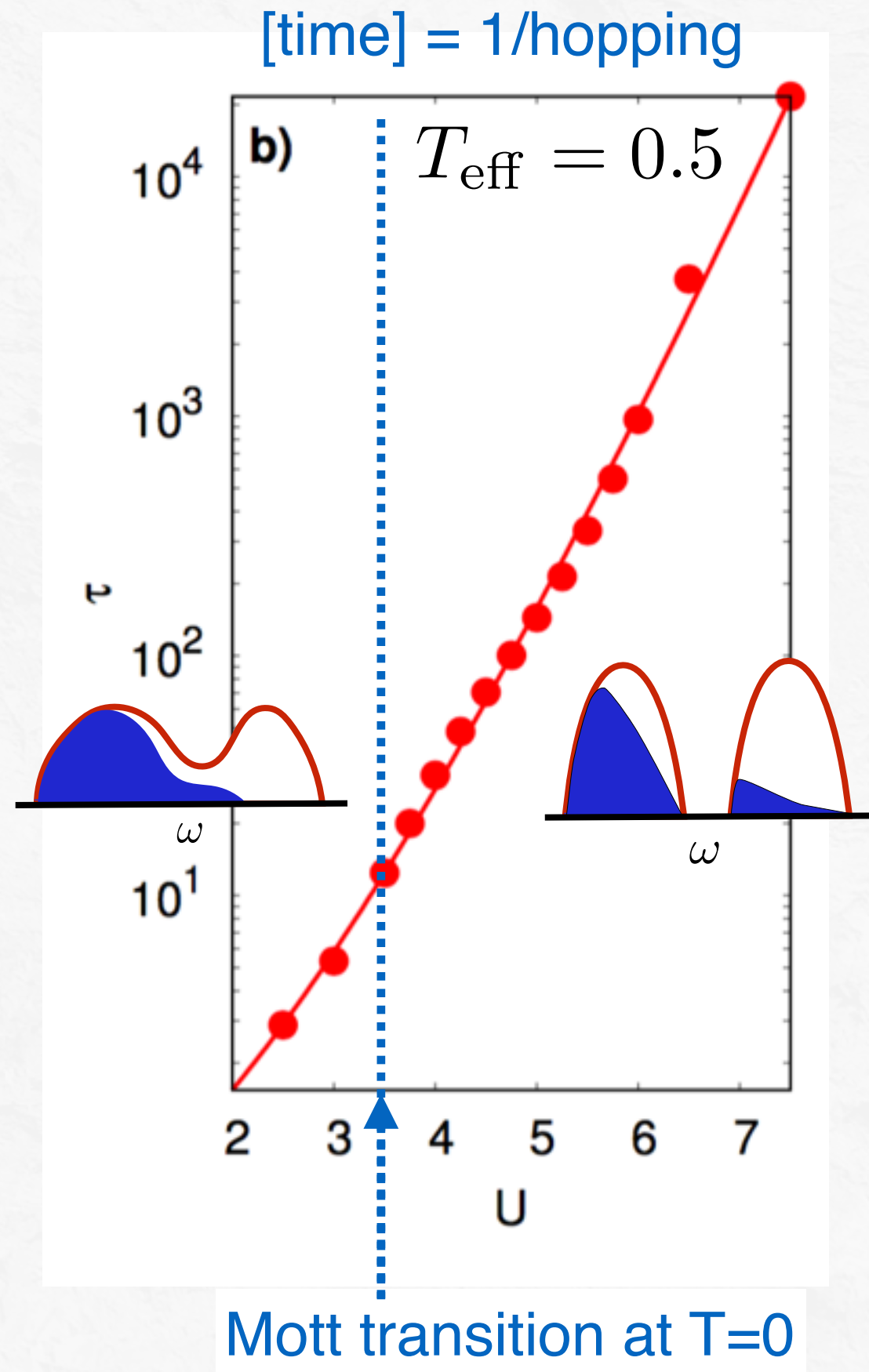
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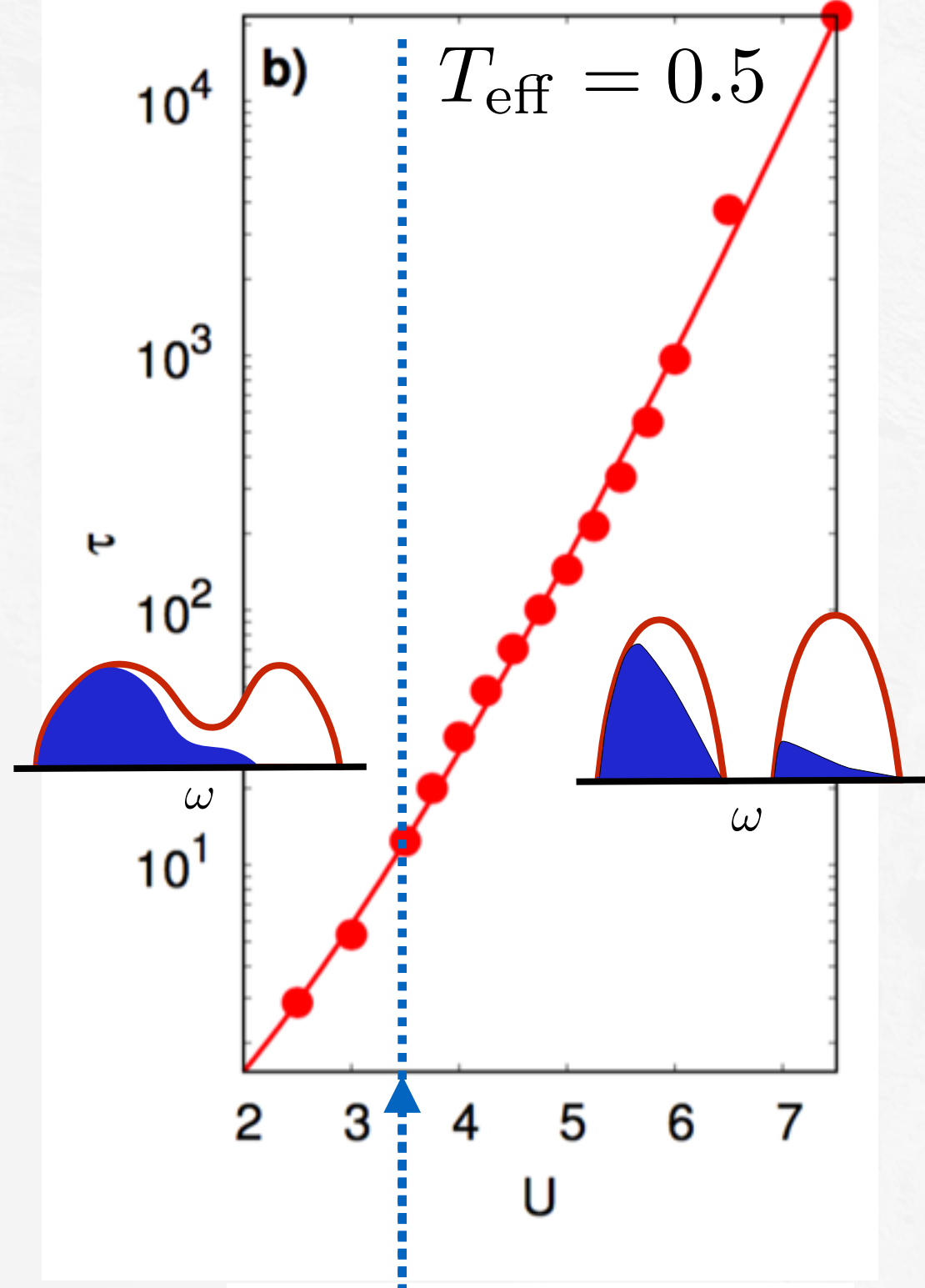
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Lifetime of photo-doped states

[time] = 1/hopping

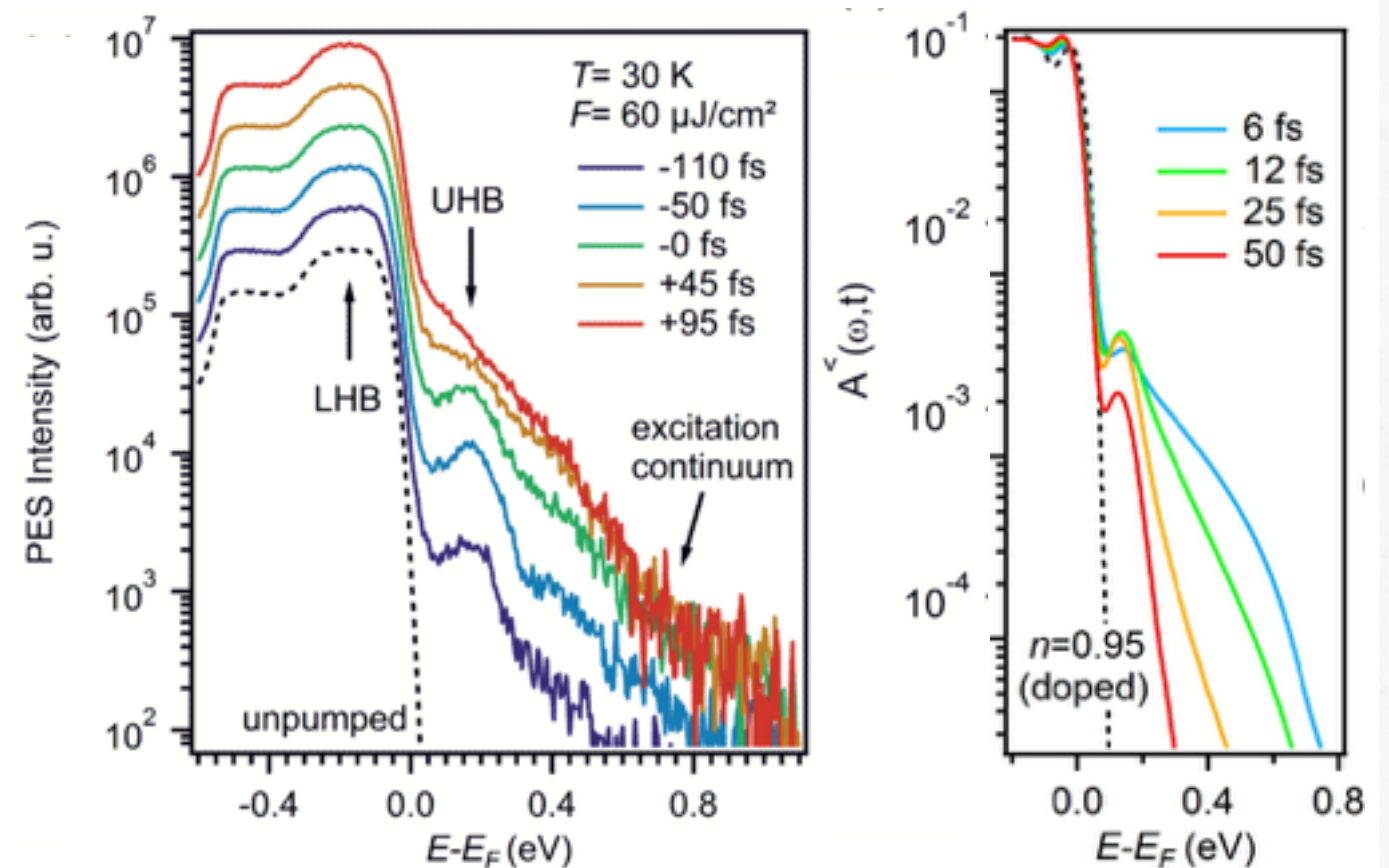


Mott transition at $T=0$

- ps ... ns lifetime of carriers in Mott.



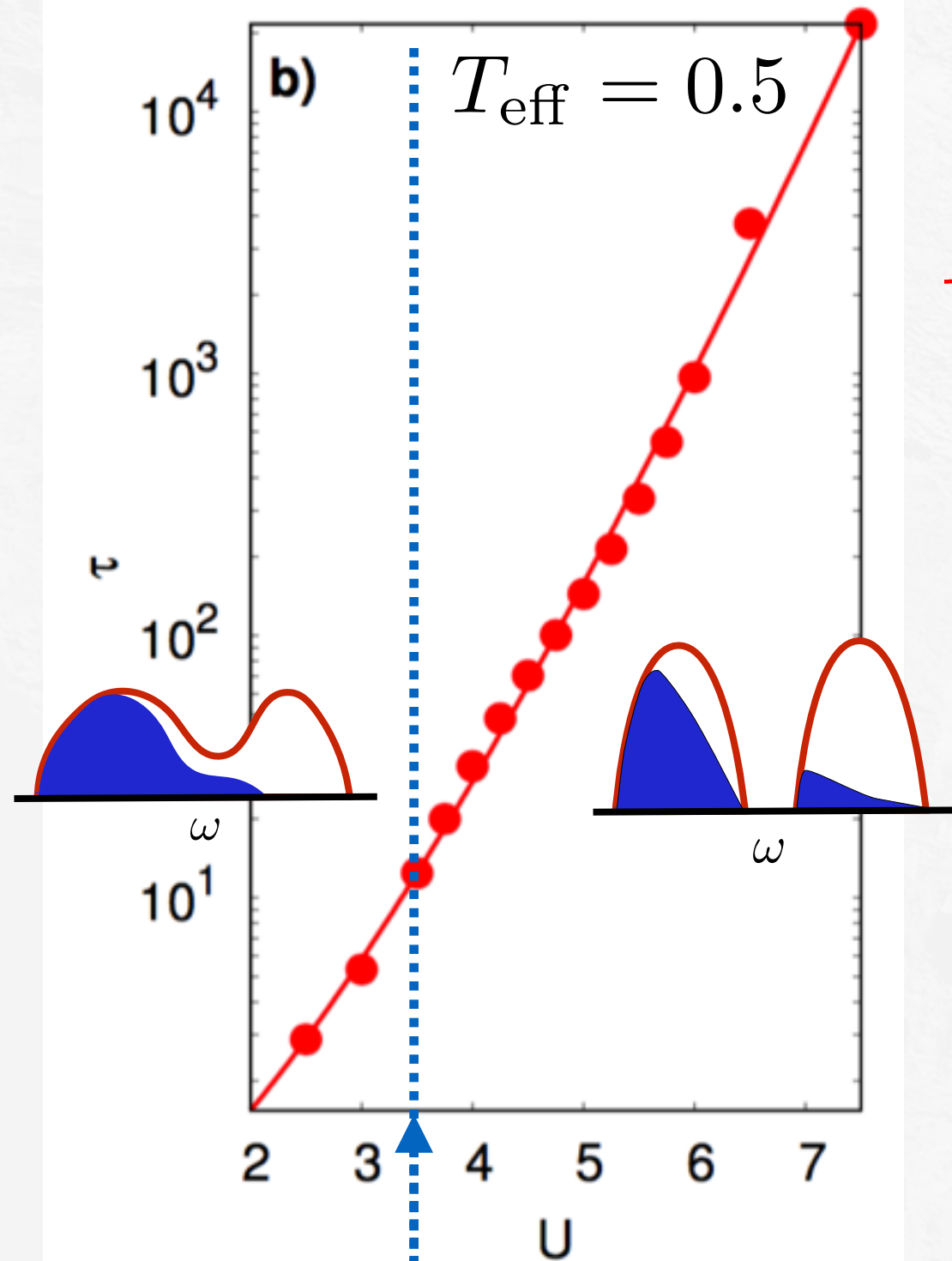
- Fast Thermalization in small gap Mott insulators: 1T-TaS₂:



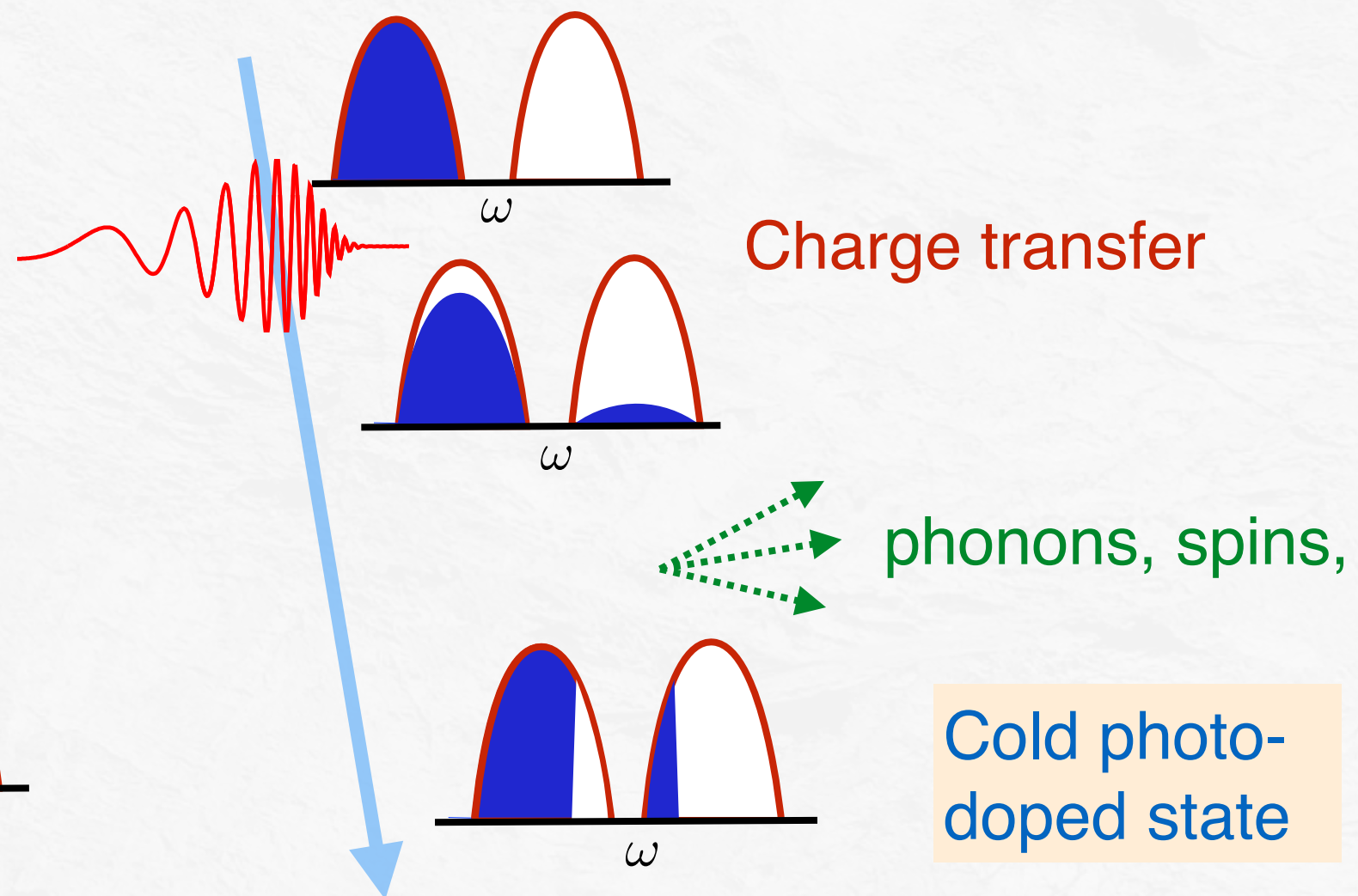
Ligges et al., PRL (2018)

Lifetime of photo-doped states

[time] = 1/hopping



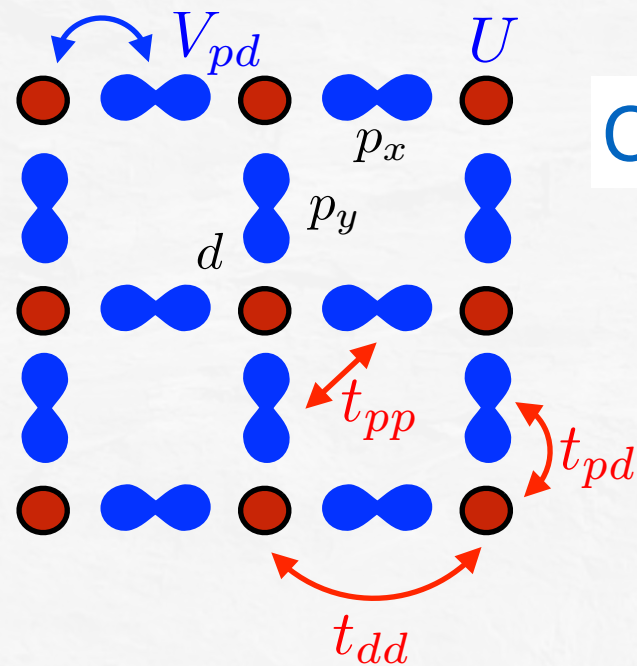
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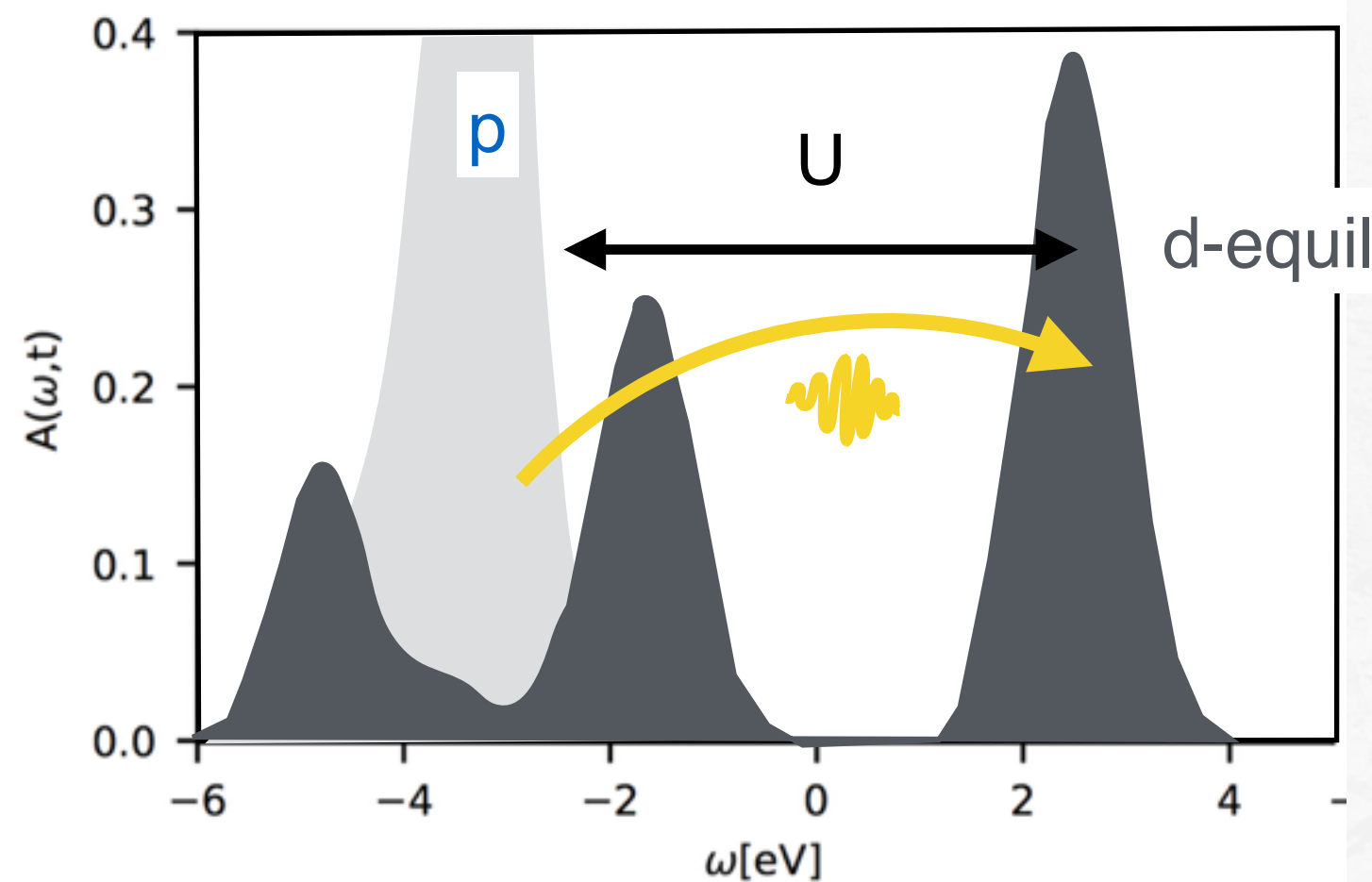
Few % charge transfer can substantially modify electronic structure:



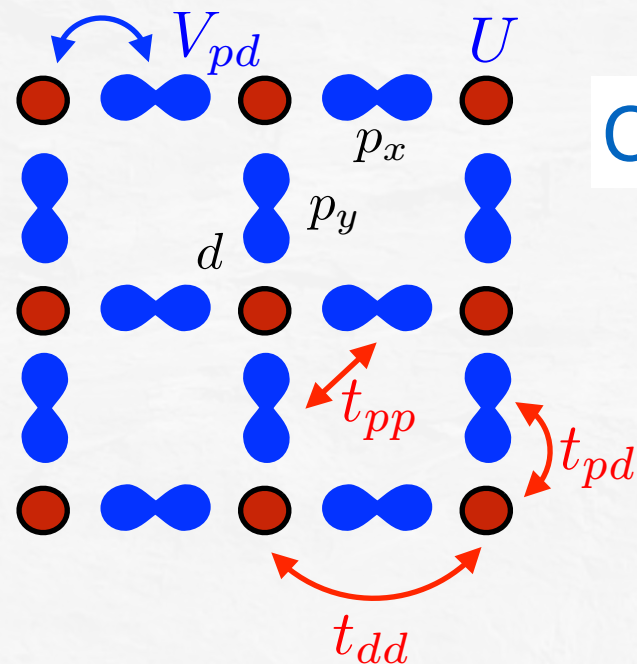
Few % charge transfer can substantially modify electronic structure:



Charge transfer insulator real-time GW+DMFT

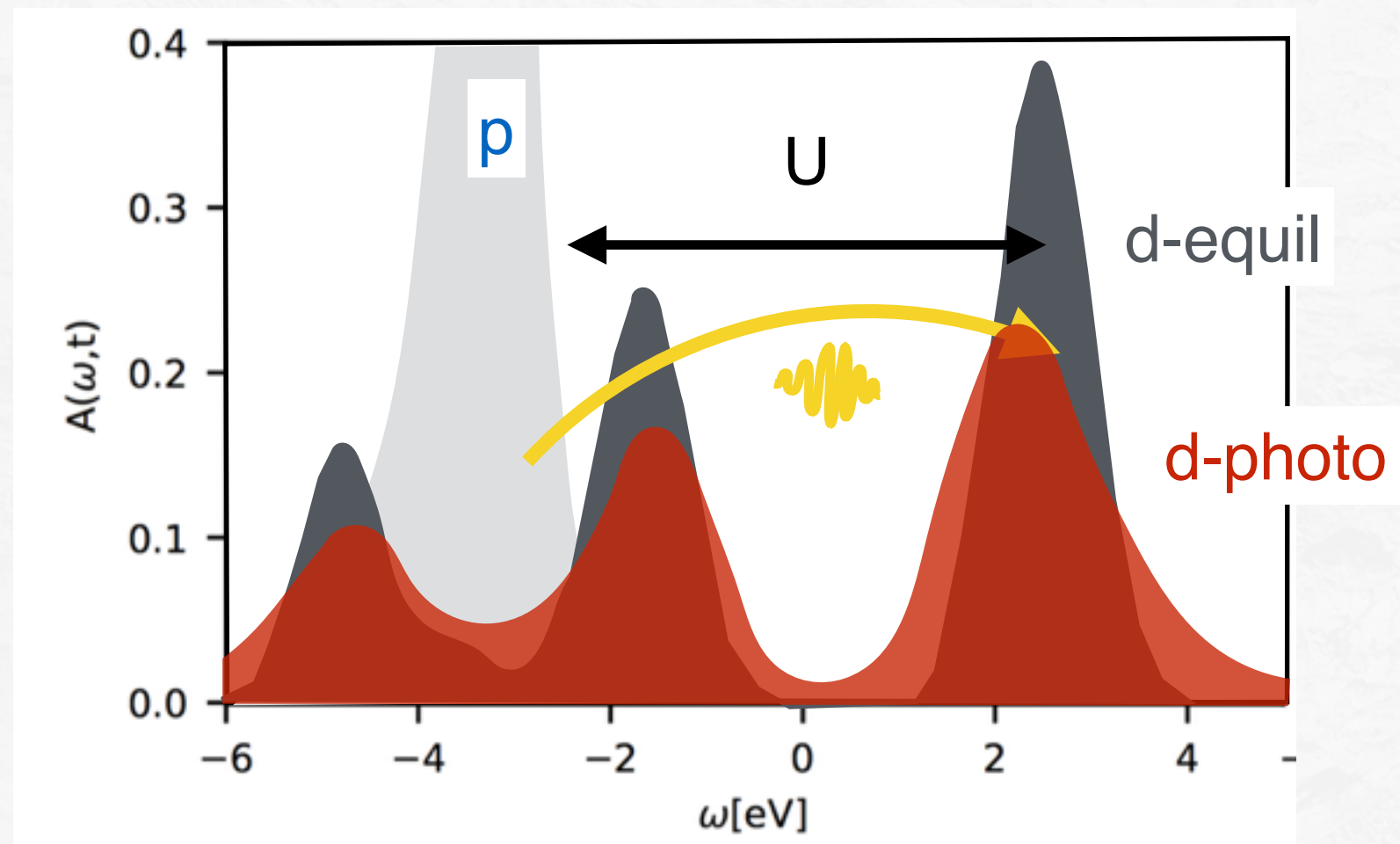


Few % charge transfer can substantially modify electronic structure:



Charge transfer insulator

real-time GW+DMFT



Shift (mean-field + dynamical screening
strong broadening (activation of scattering))

Band shifts: “time-dependent U ”, tdDFT, Tancogne-Dejean, Sentef, and Rubio (2018).

Dynamics of inter-twined orders? “Hidden states”

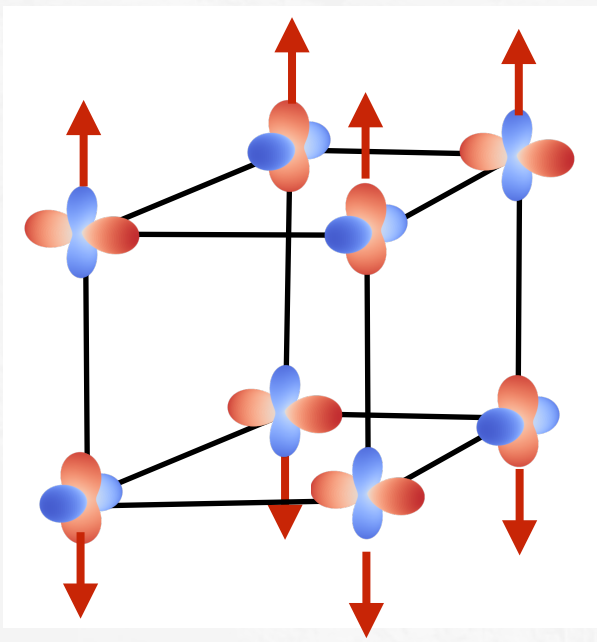




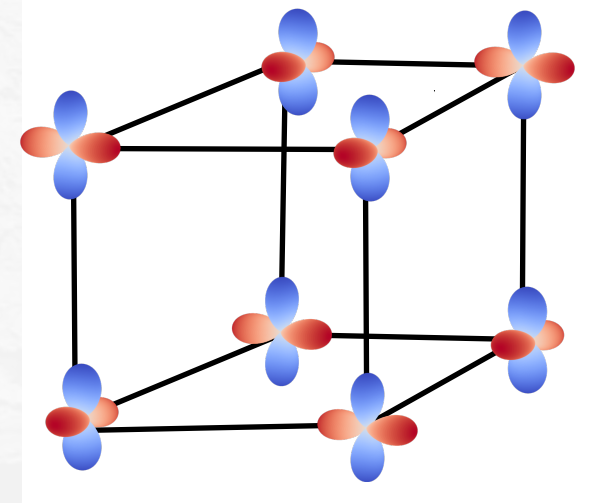
Dynamics of inter-twined orders? “Hidden states”

Mott insulator with degenerate Orbitals:

Spin & orbital order: Kugel & Khomskii 1982

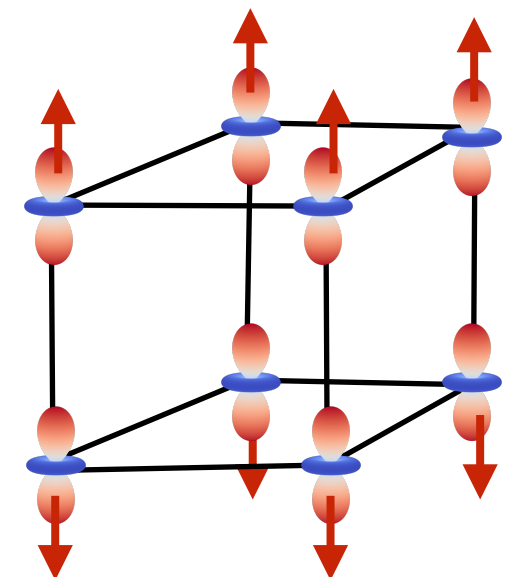


Heating: Melting orders by
population of (orbital) spin waves:

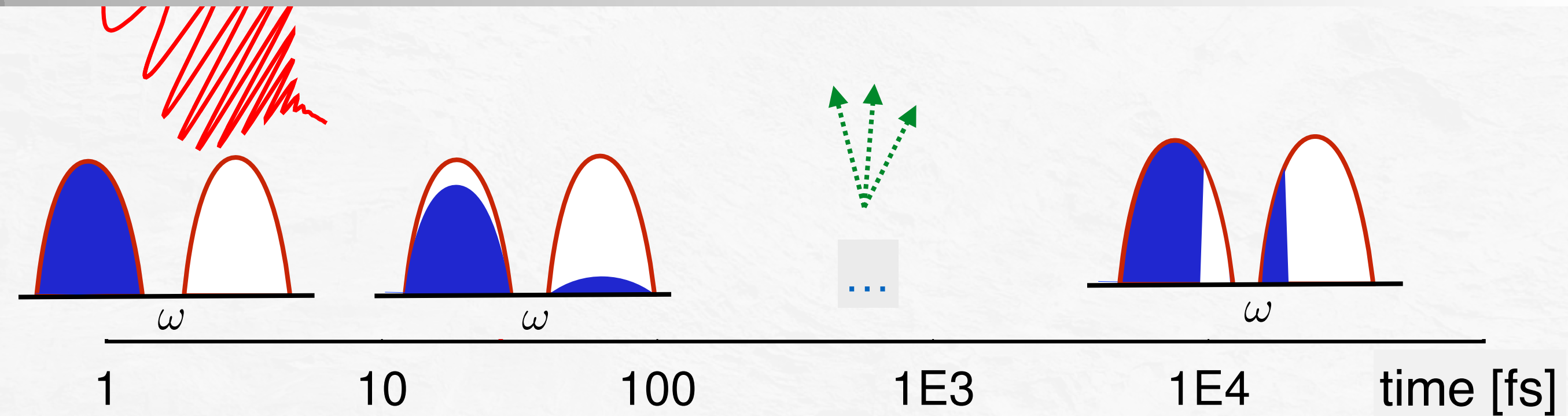


suppressed OO in
presence of AFM

quenching orders by motion
of photo-doped carriers

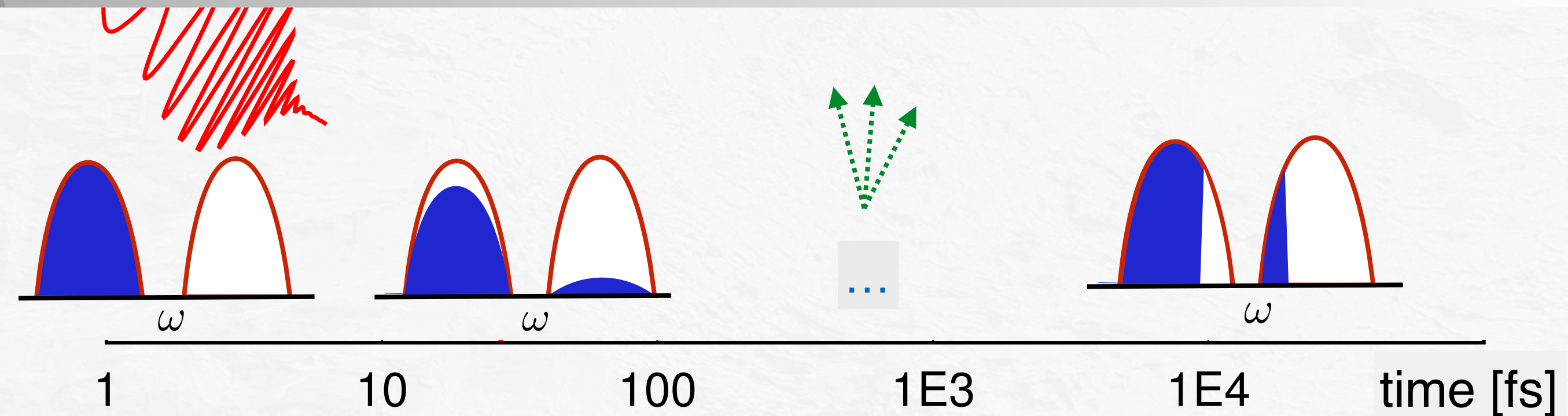


Steady state representation of photo-doped states



Simulate the time evolution?
(usually numerically costly)

Steady state representation of photo-doped states



Simulate the time evolution?
(usually numerically costly)

$$G(t, t') \approx G(t - t')$$

Non-equilibrium steady state
determined by effective
Temperature and “photo-doping”?

Steady state non-equilibrium Green's functions

Non-equilibrium steady state:

time-translational invariance
no fluctuation dissipation theorem

$$-i\langle c(t)c(t')^\dagger \rangle = G^>(t, t') \approx G^>(t - t') \leftrightarrow G^>(\omega) = -iA(\omega)(1 - F(\omega))$$

$$i\langle c(t')^\dagger c(t) \rangle = G^<(t, t') \approx G^<(t - t') \leftrightarrow G^<(\omega) = iA(\omega)F(\omega)$$

Keldysh formalism:

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



Interaction self-energy
External bath

NESS - DMFT (strong coupling impurity solver)

Steady state non-equilibrium Green's functions

Non-equilibrium steady state:

time-translational invariance
no fluctuation dissipation theorem

dissipation + continuous driving

$$-i\langle c(t)c(t')^\dagger \rangle = G^>(t, t') \approx G^>(t - t') \leftrightarrow G^>(\omega) = -iA(\omega)(1 - F(\omega))$$

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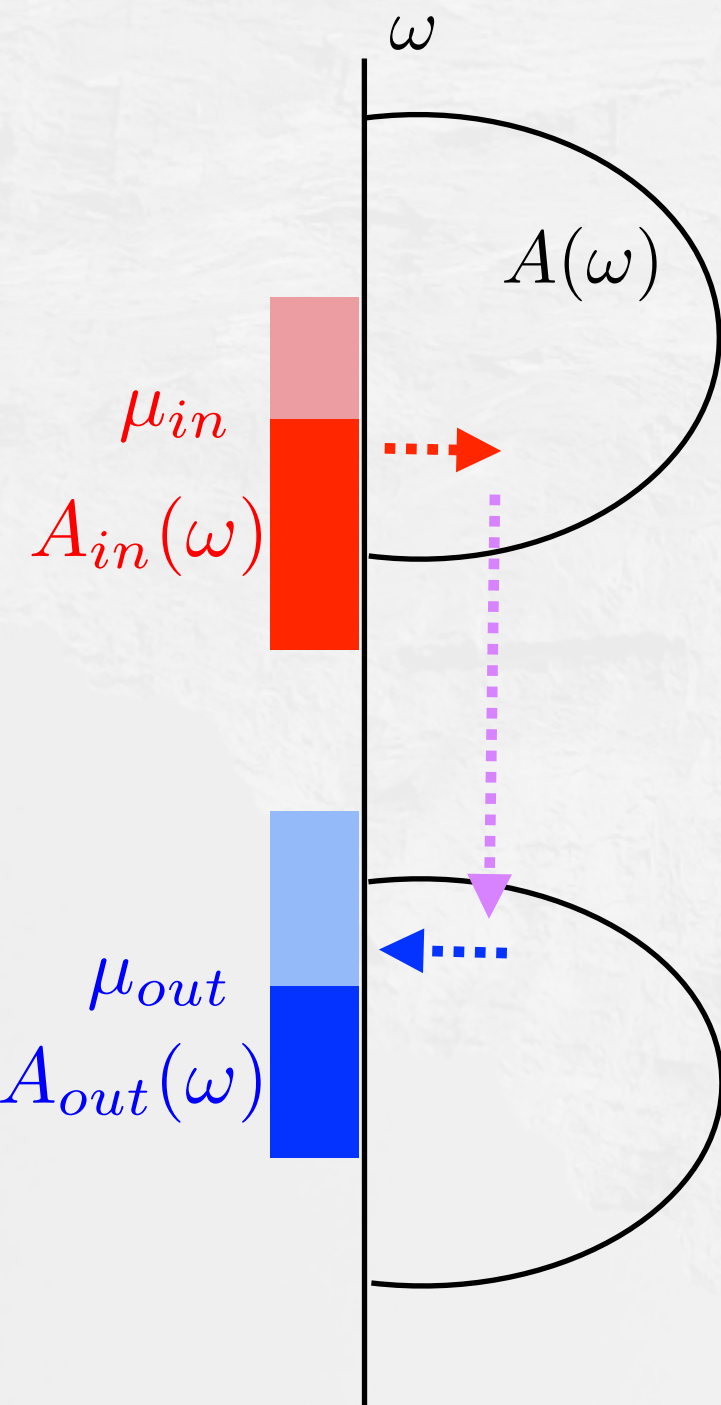
Interaction self-energy
External bath

NESS - DMFT (strong coupling impurity solver)

Photo-doping by bath

Non-equilibrium steady state:

Two bath coupled to every site of the lattice:

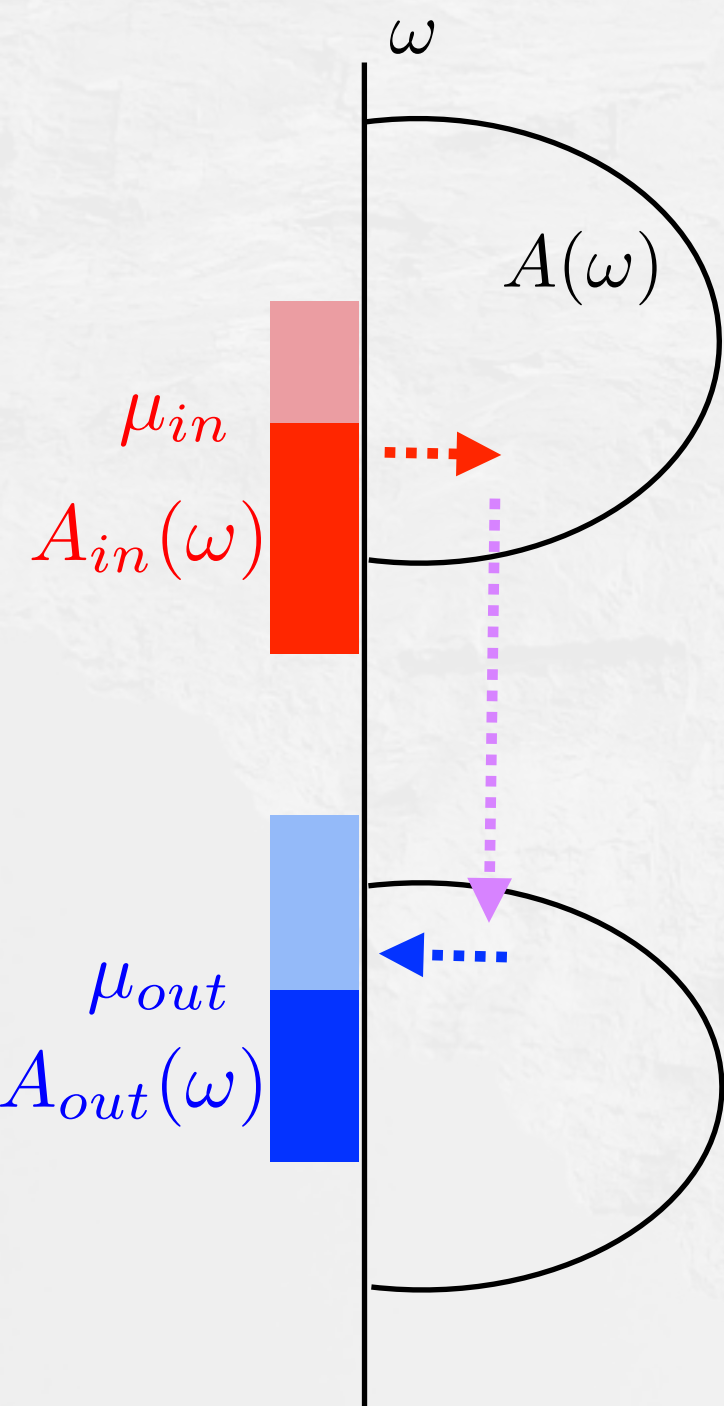


$$H = H_{System} + \sum_i \sum_{\nu=in,out} \left[\int d\omega A_\nu(\omega) \left(c_i^\dagger \psi_{i,\nu}(\omega) + h.c. \right) + \int d\omega (\omega - \mu_\nu) \psi_{i,\nu}(\omega)^\dagger \psi_{i,\nu}(\omega) \right]$$

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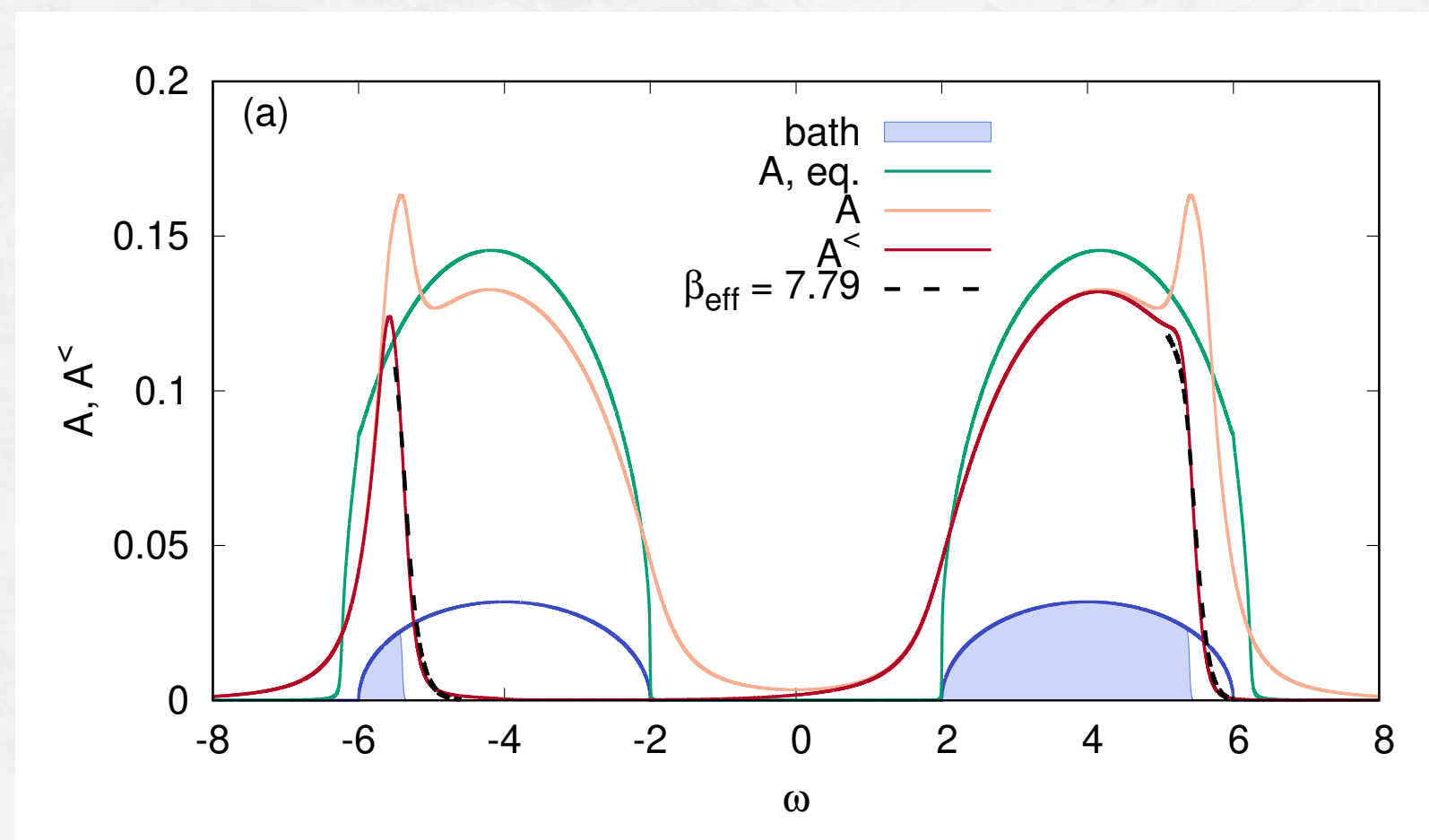
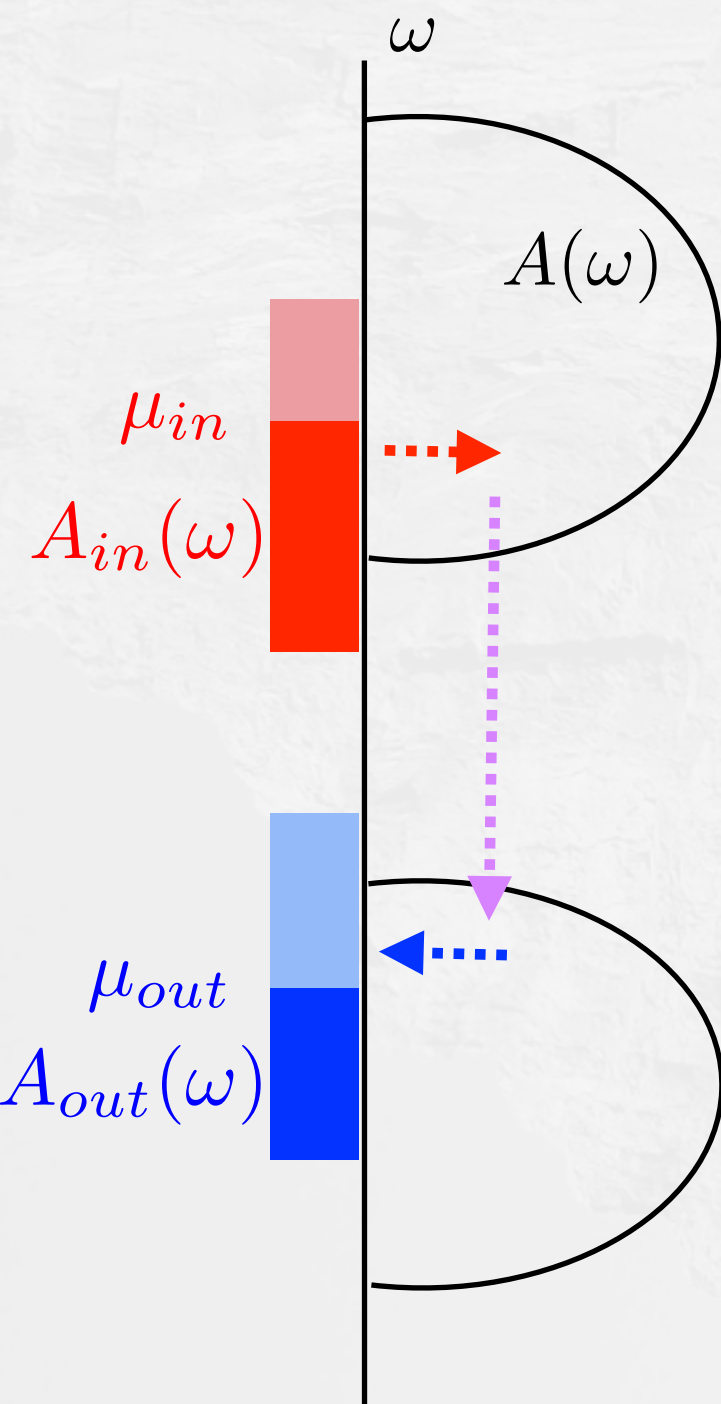


Photo-doping by bath

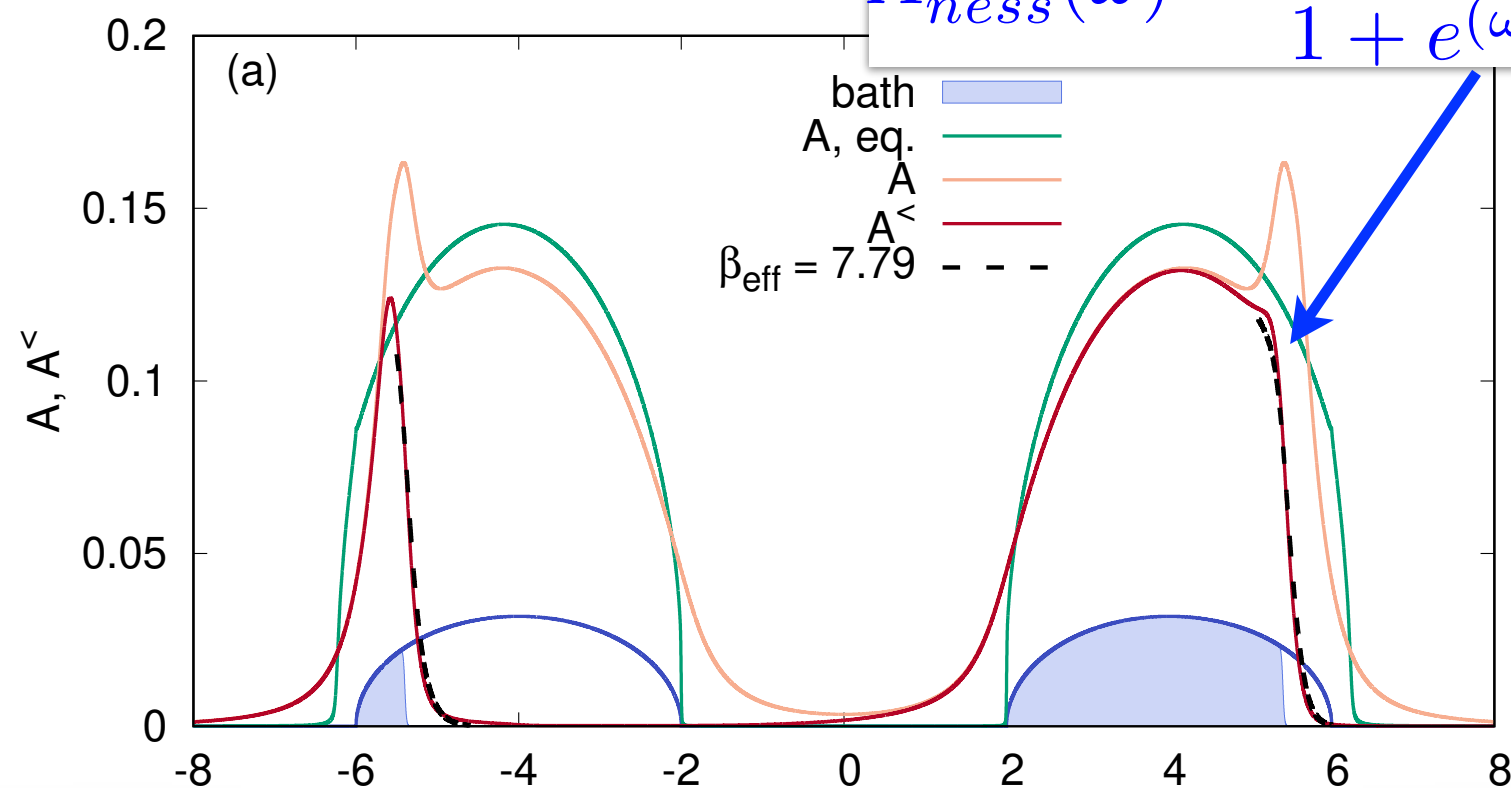
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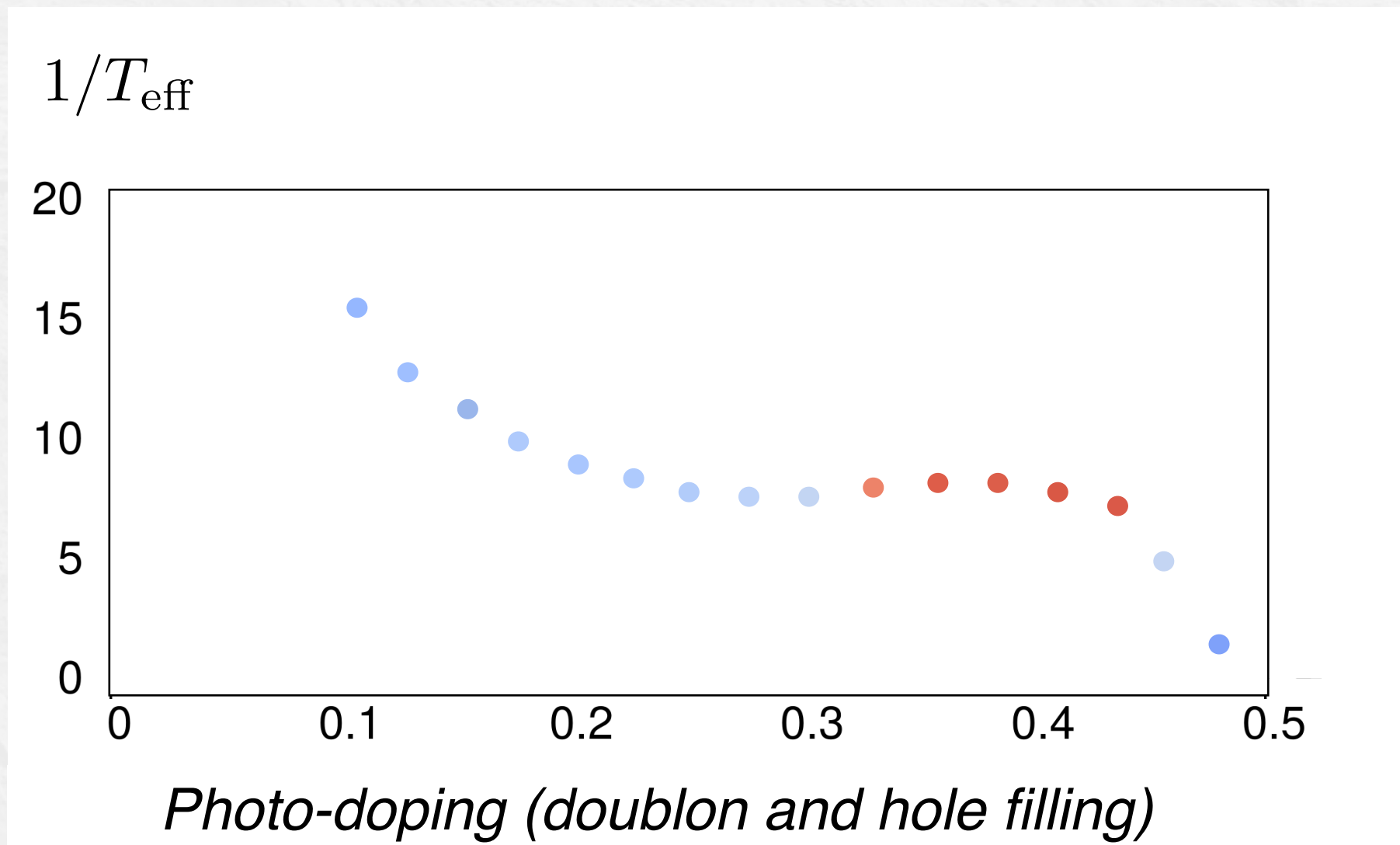
$$A_{ness}^{<}(\omega) = \frac{A_{ness}(\omega)}{1 + e^{(\omega - \mu)/T_{eff}}}$$



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

Phase-diagram of photodoped system

Scan for different μ_{bath} :



Instabilities?

AFM, CDW, SC, ...

$$H = H_{\text{System+Bath}} + h_{\eta} \sum_j \eta_j^x \quad \chi_{\eta} = \left. \frac{\langle \eta_j^x \rangle_{\text{ness}}}{h_{\eta}} \right|_{h \rightarrow 0}$$

η -pairing superconductivity

Hubbard model on a bi-partite lattice

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

SU(2)_s symmetry

$$\begin{aligned} S_j^+ &= c_{j\uparrow}^\dagger c_{j\downarrow} \\ S_j^- &= c_{j\downarrow}^\dagger c_{j\uparrow} \\ S_j^z &= \frac{1}{2}(n_{j\uparrow} - n_{j\downarrow}) \end{aligned} \quad \left(\begin{array}{c} | \uparrow \rangle \\ | \downarrow \rangle \end{array} \right)$$

Antiferromagnetism

η -pairing superconductivity

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Antiferromagnetism

$SU(2)_c$ symmetry

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

$$\begin{aligned} \eta_j^+ &= (-1)^j c_{j\uparrow}^\dagger c_{j\downarrow}^\dagger \\ \eta_j^- &= (-1)^j c_{j\downarrow} c_{j\uparrow} \\ \eta_j^z &= \frac{1}{2}(n_j - 1) \end{aligned} \quad \left(\begin{array}{c} |\uparrow\downarrow\rangle \\ |0\rangle \end{array} \right)$$

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

$\Rightarrow$ Staggered phase Superconductivity

(special excited state on bipartite lattice,
where η -pairing is conserved)

η -pairing superconductivity

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η -SC in attractive U: Rotating CDW SC, Dissipative state engineering

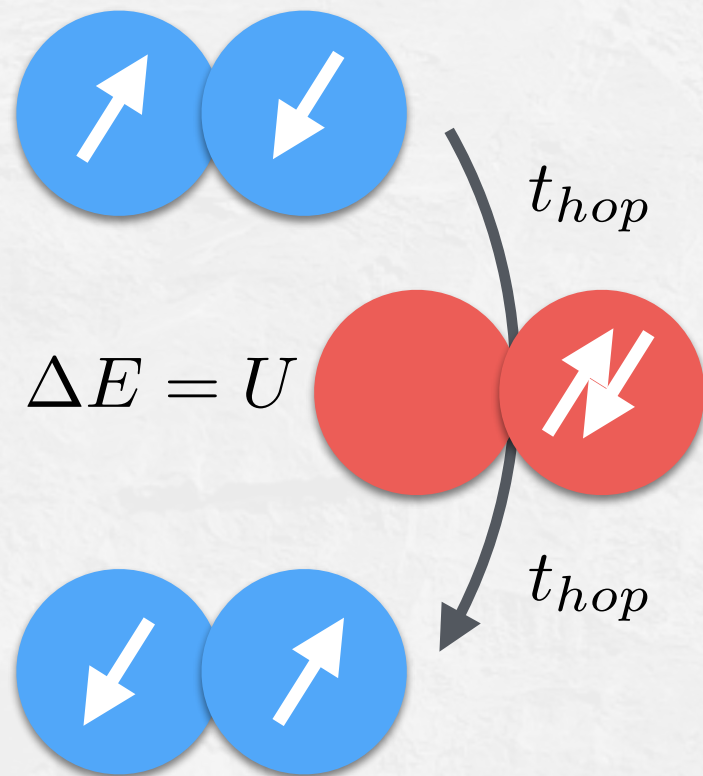
Kitamura and Aoki (2016), Sentef et al (2017) Bernier, Kollath (2016), Tindall et al (2019)

η -pairing superconductivity

Large U limit:

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

only singly occupied sites

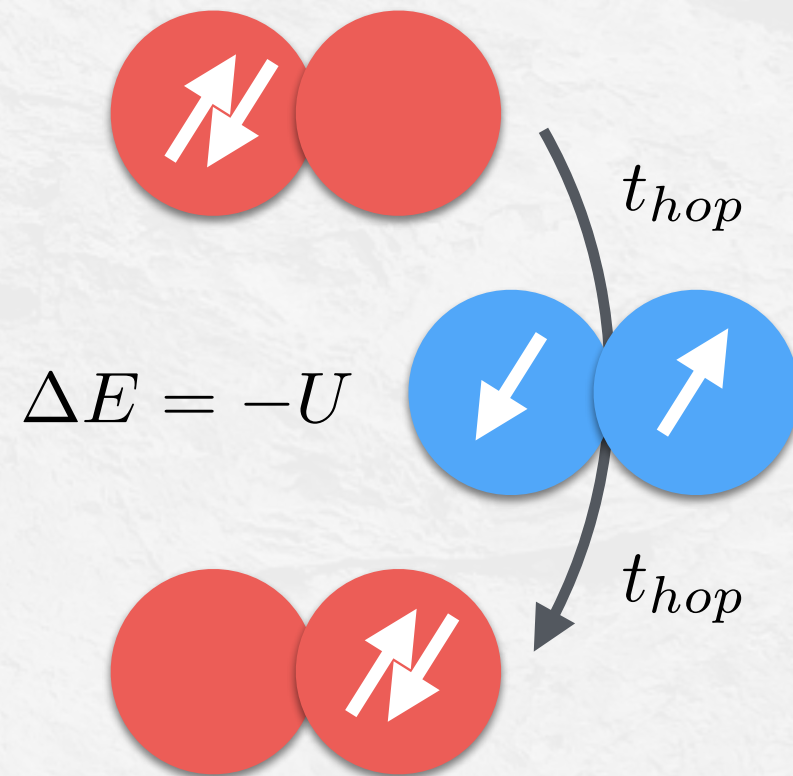


$$H^{\text{eff}} = \sum_{\langle ij \rangle} J_{\text{ex}} \mathbf{S}_i \cdot \mathbf{S}_j$$

Antiferromagnetism

$$T_N \sim t_{hop}^2 / U$$

only doubly occupied/empty sites



$$H^{\text{eff}} = - \sum_{\langle ij \rangle} J_{\text{ex}} \boldsymbol{\eta}_i \cdot \boldsymbol{\eta}_j$$

Needed nonequilibrium population of doublons

$$T_{\text{eff}} \sim t_{hop}^2 / U$$

Rosch et al, PRL (2008)

η -pairing superconductivity

*Only singly
occupied sites*

$$H^{\text{eff}} = \sum_{\langle ij \rangle} J_{\text{ex}} \mathbf{S}_i \cdot \mathbf{S}_j$$

inverse temperature

AFM

?

η

*Only doublons
& holes*

$$H^{\text{eff}} = - \sum_{\langle ij \rangle} J_{\text{ex}} \eta_i \cdot \eta_j$$

Rosch et al, PRL (2008)

0

0.1

0.2

0.3

0.4

0.5

Photo-doping (doublon and hole filling)

η -pairing superconductivity

Only singly
occupied sites

$$H^{\text{eff}} = \sum_{\langle ij \rangle} J_{\text{ex}} \mathbf{S}_i \cdot \mathbf{S}_j$$

inverse temperature

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Photo-doping (doublon and hole filling)

⇒ Use photo-excitation to “over-populate” doublons?

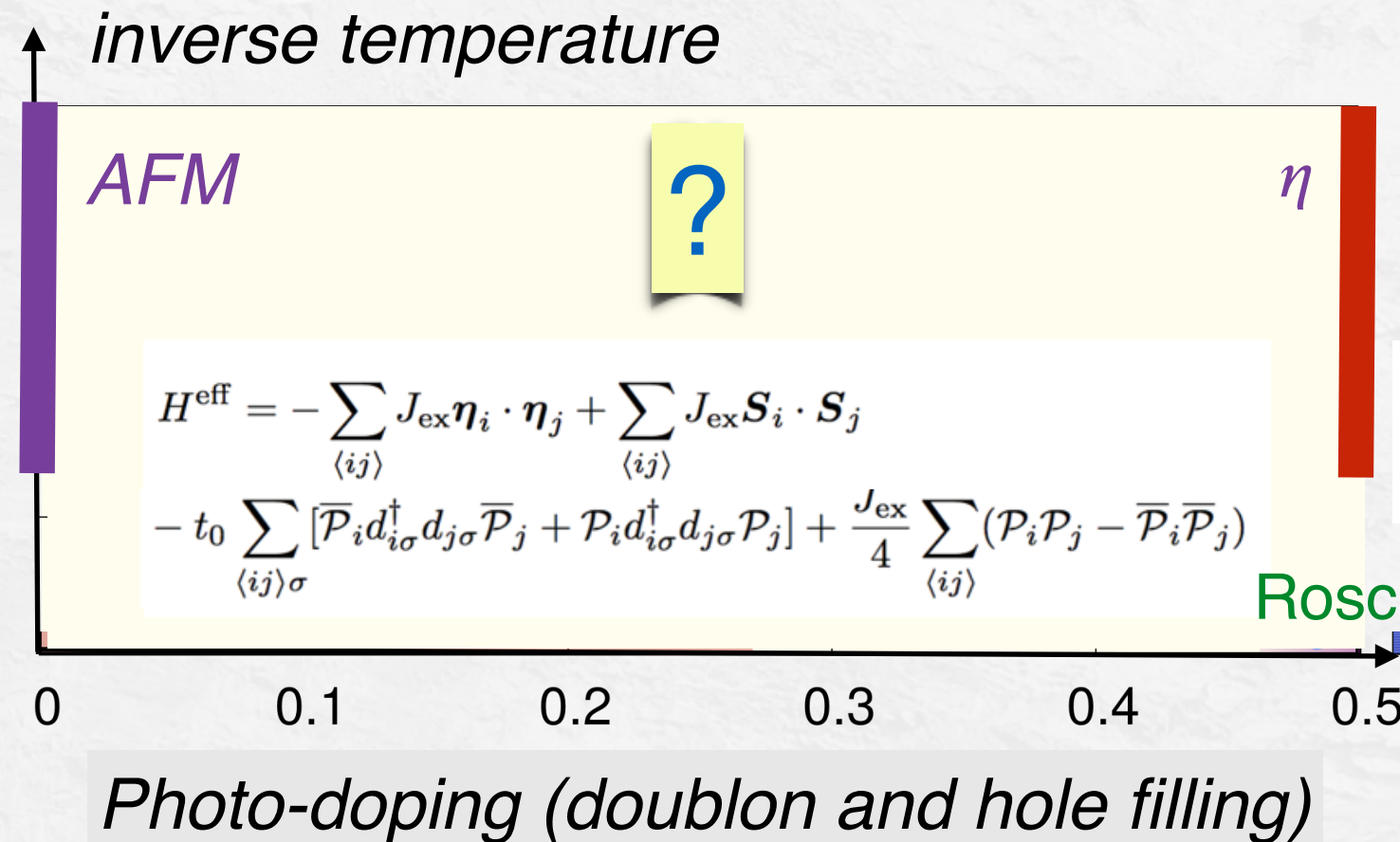
finite size simulations: Enhancing of η -correlations Kaneko et al, PRL 2018

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

η -pairing superconductivity

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Only doublons
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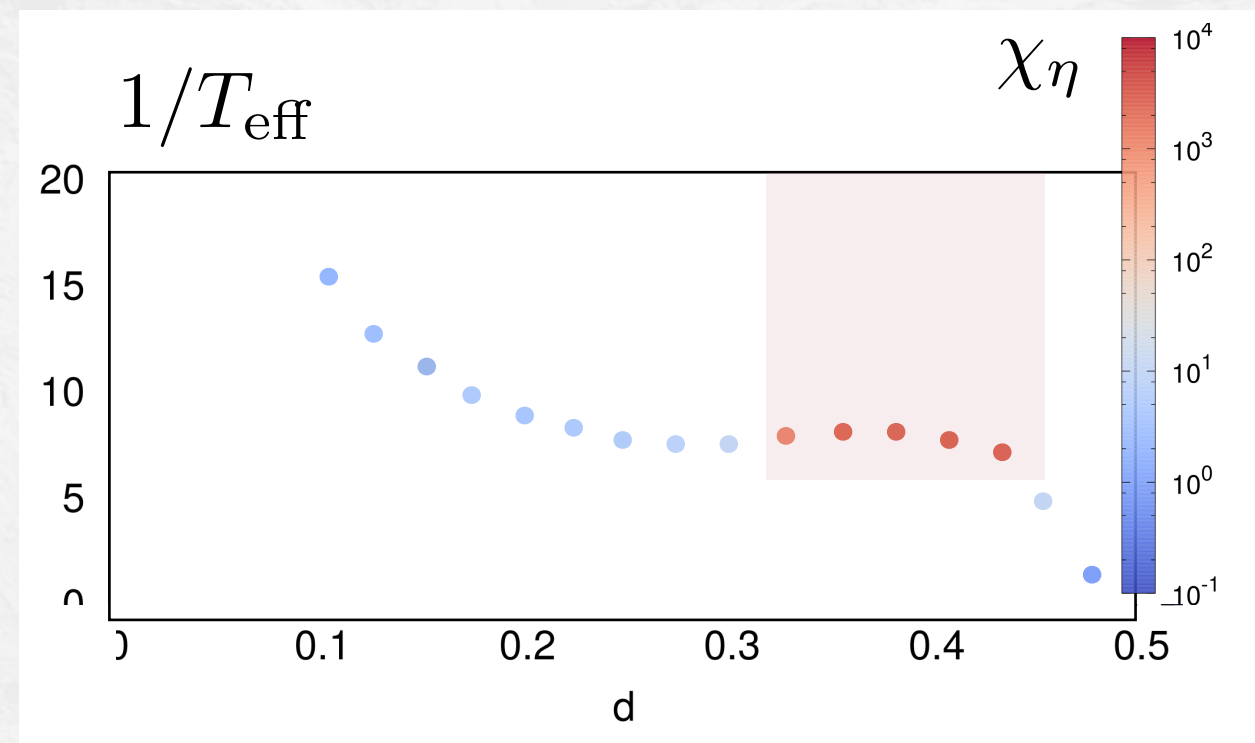
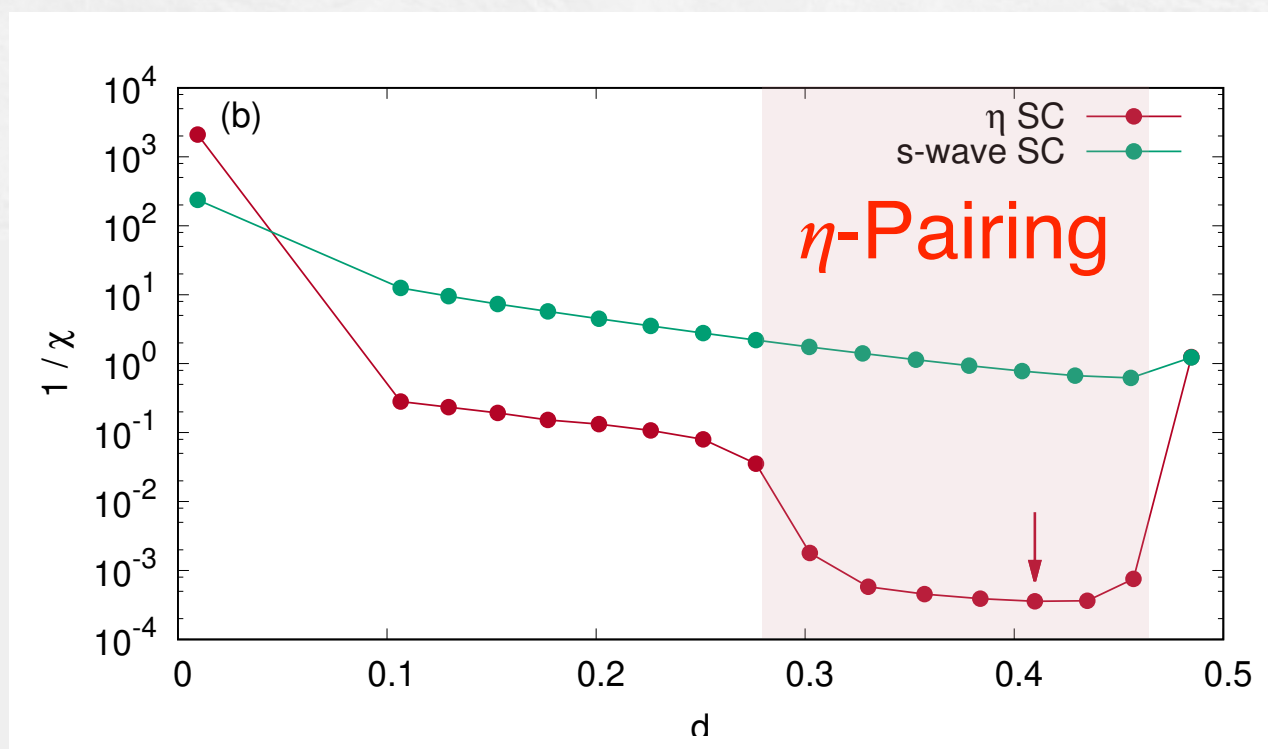
Phase-diagram of photodoped system

η -Pairing susceptibility in NESS:

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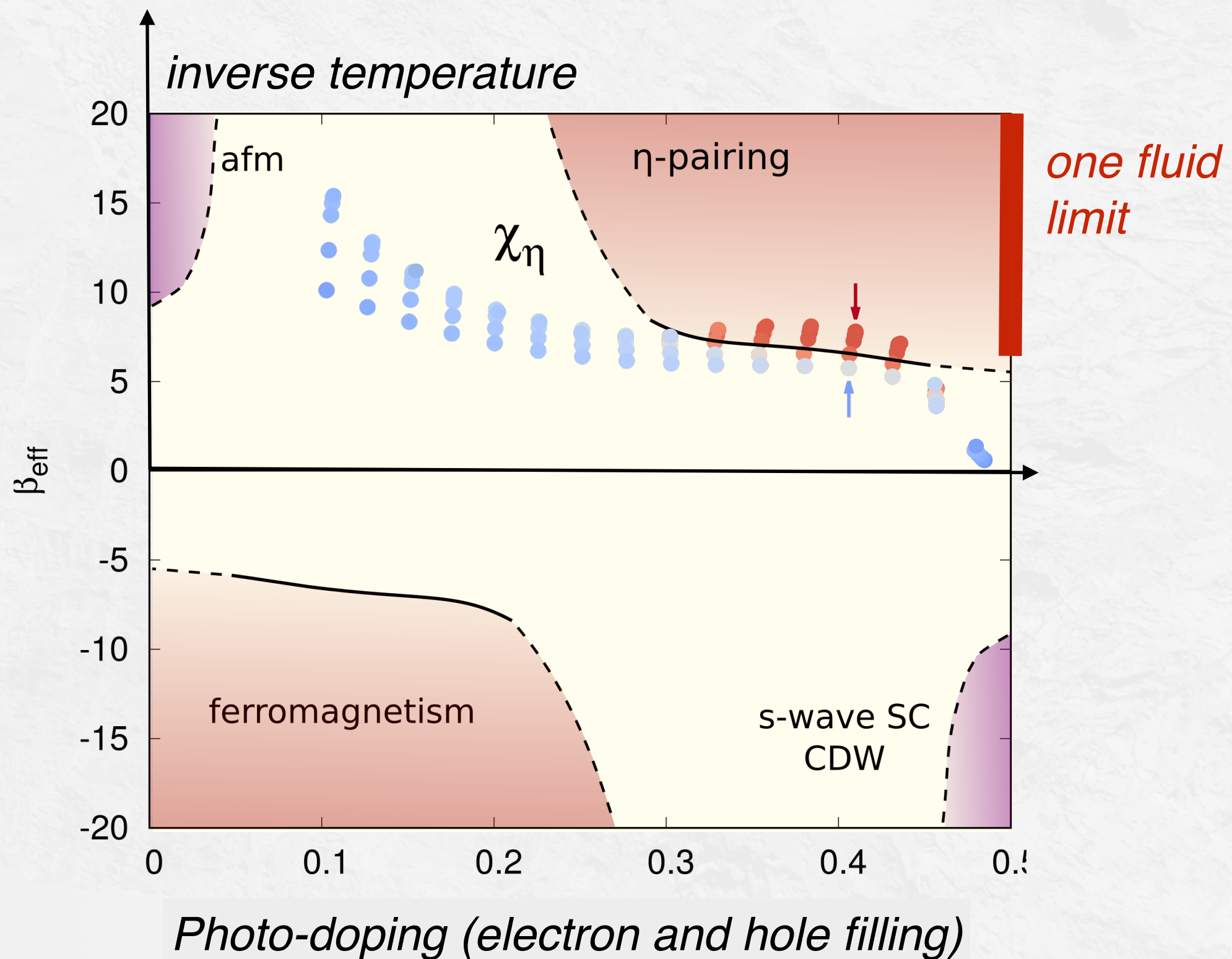
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Scan for different μ_{bath} :



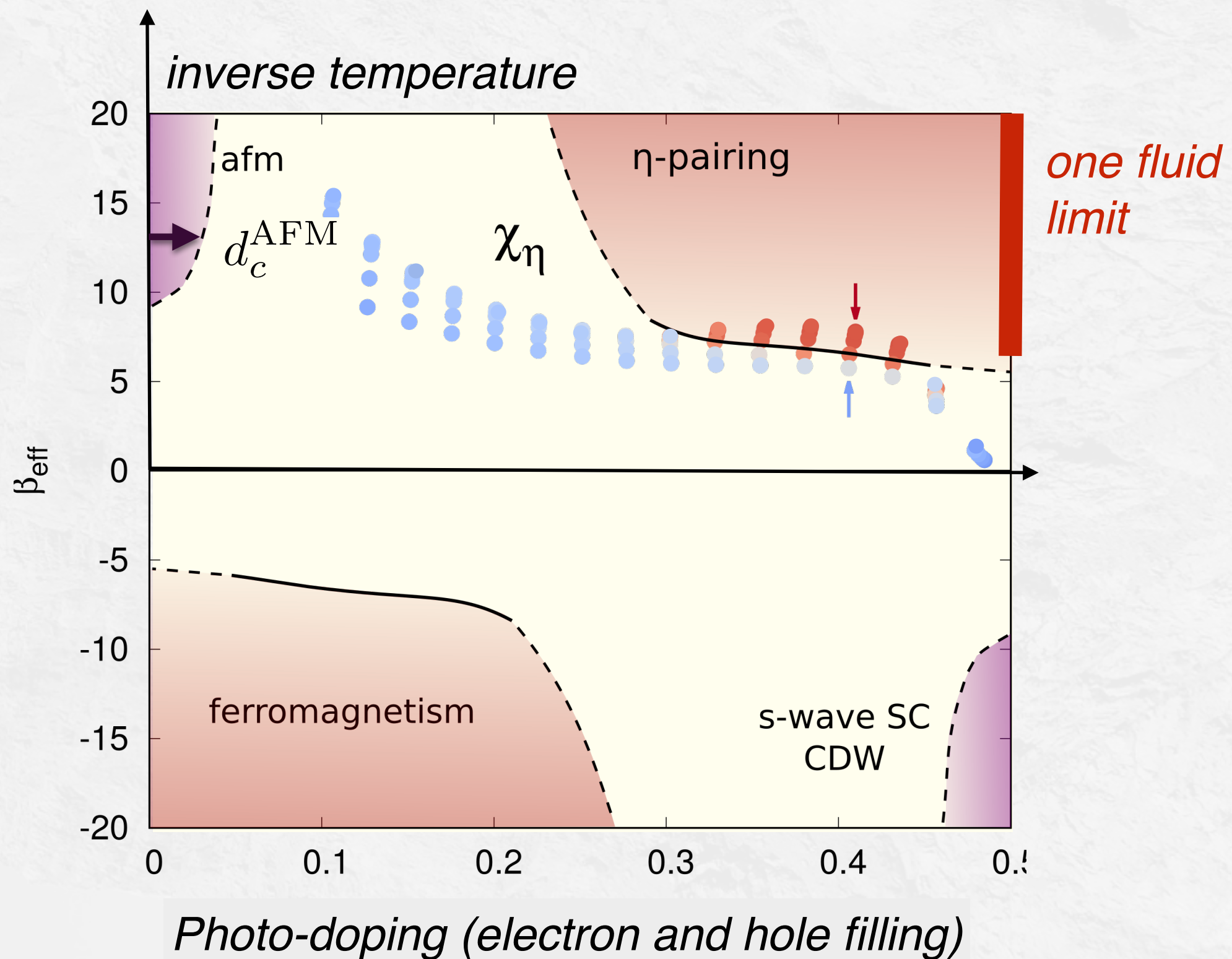
Phase-diagram of photo-doped system

Phase-diagram of photo-doped Hubbard model



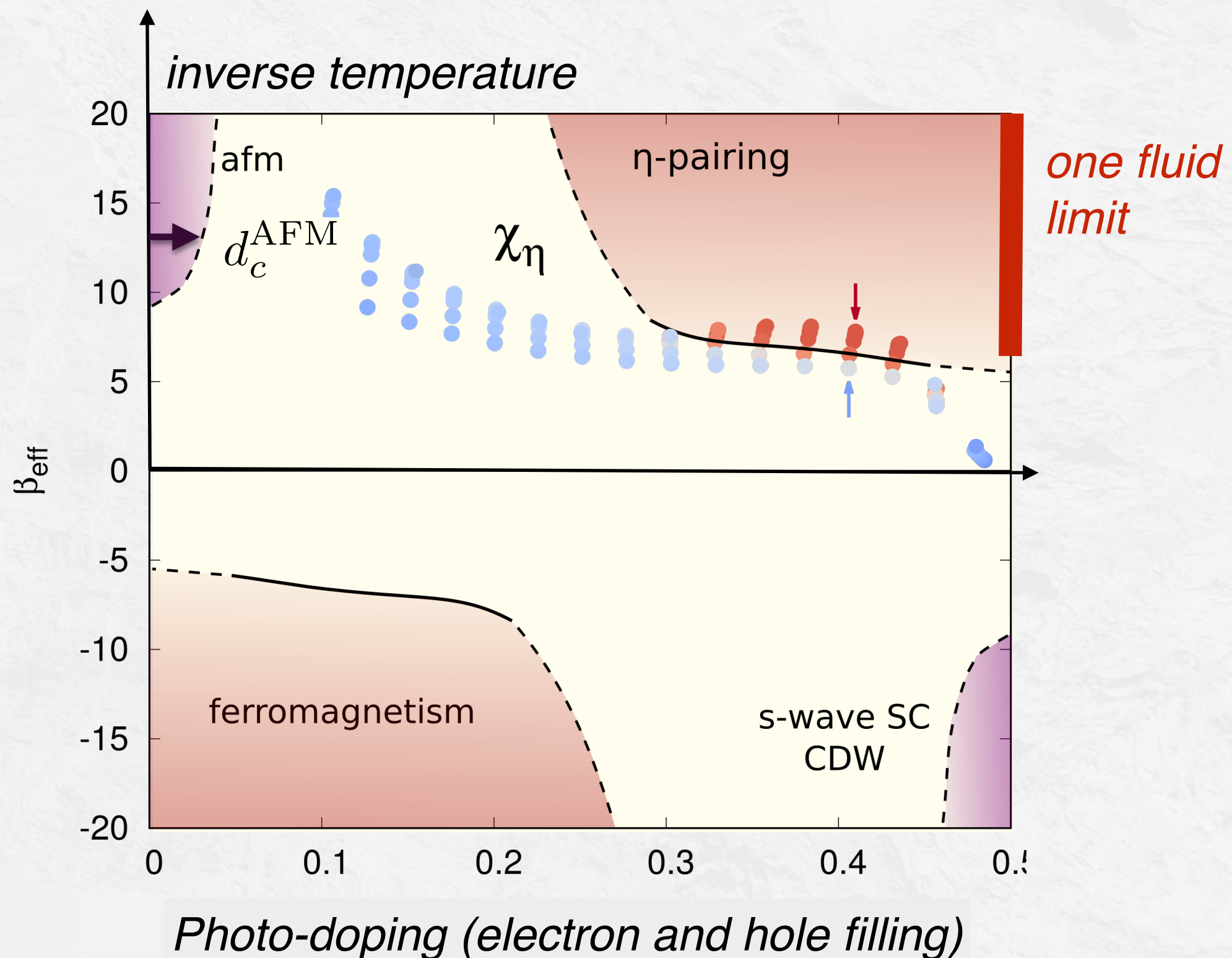
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Phase-diagram of photo-doped system

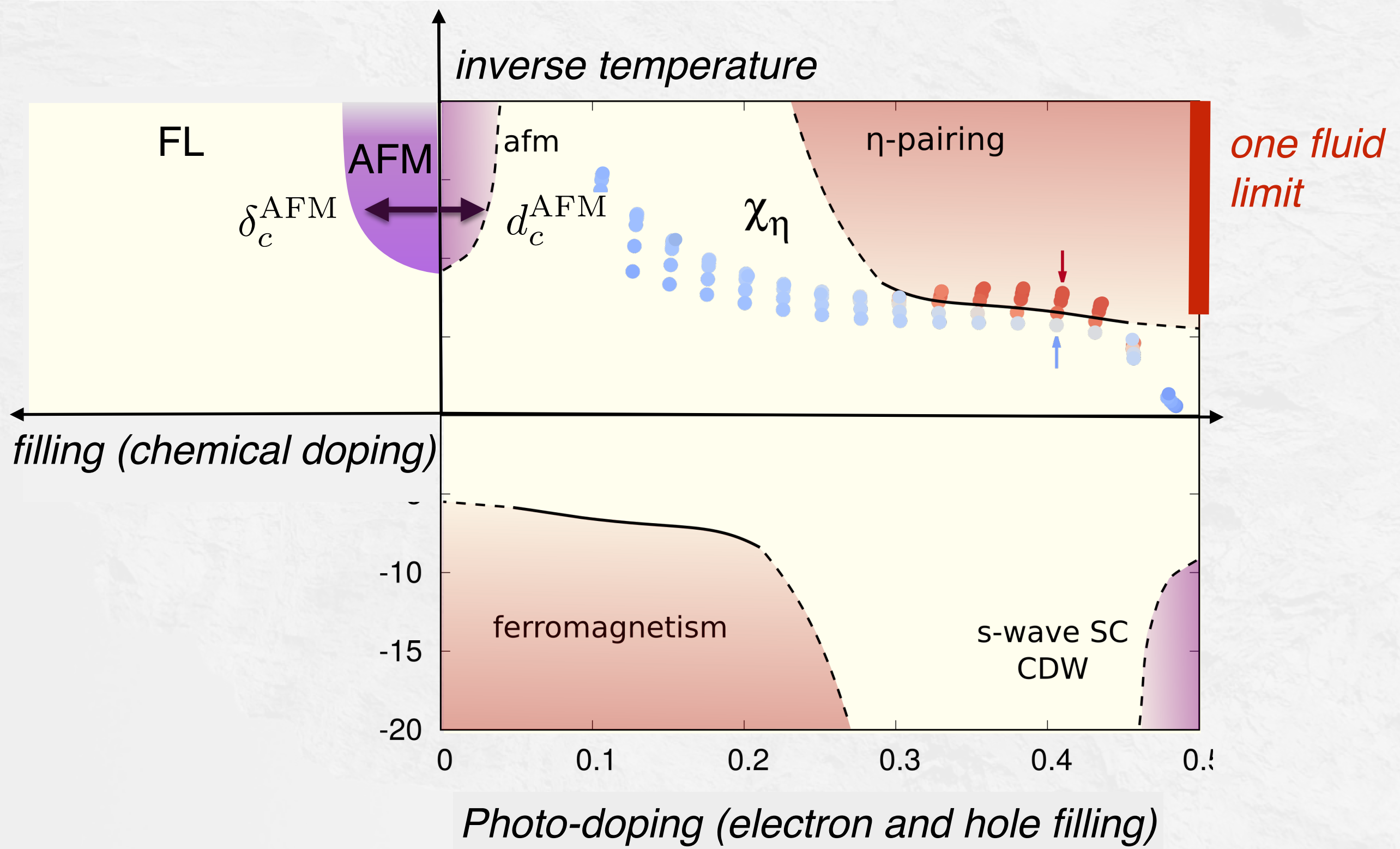
Phase-diagram of photo-doped Hubbard model



stable against NNN hopping

Phase-diagram of photo-doped system

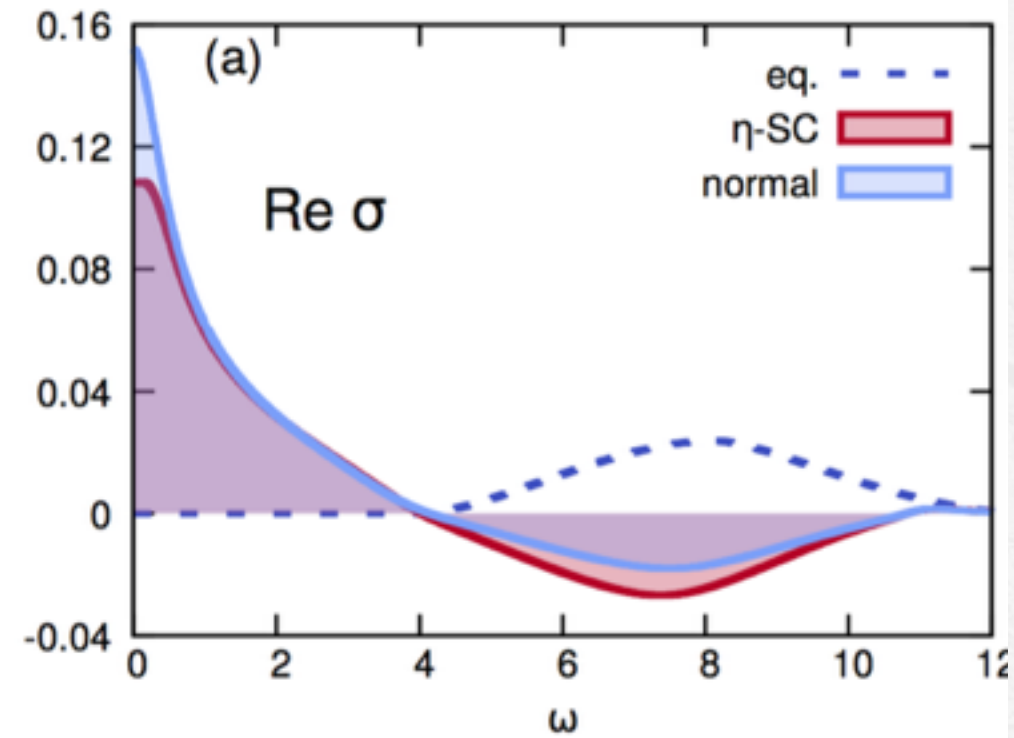
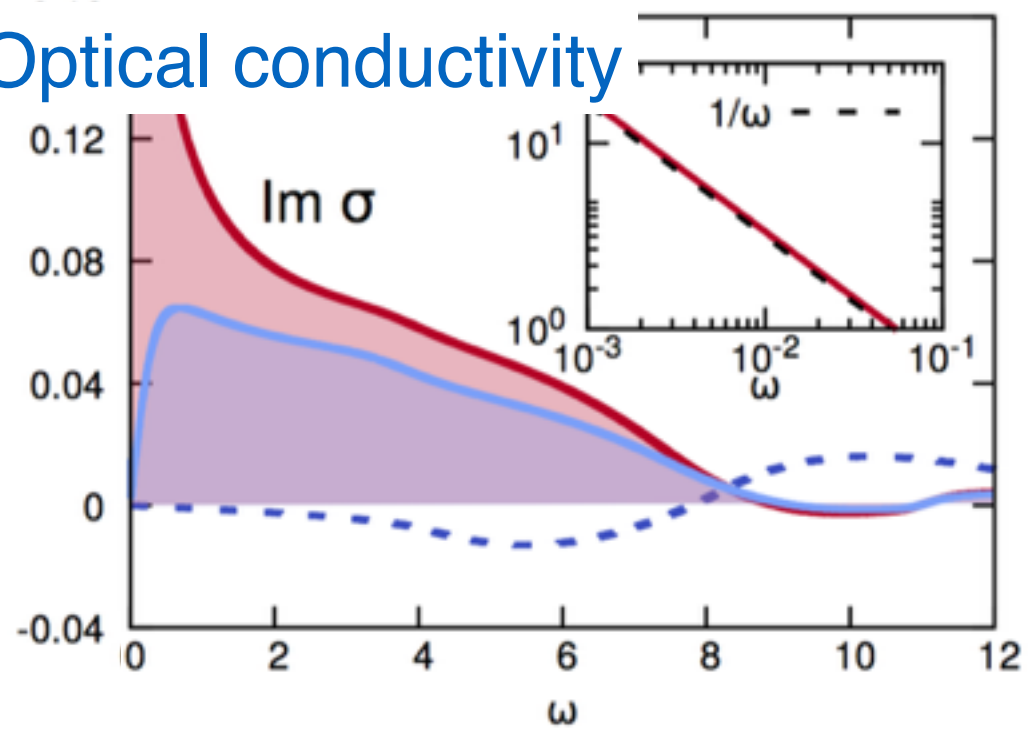
Phase-diagram of photo-doped Hubbard model



stable against NNN hopping

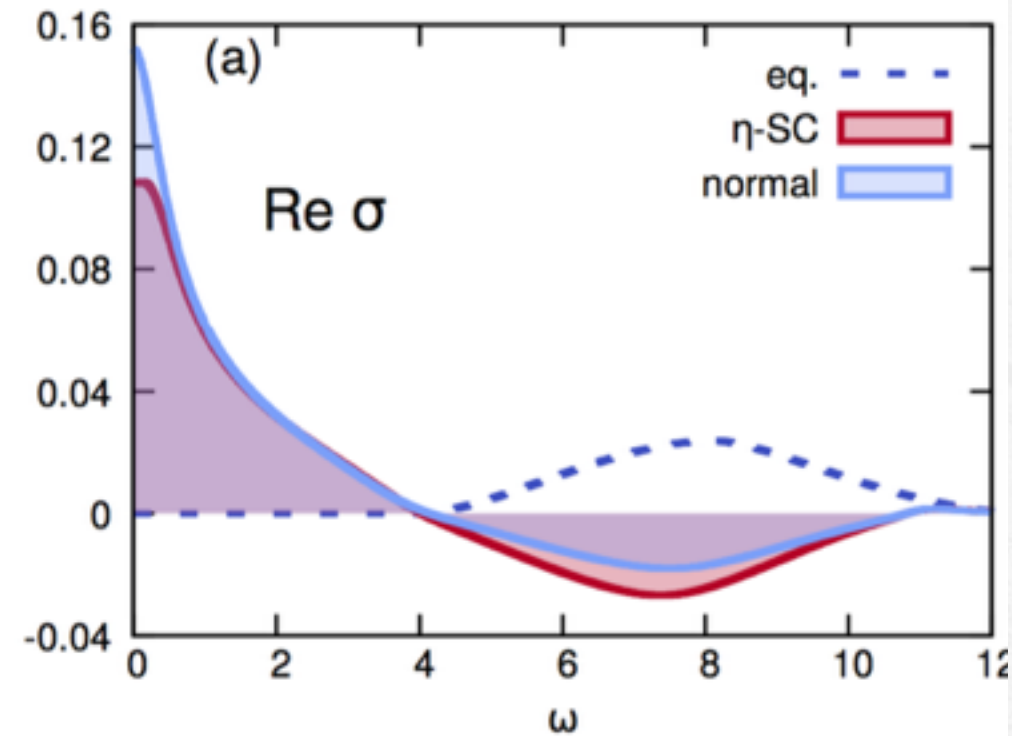
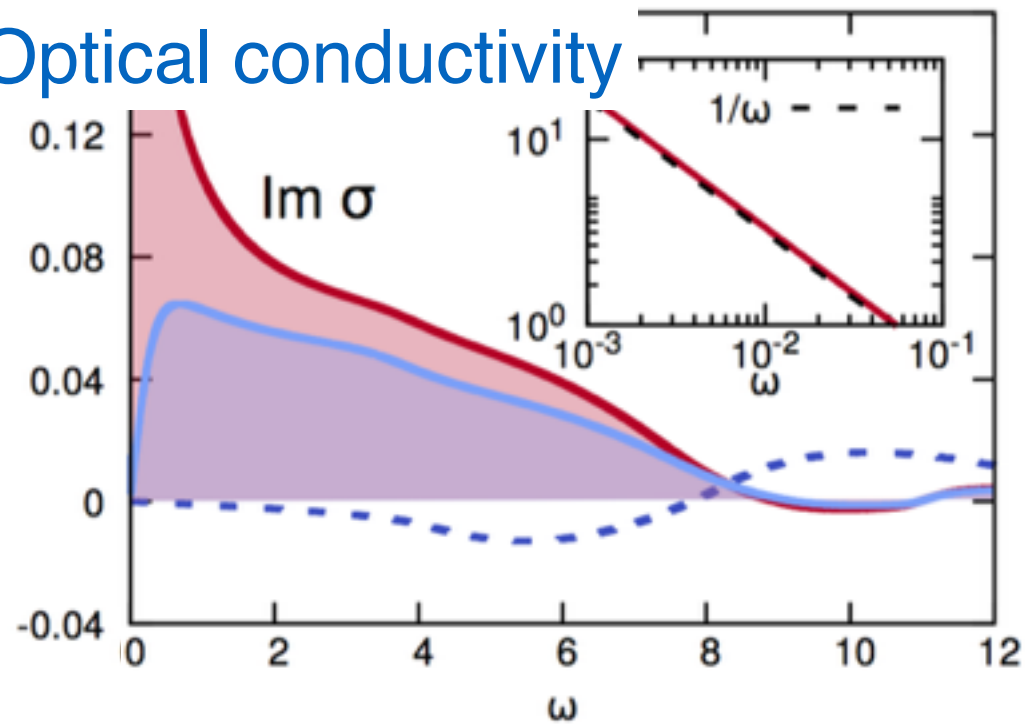
Meissner Effect

Optical conductivity

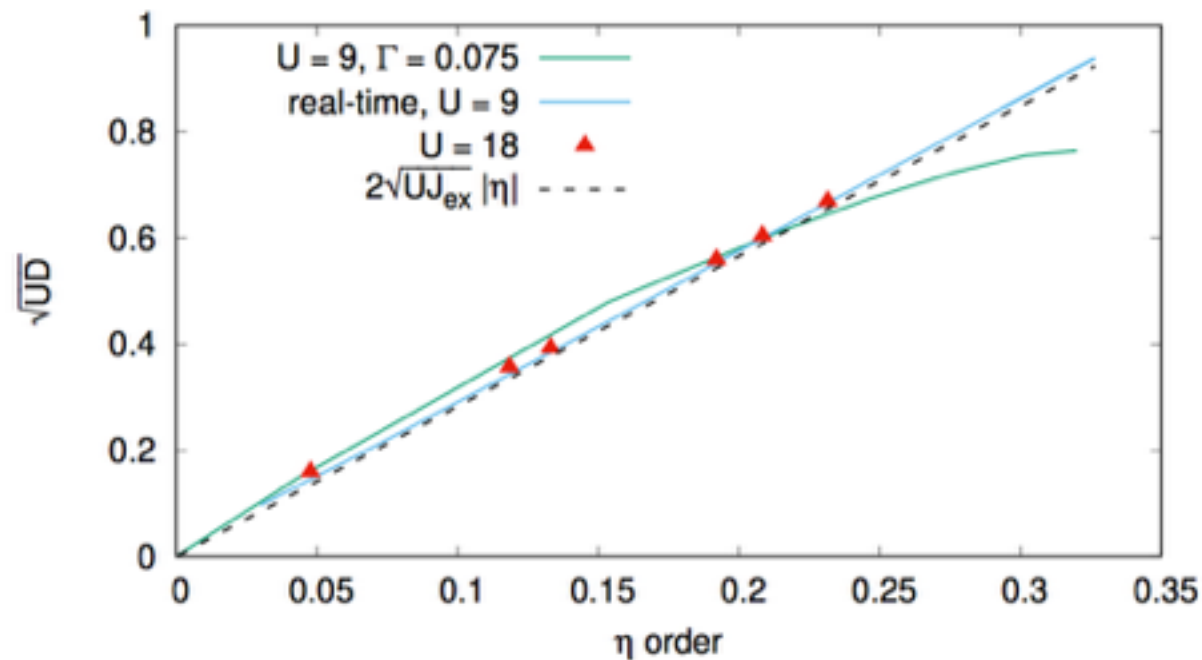


Meissner Effect

Optical conductivity



η -pairing phase: phase stiffness implies Meissner effect,



Drude weight ... from:

Sum rule: $\int_0^\infty d\omega \sigma(\omega) = -\frac{\pi}{4} E_{kin}$

$1/\omega$ behavior

real time $\partial j / \partial A$

Strong-coupling result: $D = 4J_{ex} |\eta|^2$

Summary

Photo doped states as NESS:

... access photo-doped phases in more complex systems ?

⇒ exploring photo-doped states in GW+EDMFT (realistic simulations)

⇒ Hidden states is systems with spin-orbital order
(Include feedback on lattice?)

⇒ Multi-orbital systems (SC in Cs_3C_{60})?

Boltzmann equation based on NESS ?

Photo doped states as NESS:

A protocol to reach cold photo-doped states

Philipp Werner, Martin Eckstein, Markus Müller, Gil Refael, *Photo-induced evaporative cooling*, arXiv:1904.00822 (Nature Comm.)

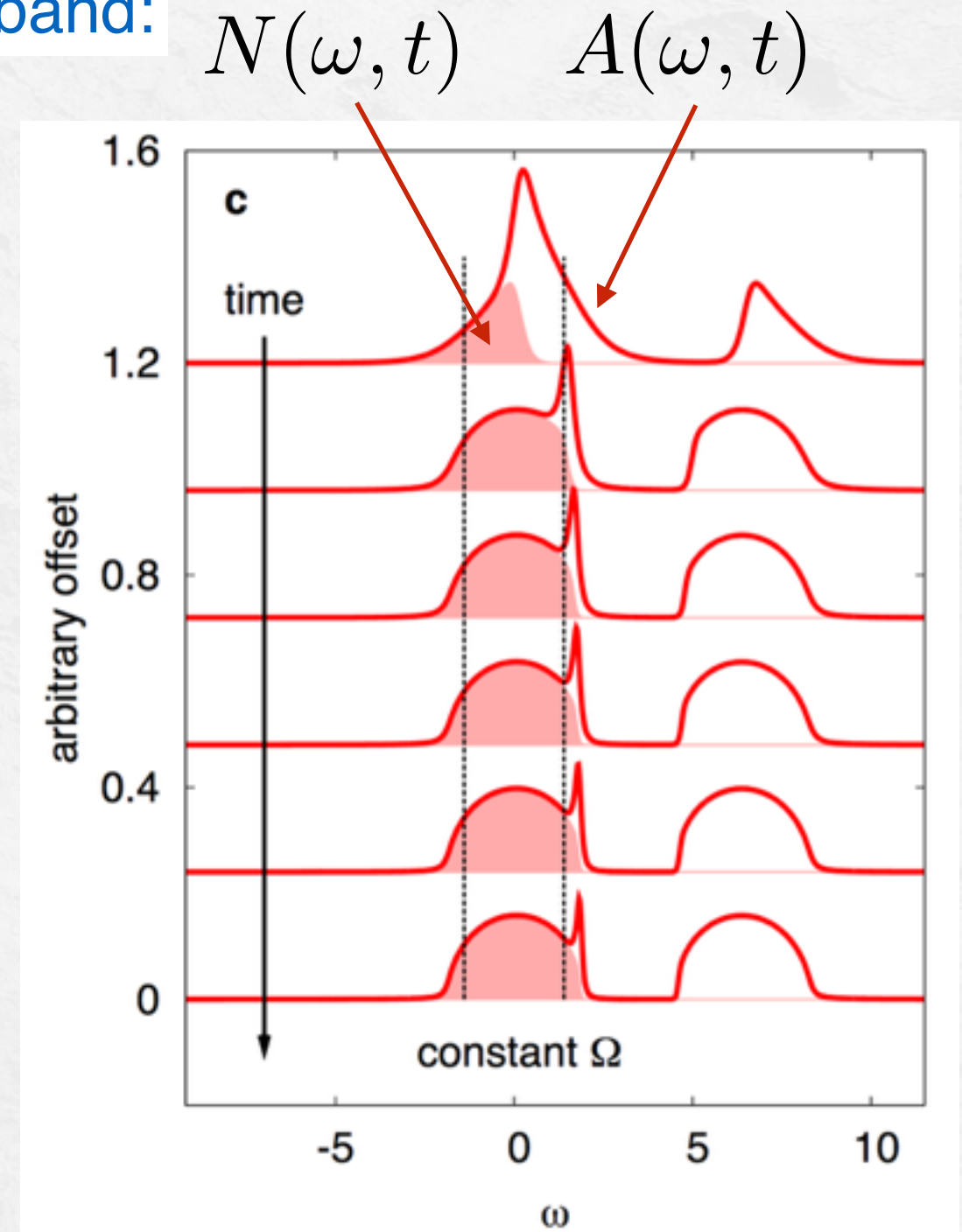
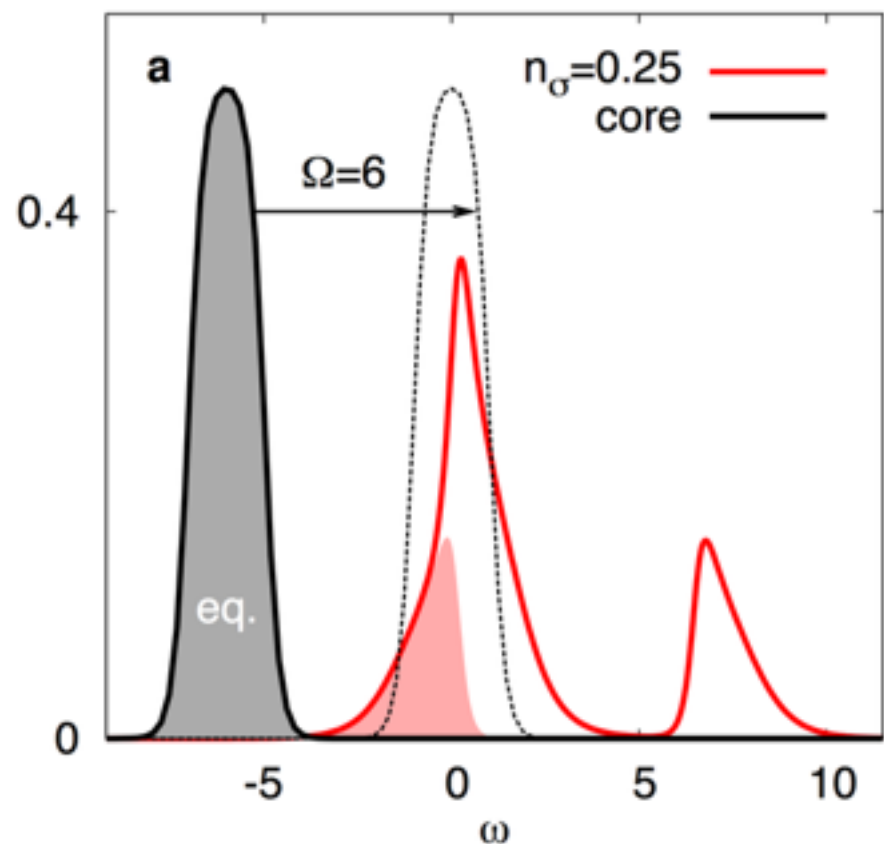


Entropic cooling

Single-band Hubbard model + (uncorrelated) band:

$$\begin{aligned}
 H = & - \sum_{ij\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow} \\
 & + \sum_{k\sigma} \epsilon_k^{core} a_{k\sigma}^\dagger a_{k\sigma} \\
 & + \sum_{k\sigma} \left(p_k e^{i\Omega t} c_{k\sigma}^\dagger a_{k\sigma} + h.c. \right)
 \end{aligned}$$

dipole transition

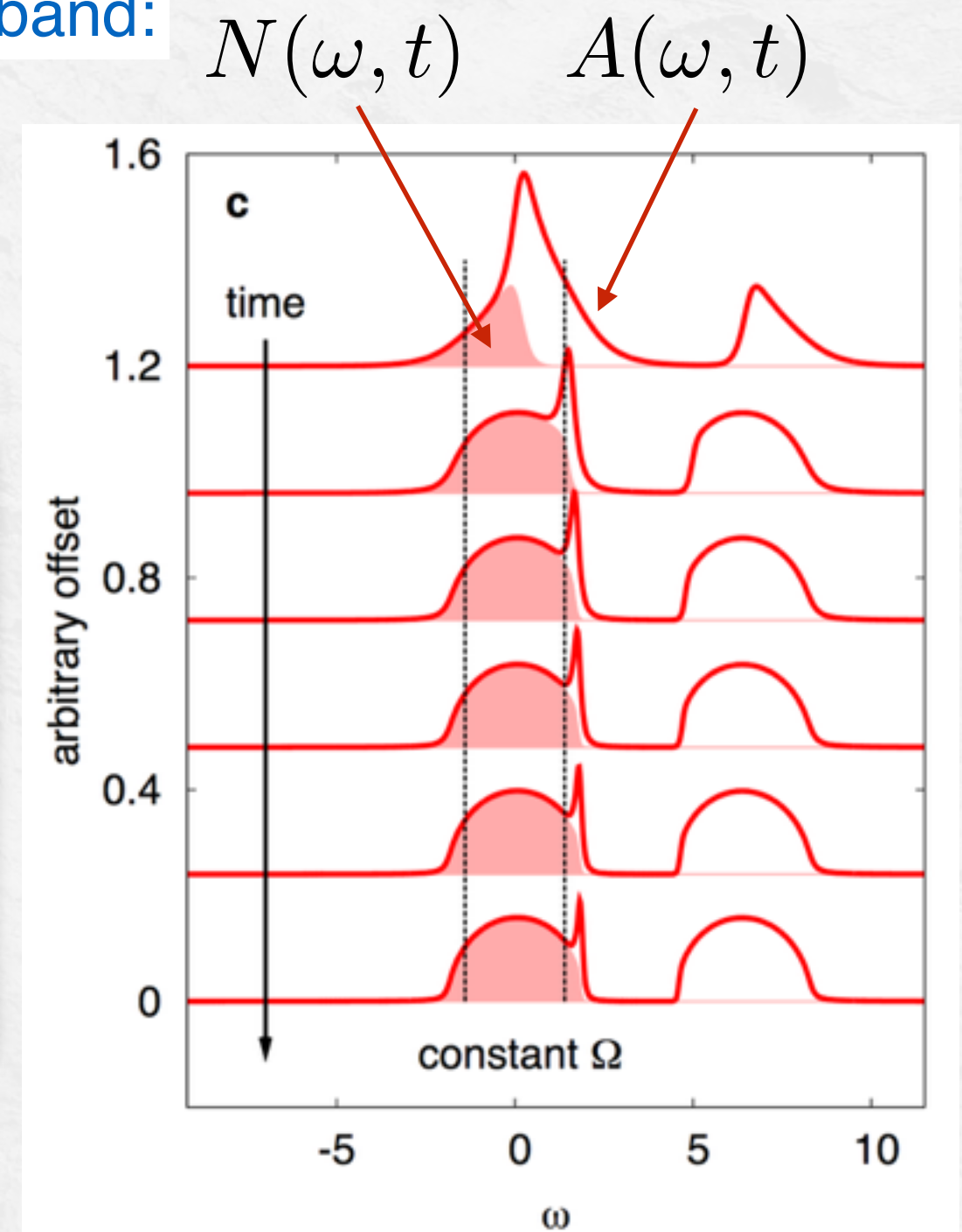
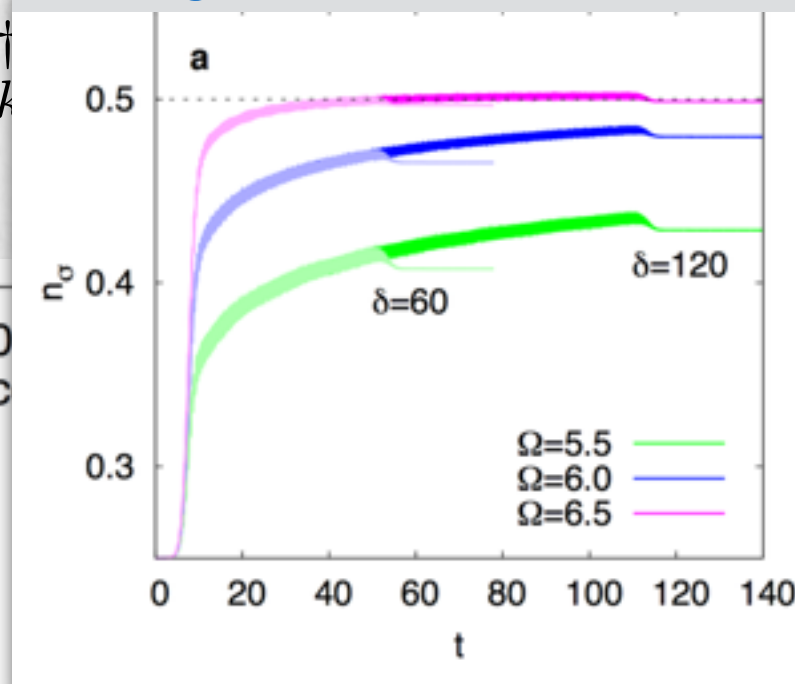
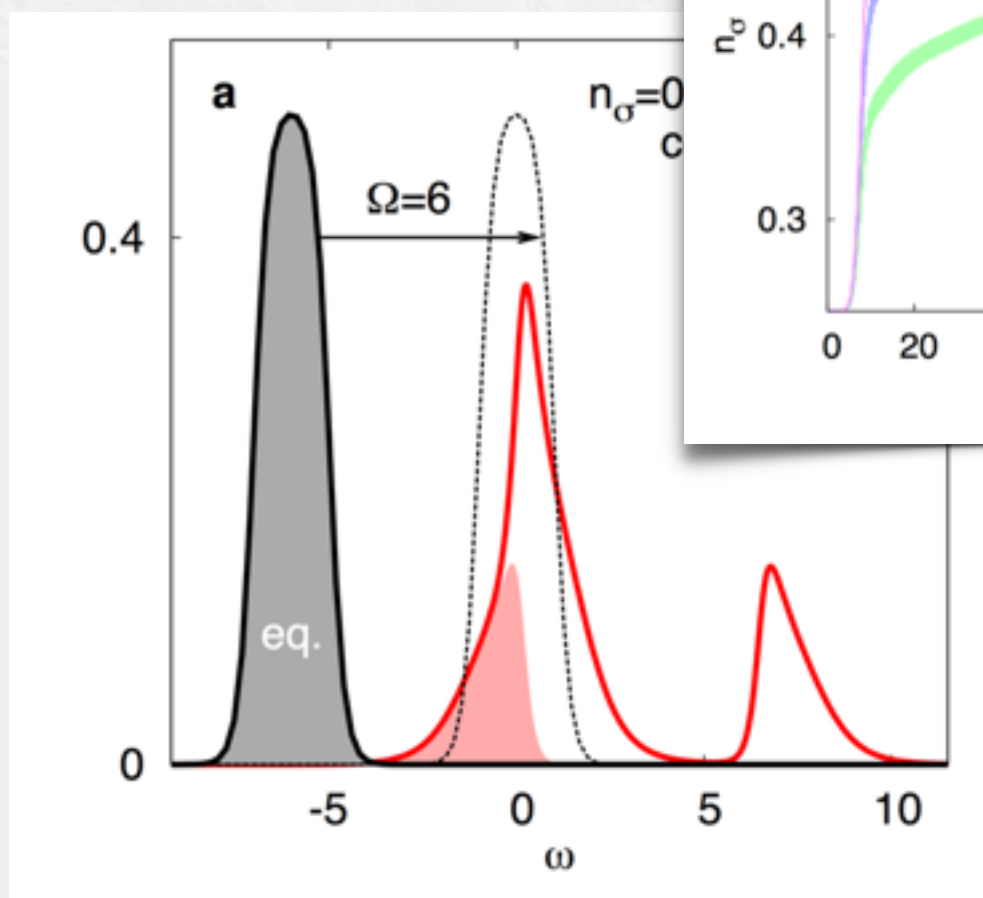


Entropic cooling

Single-band Hubbard model + (uncorrelated) band:

$$H = - \sum_{ij\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow} + \sum_{k\sigma} \epsilon_k^{core} a_{k\sigma}^\dagger a_{k\sigma} + \dots + \sum_{k\sigma} (p_k e^{i\Omega t} c_{k\sigma}^\dagger + \text{h.c.})$$

Filling (valence band)



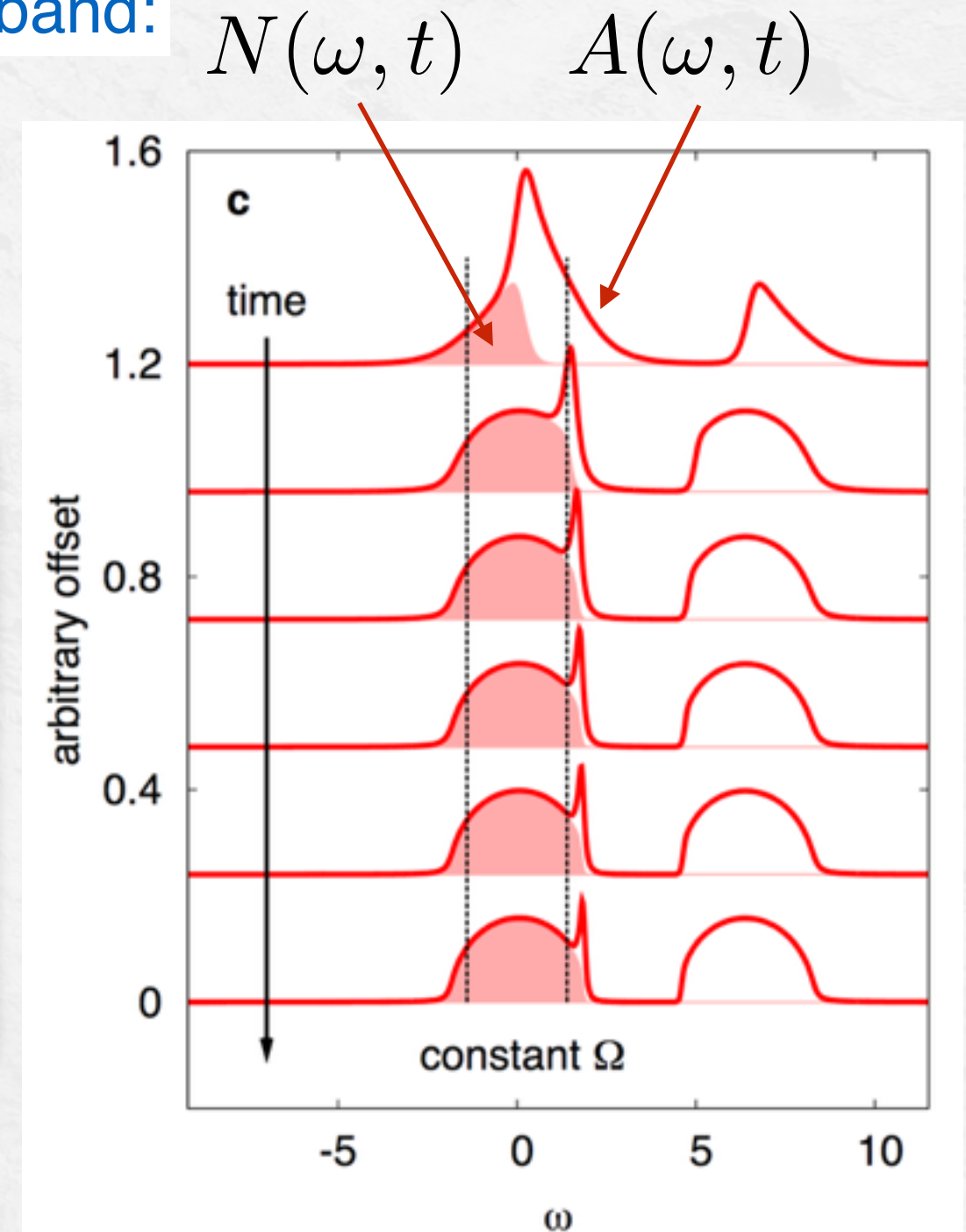
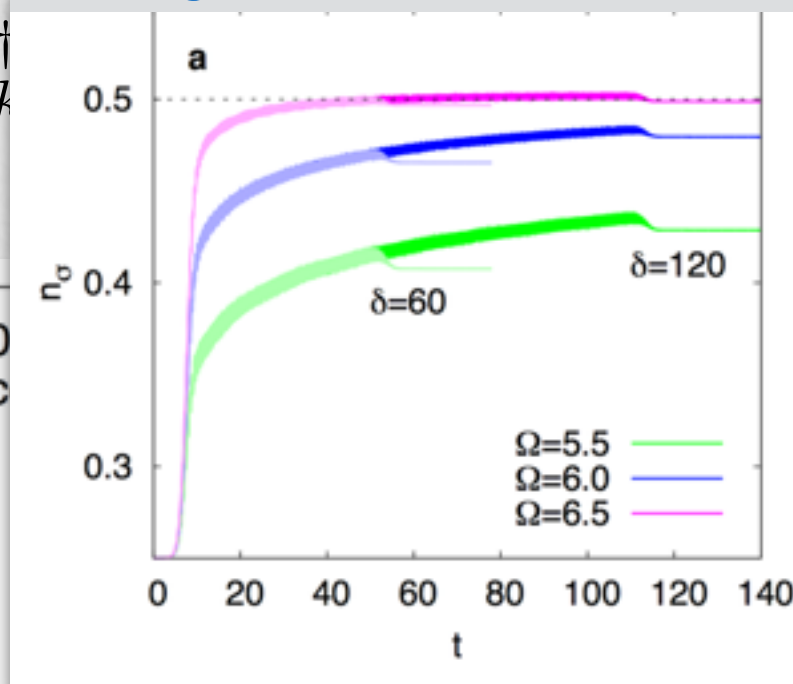
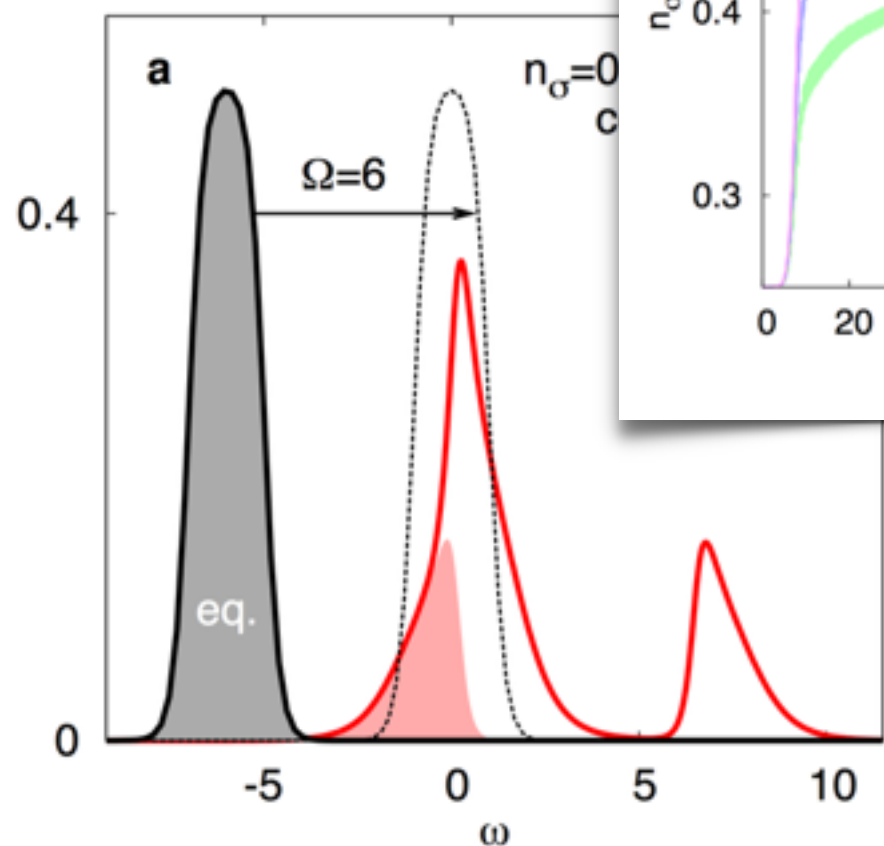
⇒ Filling increased towards $n=1/2$

Entropic cooling

Single-band Hubbard model + (uncorrelated) band:

$$H = - \sum_{ij\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + U \sum_j n_{j\uparrow} n_{j\downarrow} + \sum_{k\sigma} \epsilon_k^{core} a_{k\sigma}^\dagger a_{k\sigma} + \sum_{k\sigma} (p_k e^{i\Omega t} c_{k\sigma}^\dagger a_{k\sigma} + \text{h.c.})$$

Filling (valence band)



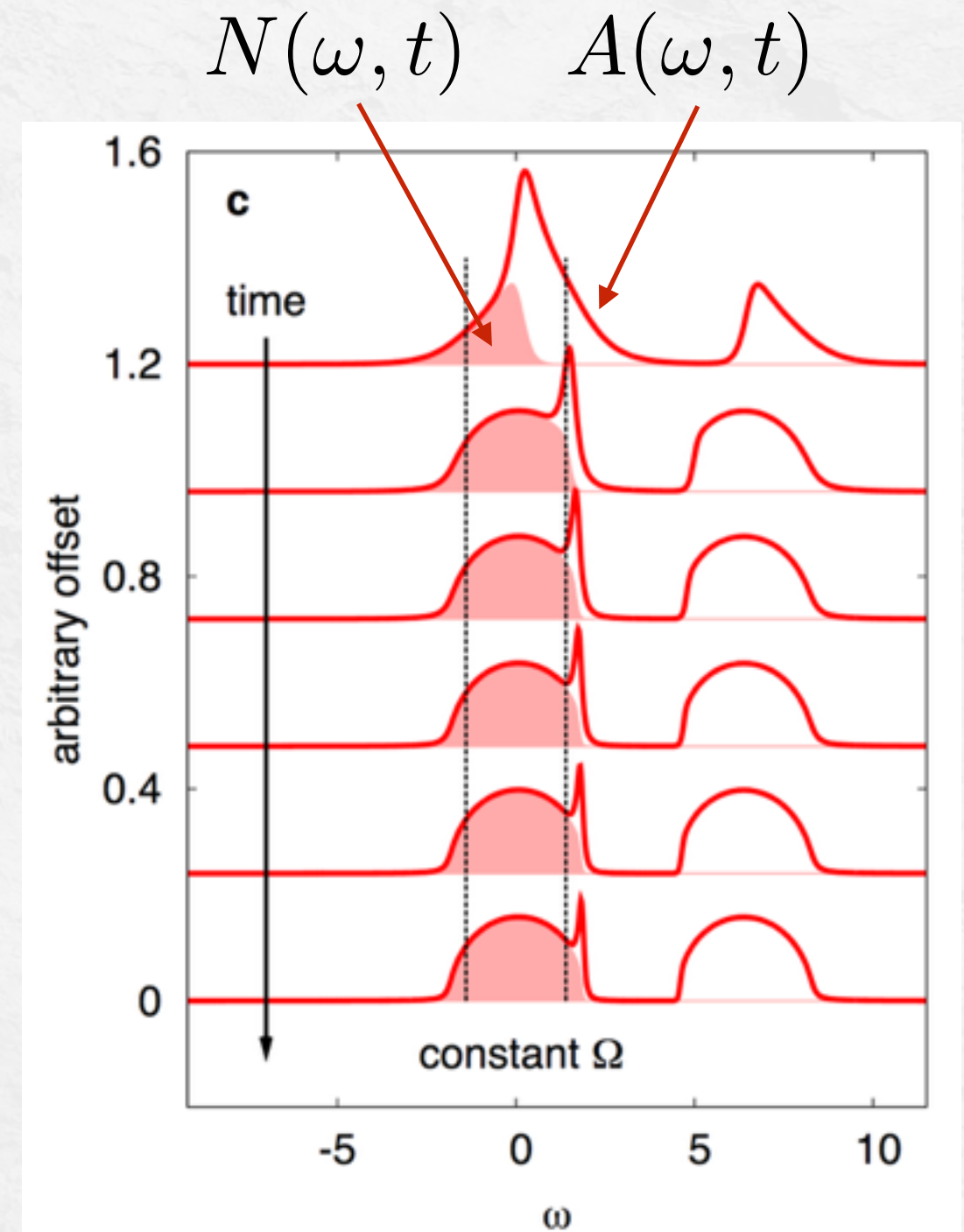
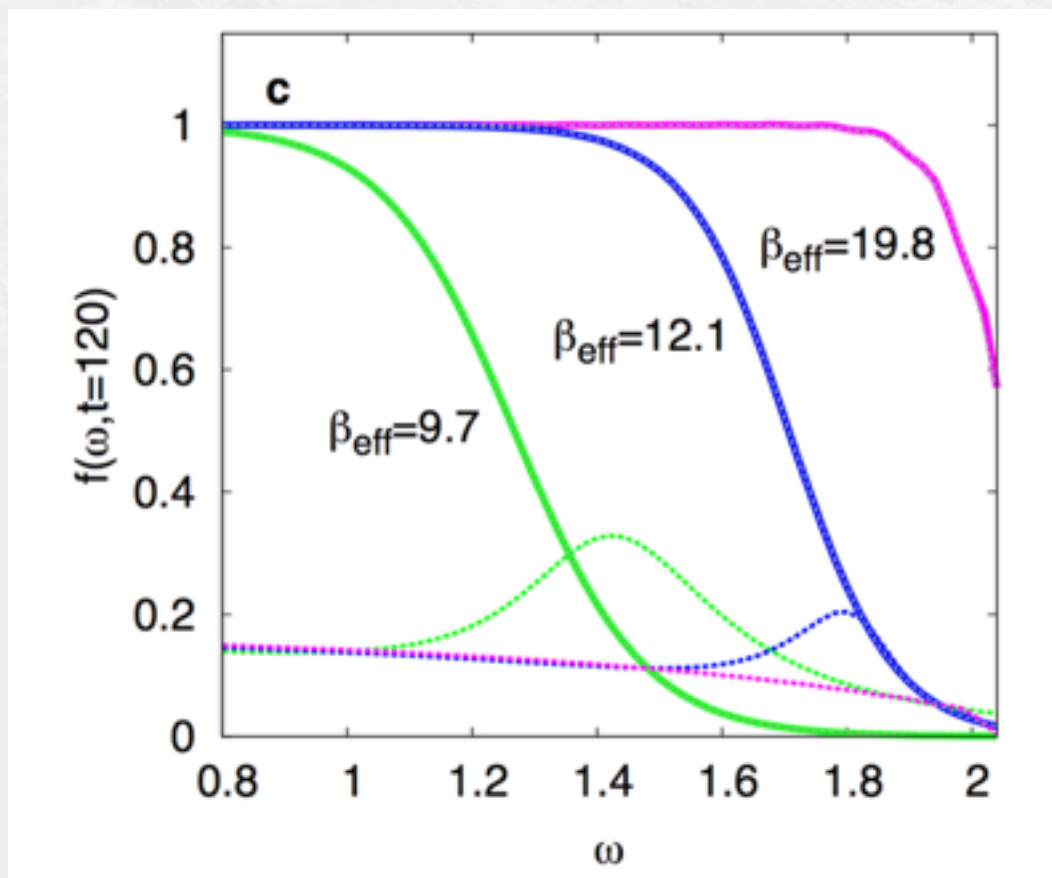
⇒ Filling increased towards $n=1/2$

⇒ System becomes more correlated

Entropic cooling

Distribution function

$$\frac{N(\omega, t)}{A(\omega, t)} = F(\omega, t)$$

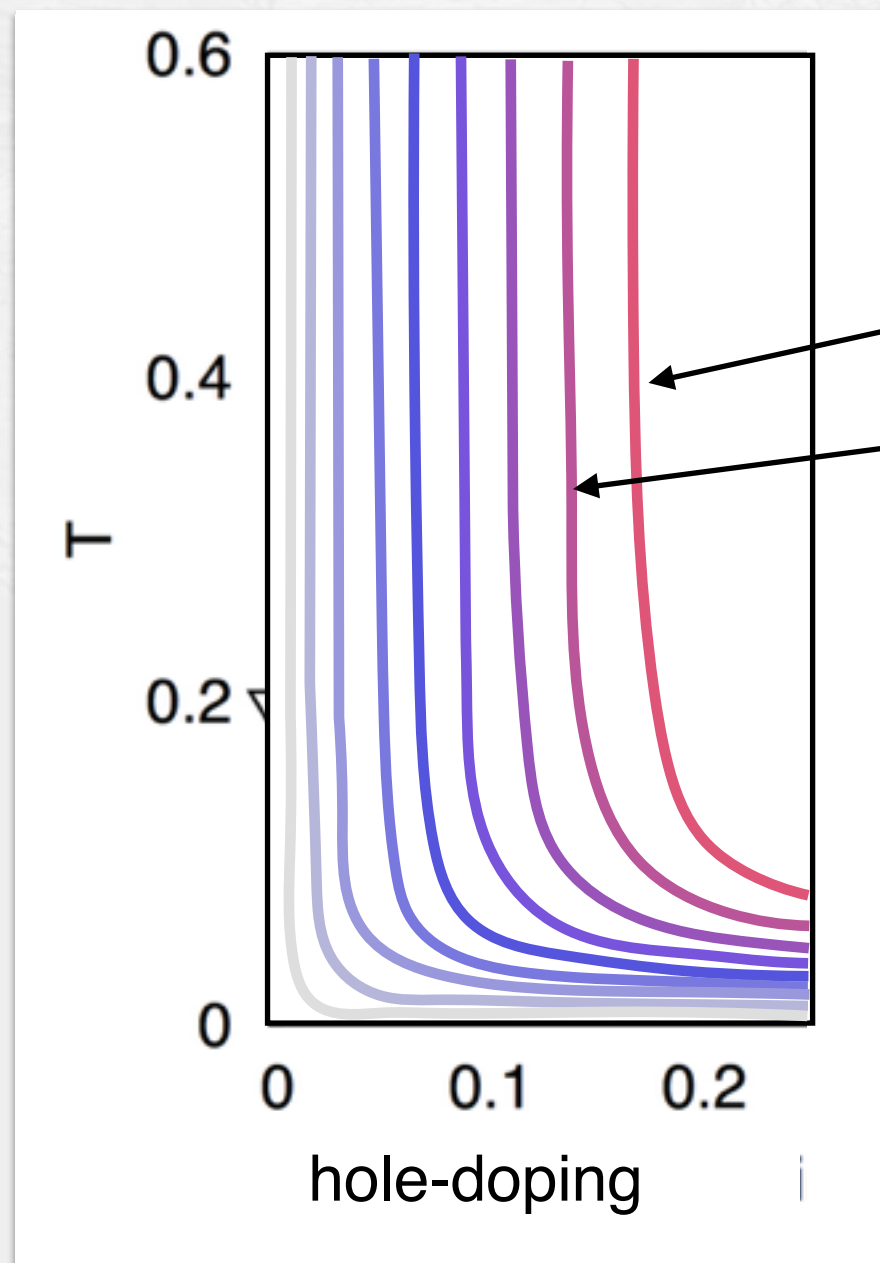


$\Rightarrow$ cooling: $T_{eff} = \frac{1}{\beta_{eff}} < T_{in}$ (initial temperature $T=0.2$, $\beta=5$)

Entropic cooling

Entropy per Volume:

Core band



$$S = 0.9$$

$$S = 0.8$$

...

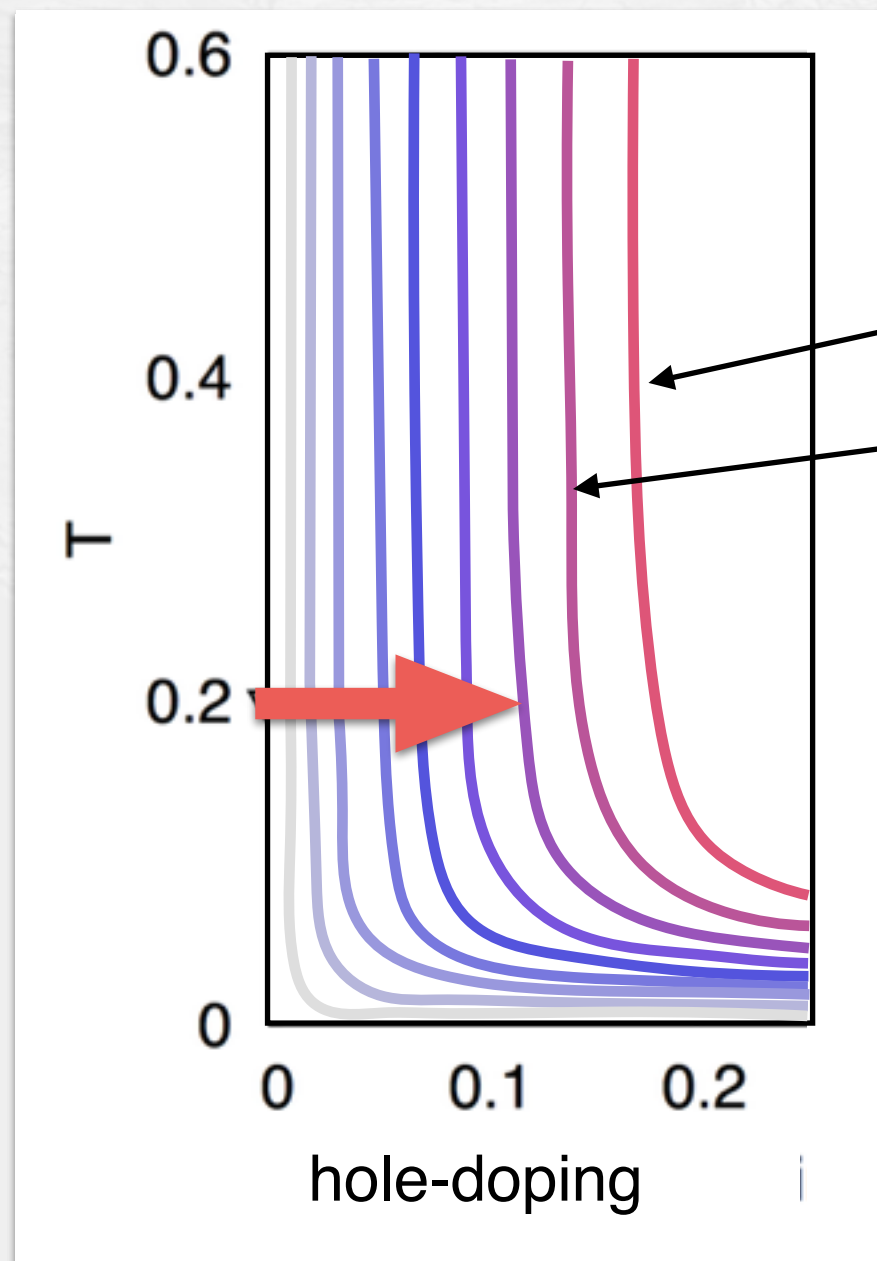
$T > \text{bandwidth}$

$$S = -2[n \log(n) + (1 - n) \log(1 - n)]$$

Entropic cooling

Entropy per Volume:

Core band



$T > \text{bandwidth}$

$$S = -2[n \log(n) + (1 - n) \log(1 - n)]$$

⇒ Strong increase with hole doping

Entropic cooling

Entropy per Volume:

$n=0.5$ (Mott):

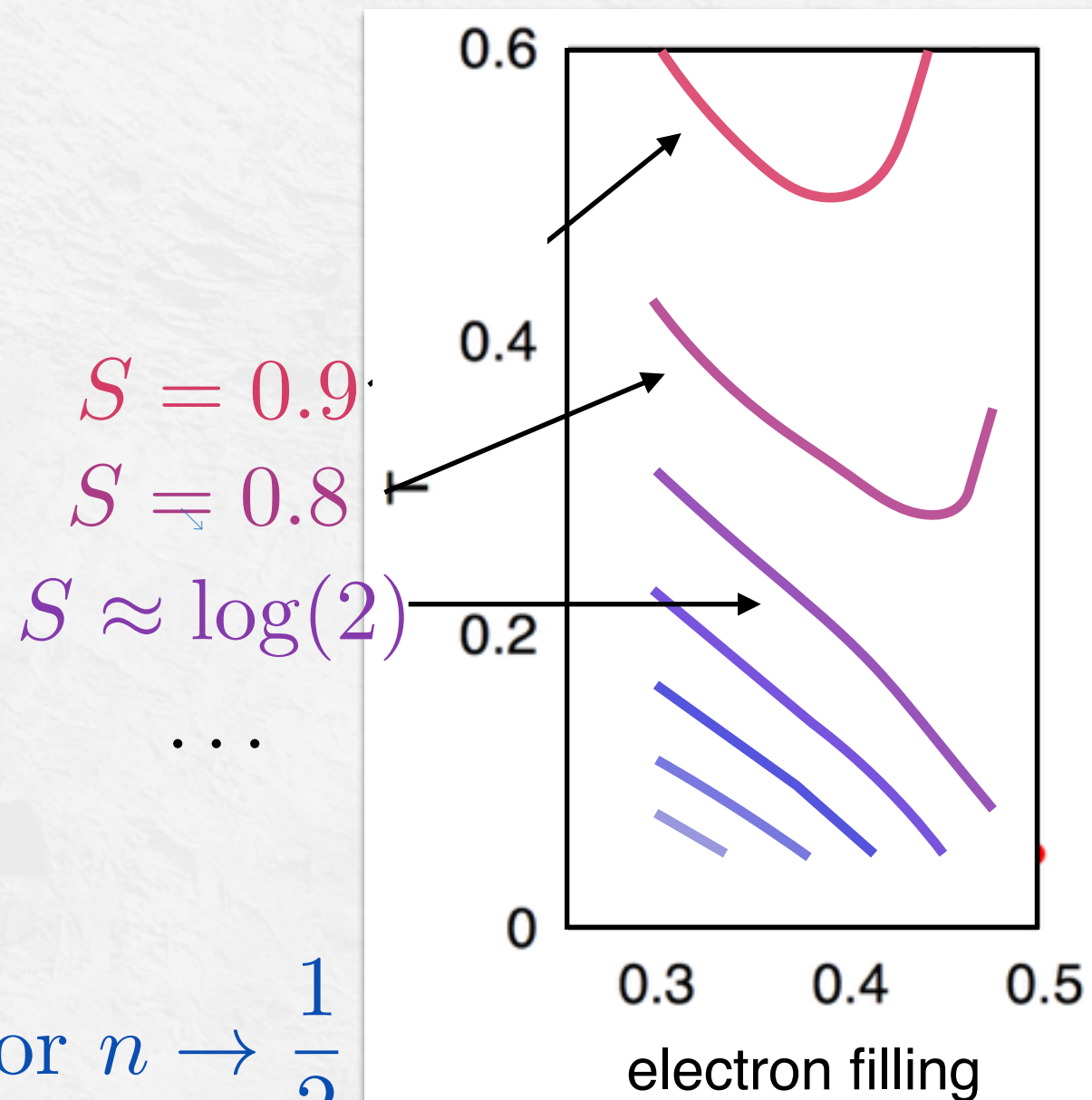
$S \approx \ln(2)$ at $T > 0$
(spin disordered state)

further increase:
Charge excitations

$n < 0.5$: $T \rightarrow 0$: Fermi liquid

$$S = \gamma(n)T \quad \gamma(n) \rightarrow \infty \text{ for } n \rightarrow \frac{1}{2}$$

Valence band



Entropic cooling

Entropy per Volume:

$n=0.5$ (Mott):

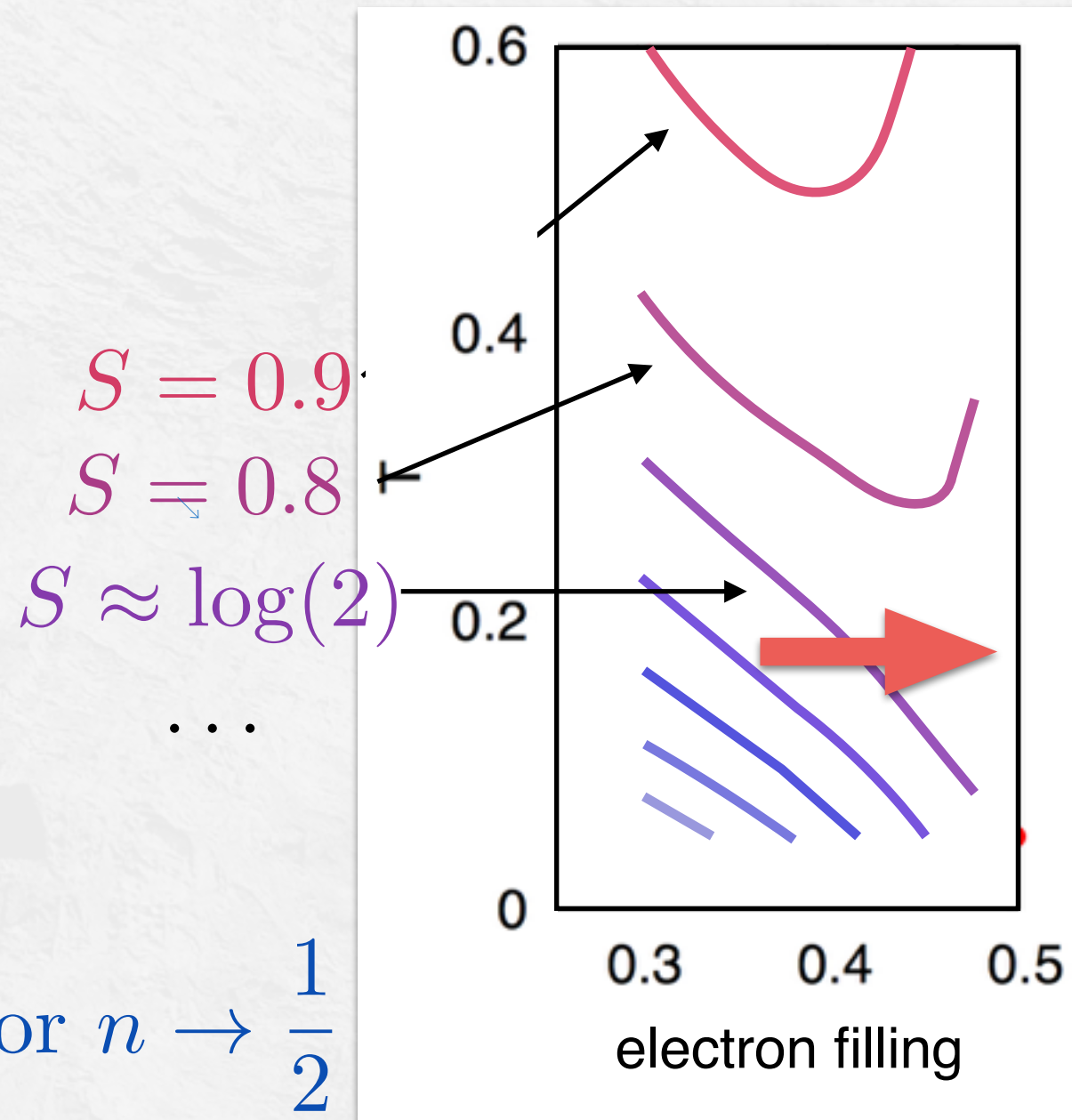
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Valence band

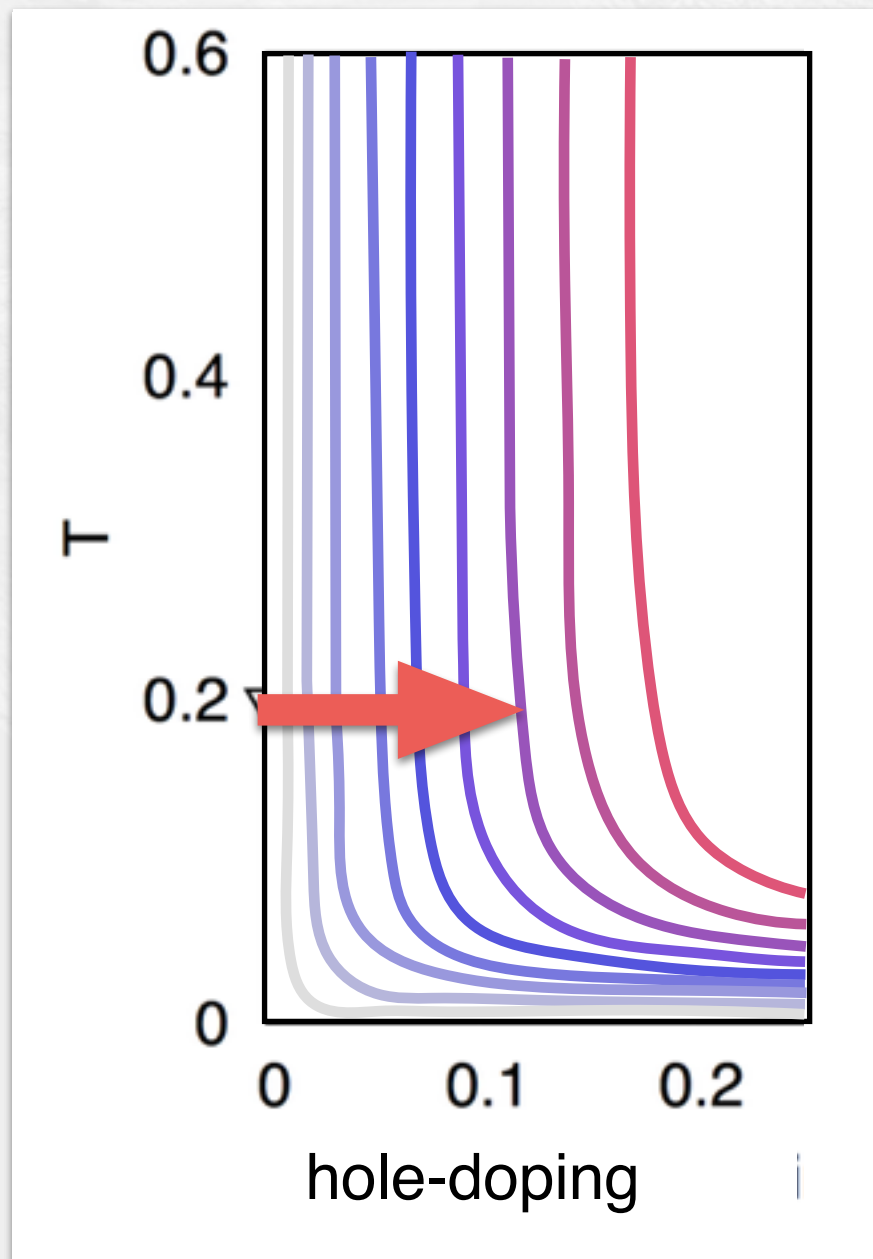


⇒ Increase for filling towards 1/2

Entropic cooling

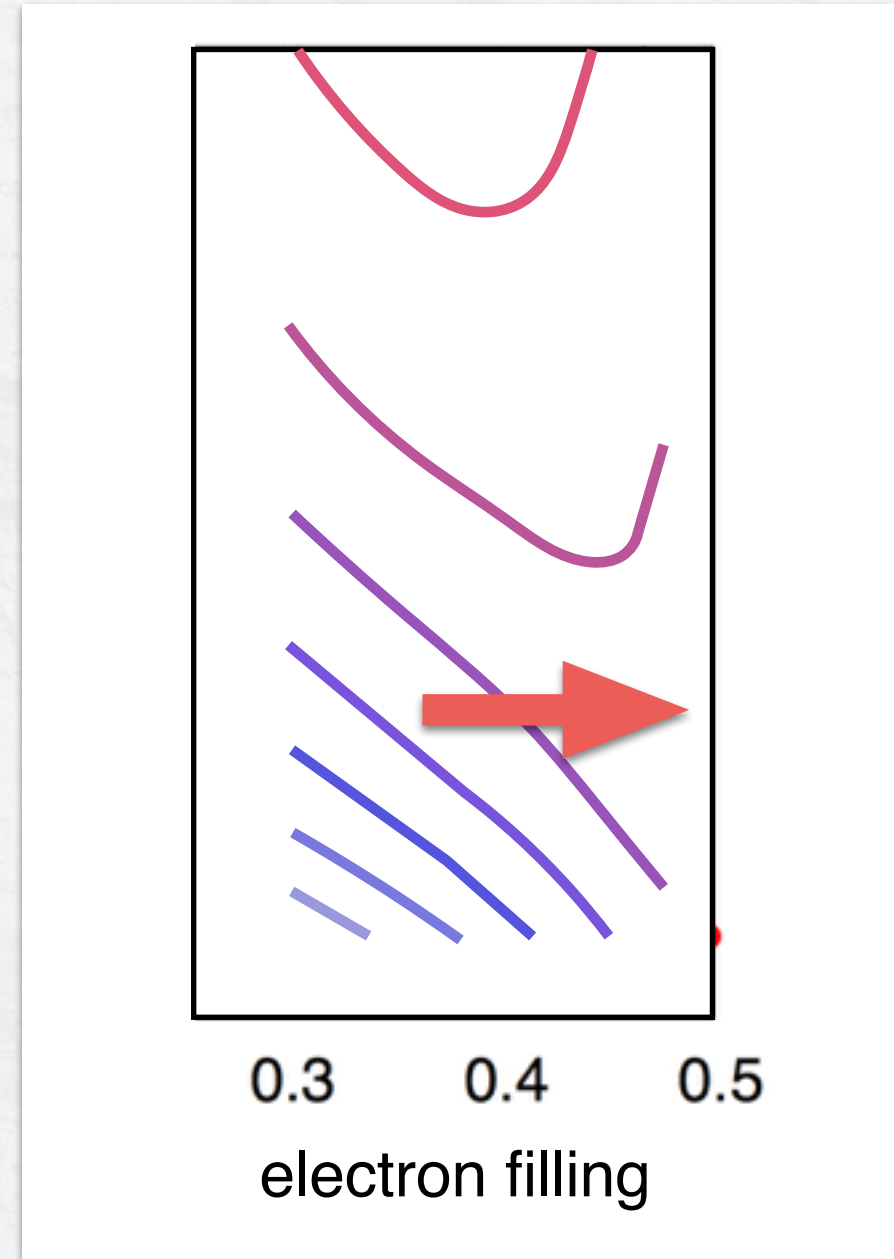
Entropy per Volume:

Core band



⇒ Strong increase with hole doping

Valence band

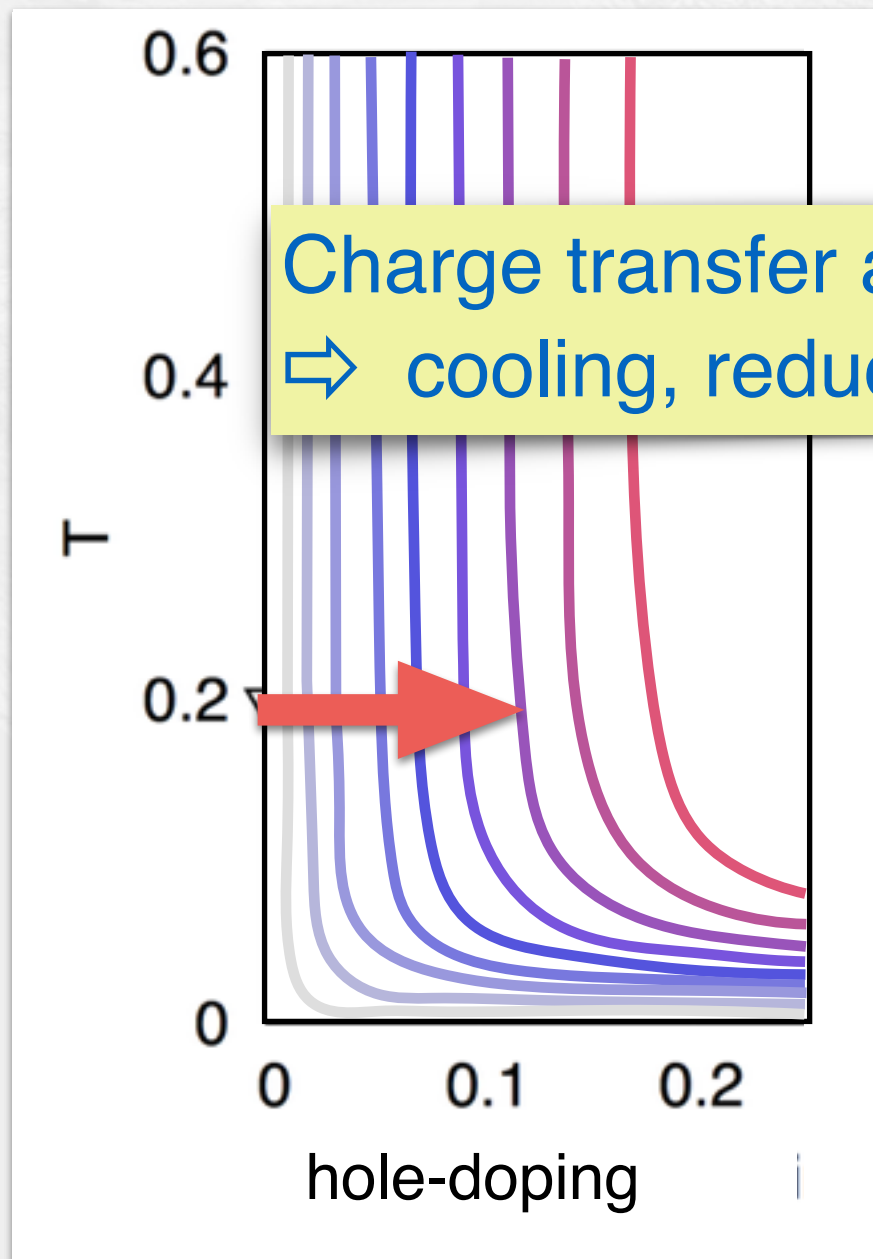


⇒ Increase for filling towards 1/2

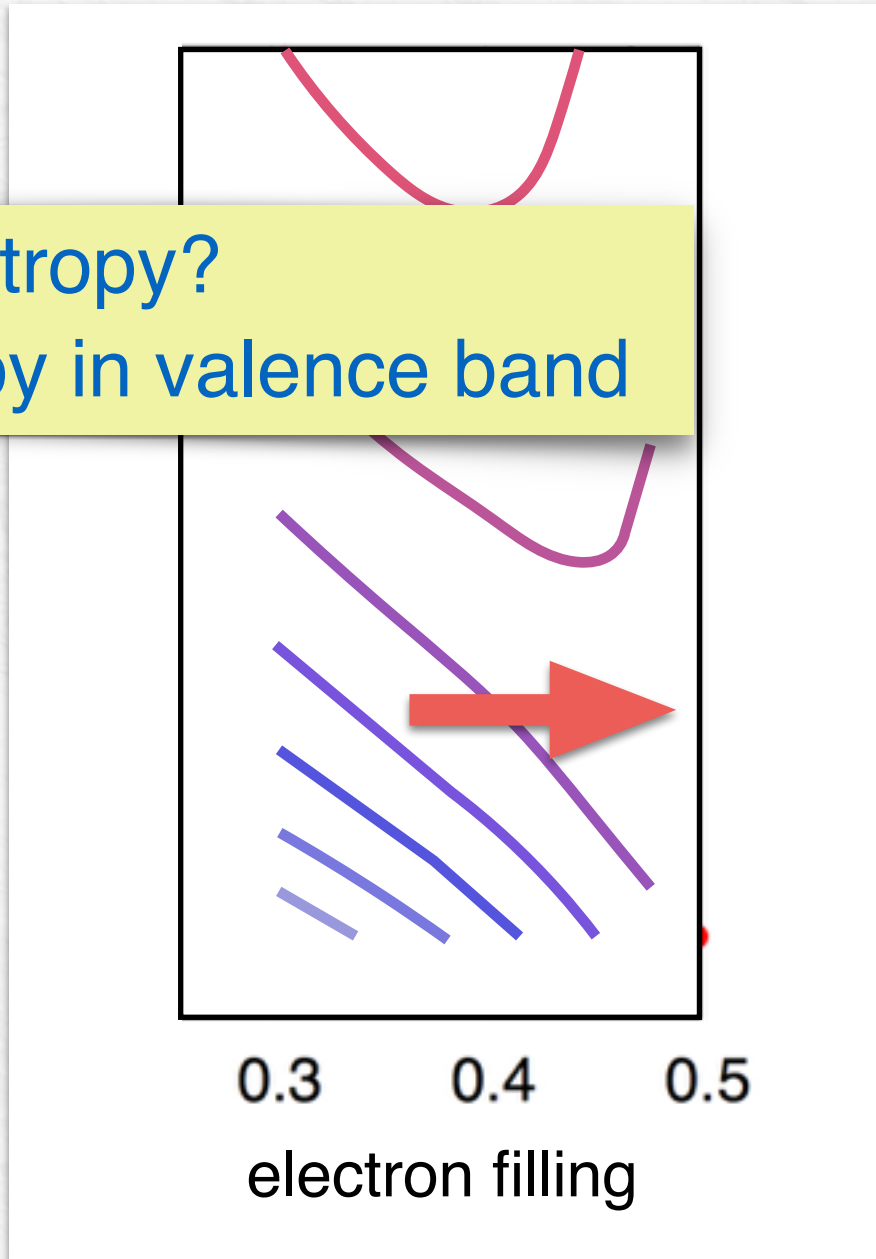
Entropic cooling

Entropy per Volume:

Core band



Valence band



Charge transfer at constant entropy?

⇒ cooling, reduction of entropy in valence band

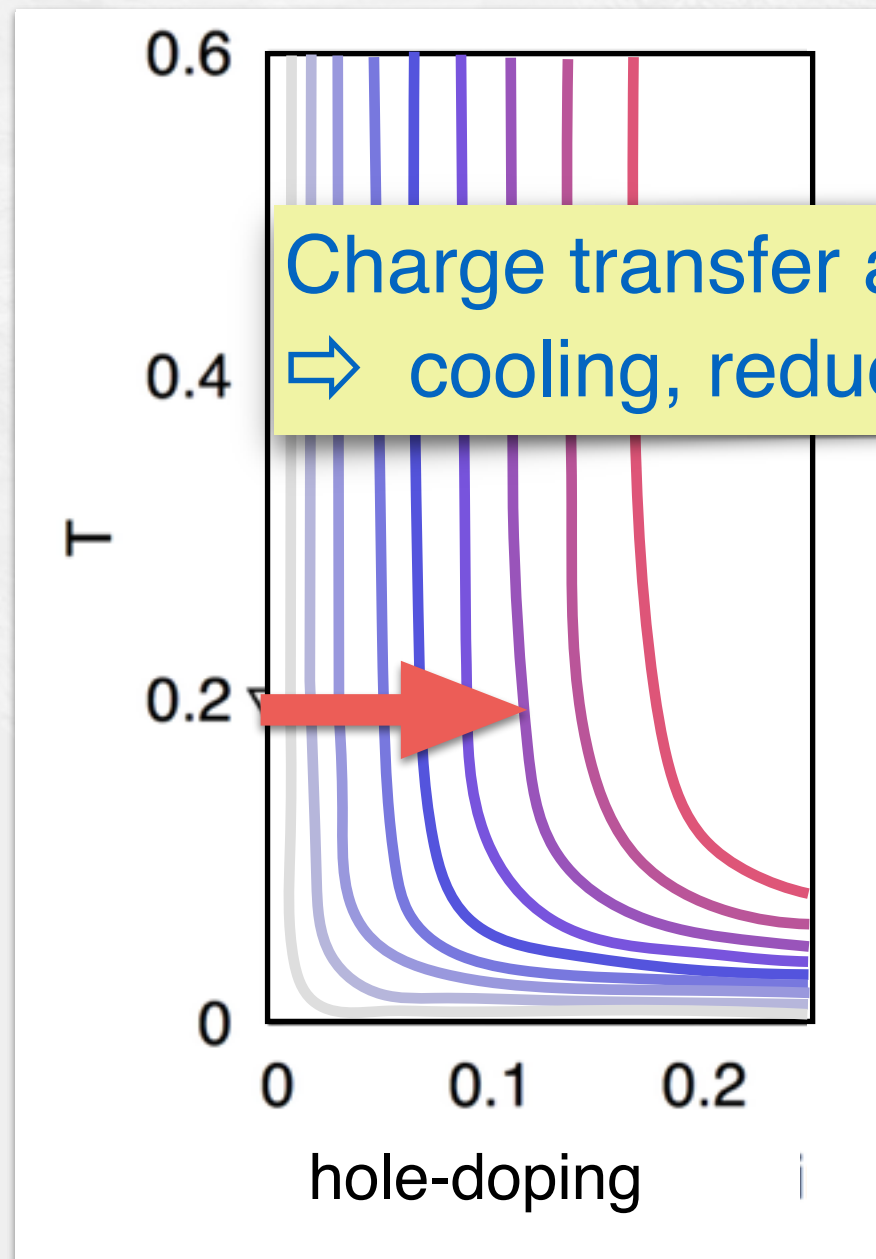
⇒ Strong increase with hole doping

⇒ Increase for filling towards 1/2

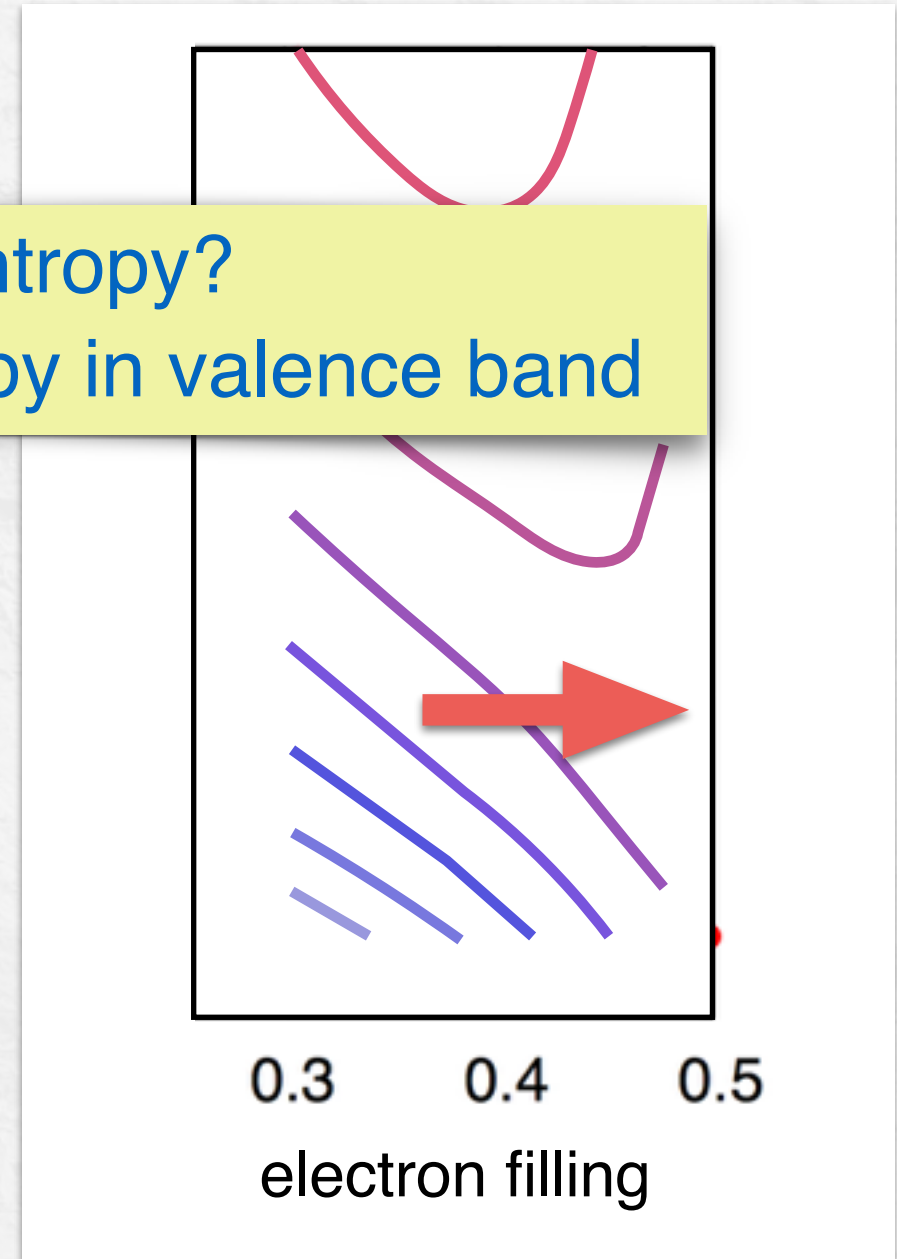
Entropic cooling

Entropy per Volume:

Core band



Valence band



Charge transfer at constant entropy?

⇒ cooling, reduction of entropy in valence band

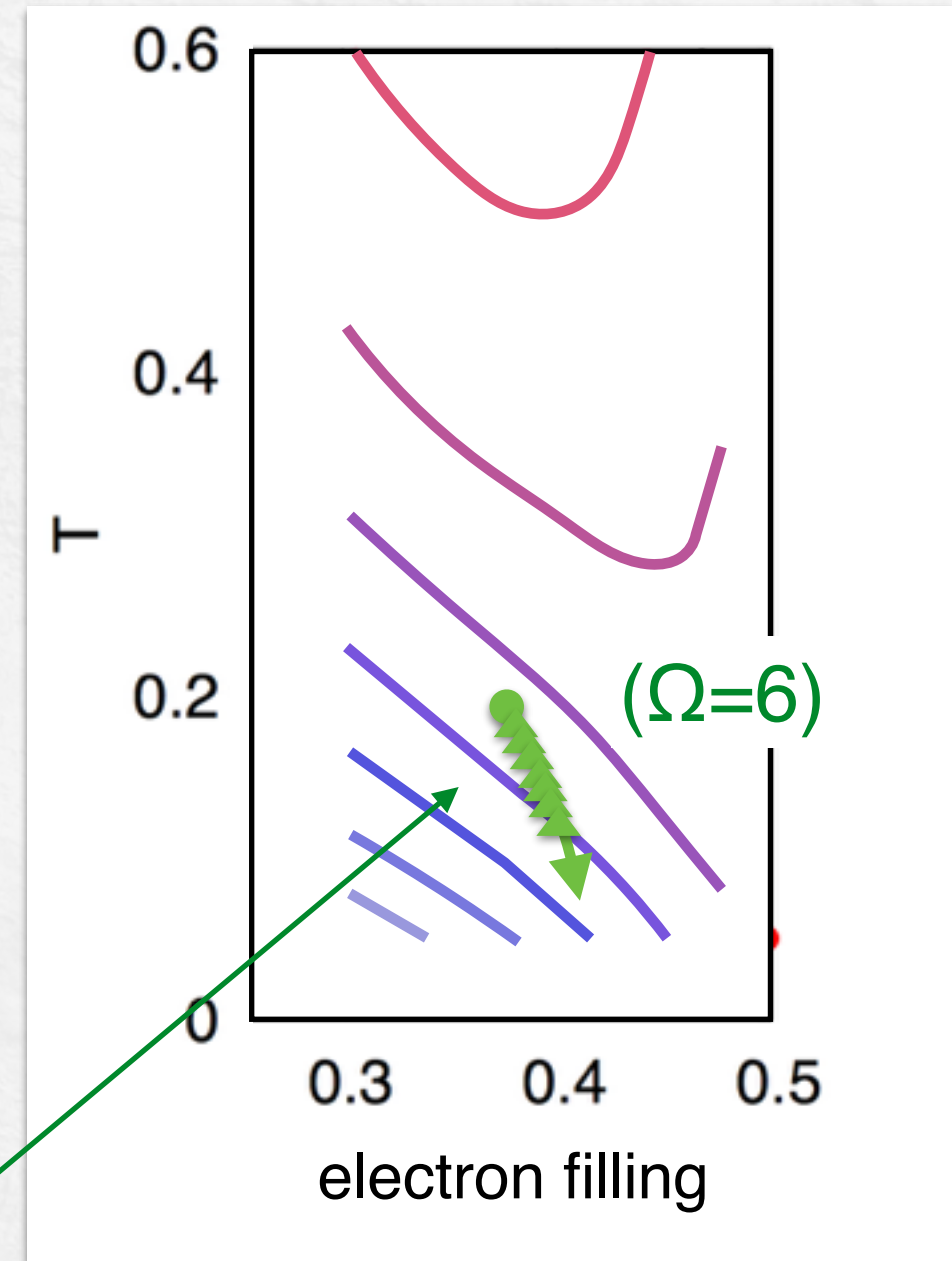
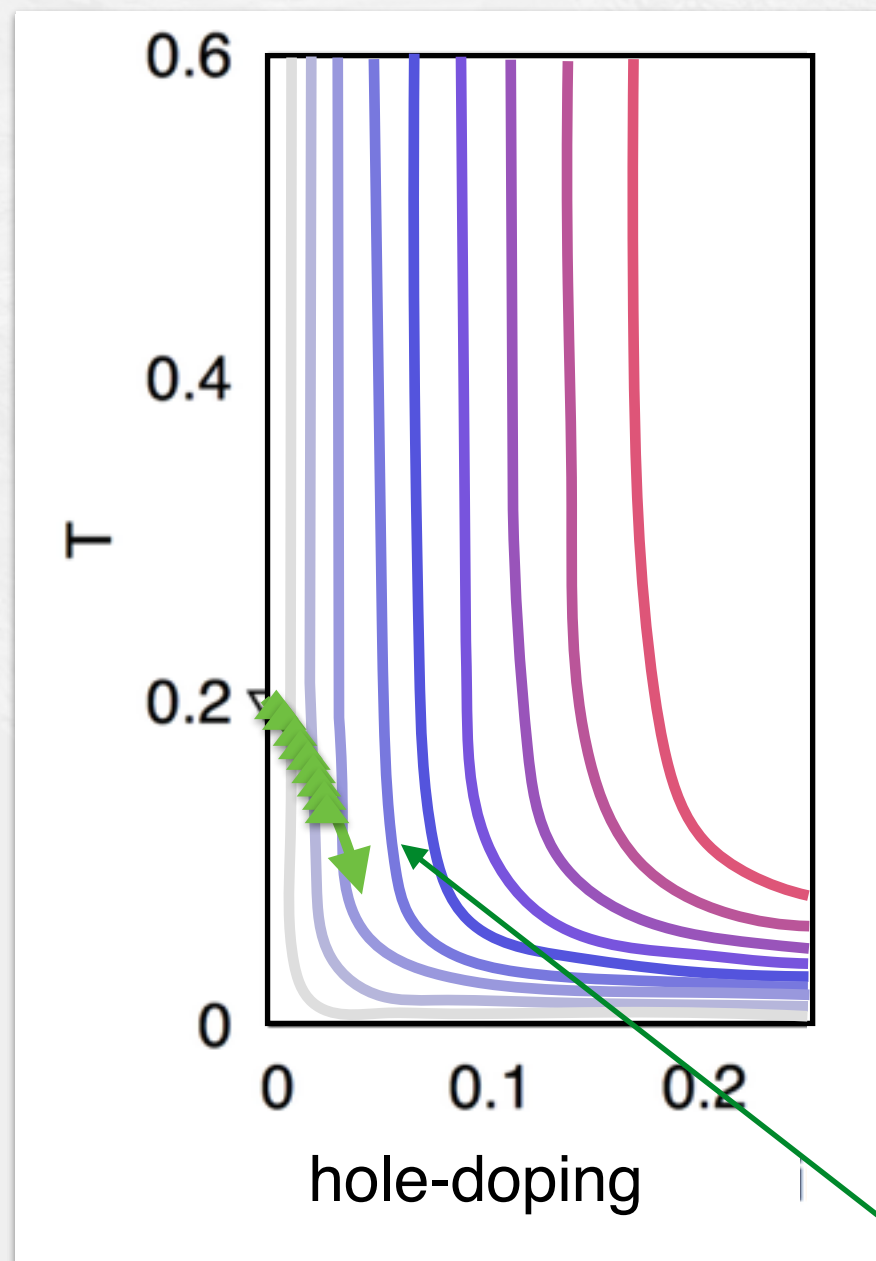


⇒ Strong increase with hole doping

⇒ Increase for filling towards 1/2

Entropic cooling

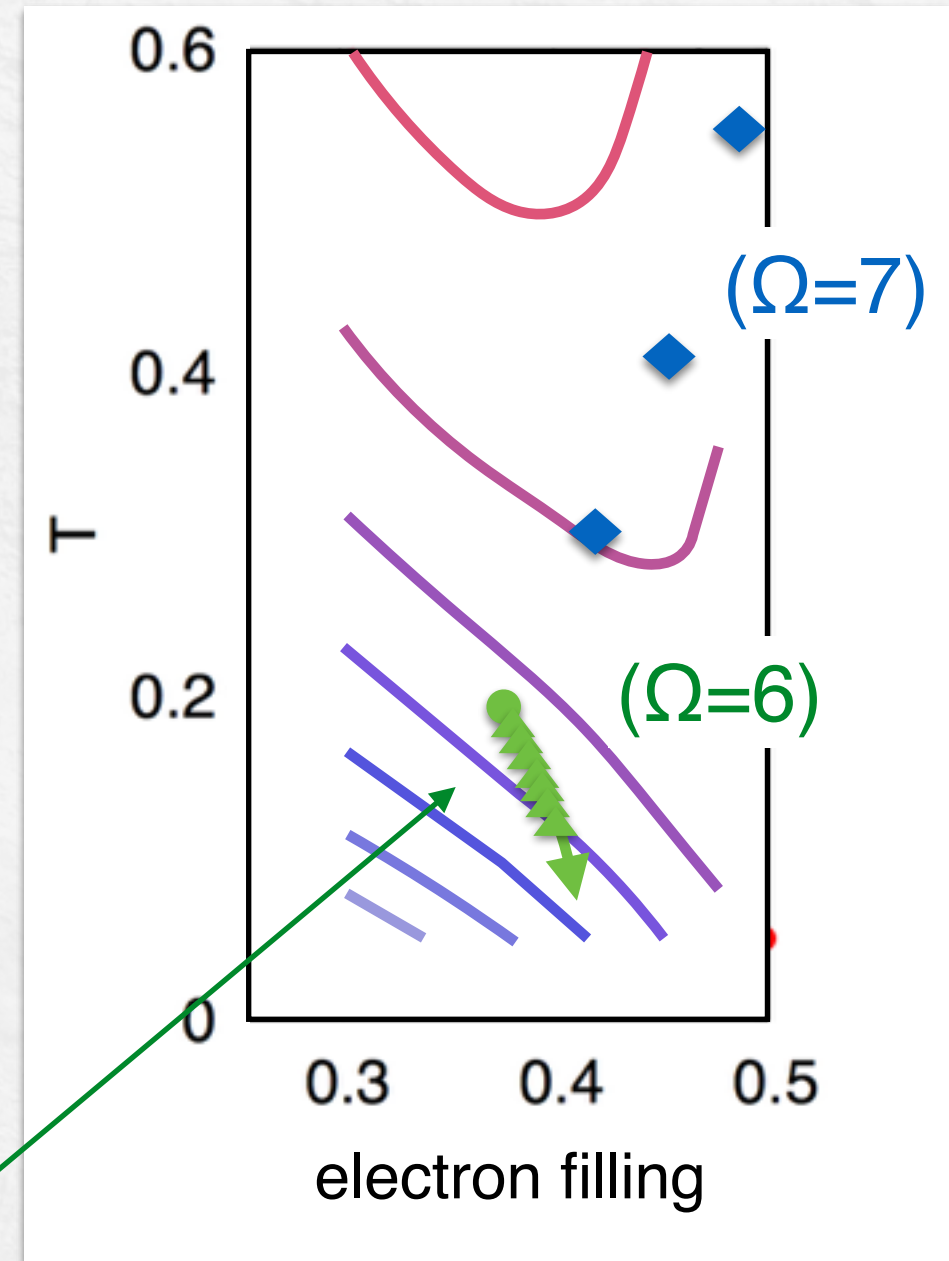
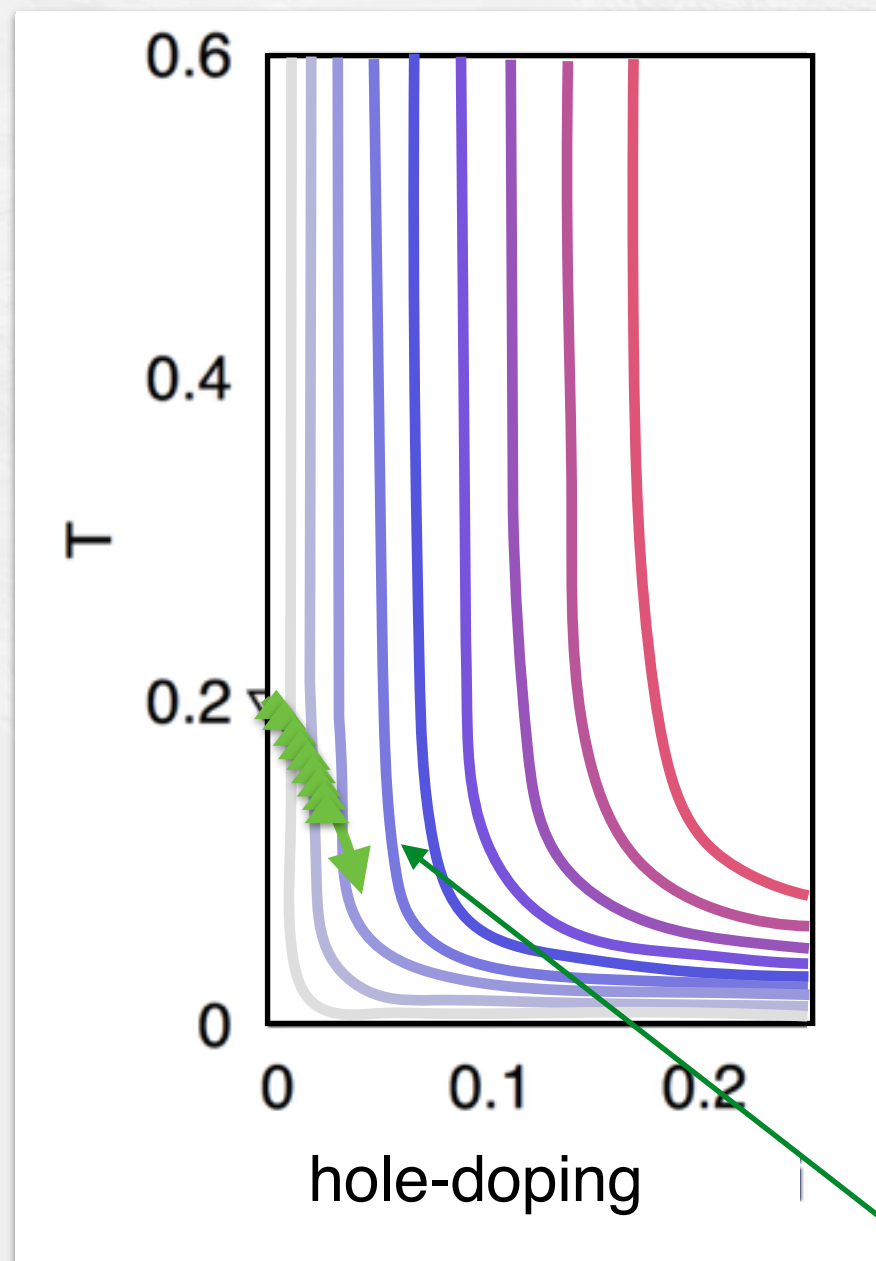
Charge transfer at constant entropy?



Effective Temperature after real (dynamic) charge transfer ($\Omega=6$)

Entropic cooling

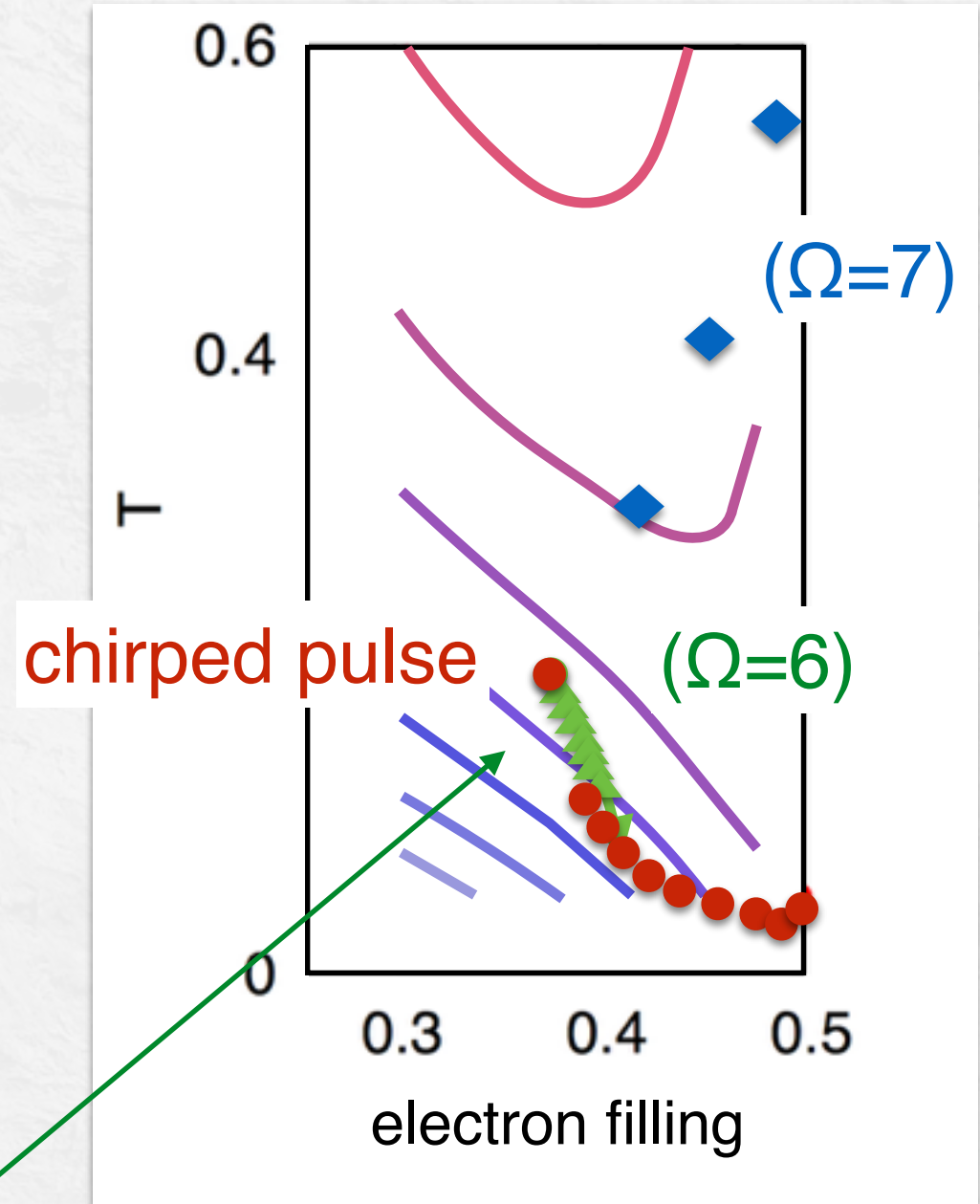
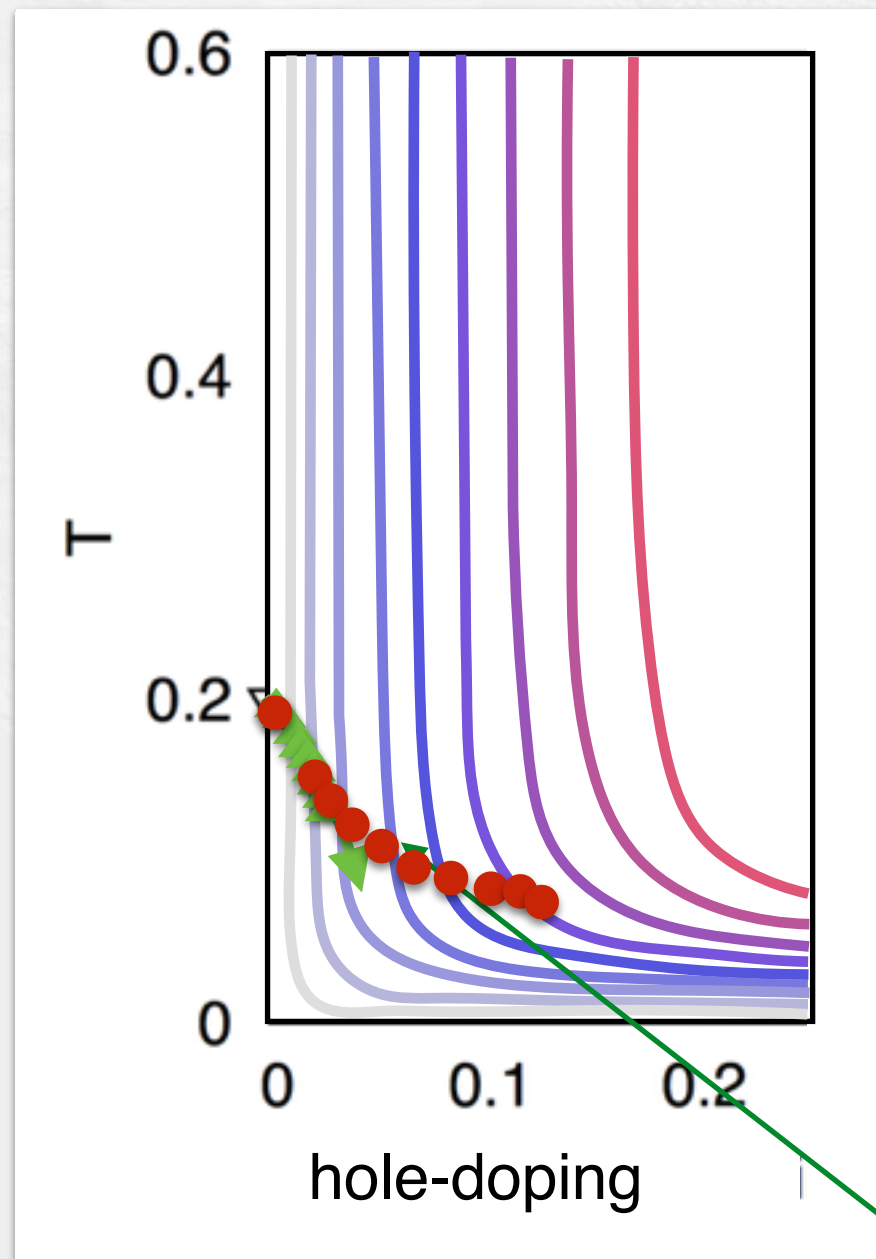
Charge transfer at constant entropy?



Effective Temperature after real (dynamic) charge transfer ($\Omega=6$)

Entropic cooling

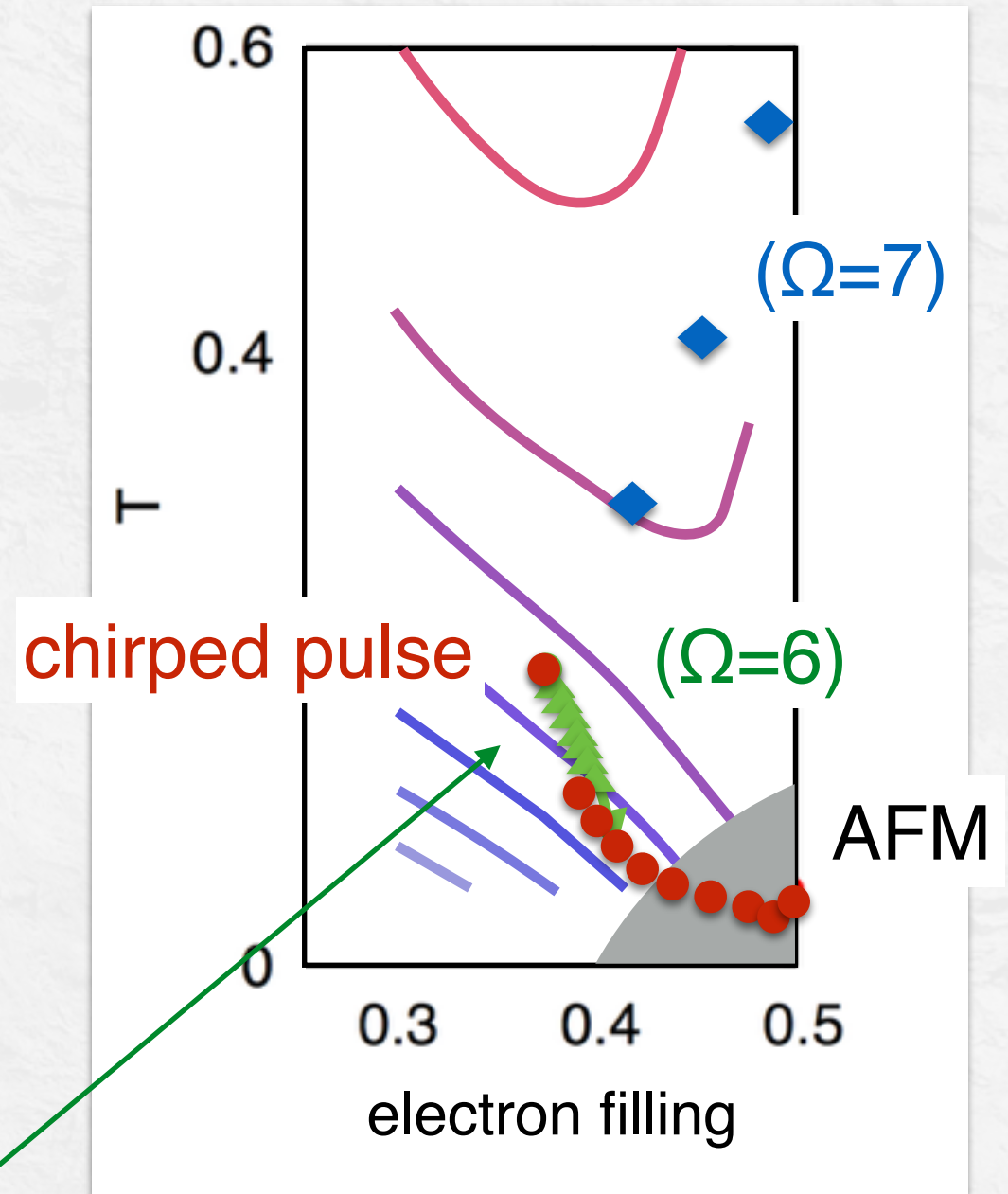
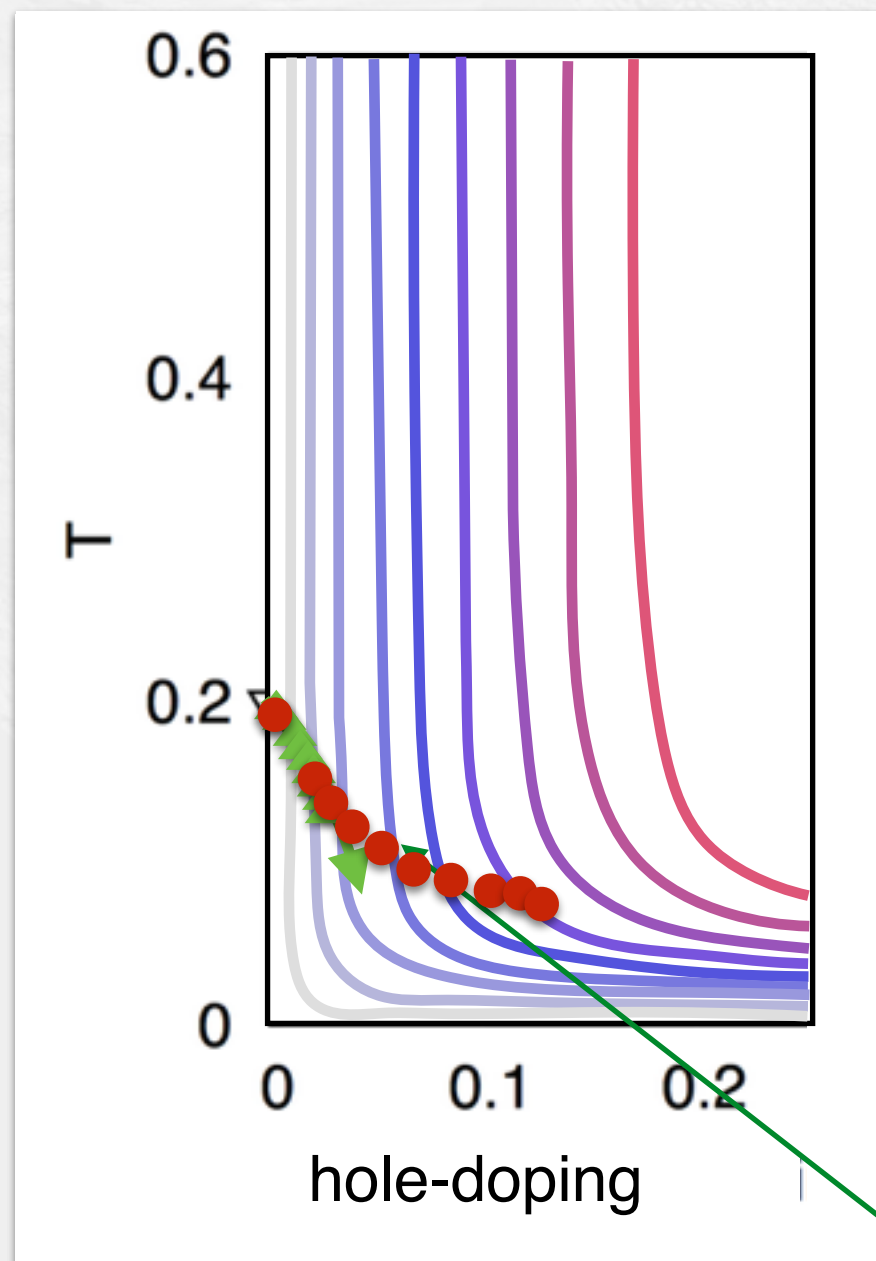
Charge transfer at constant entropy?



Effective Temperature after real (dynamic) charge transfer ($\Omega=6$)

Entropic cooling

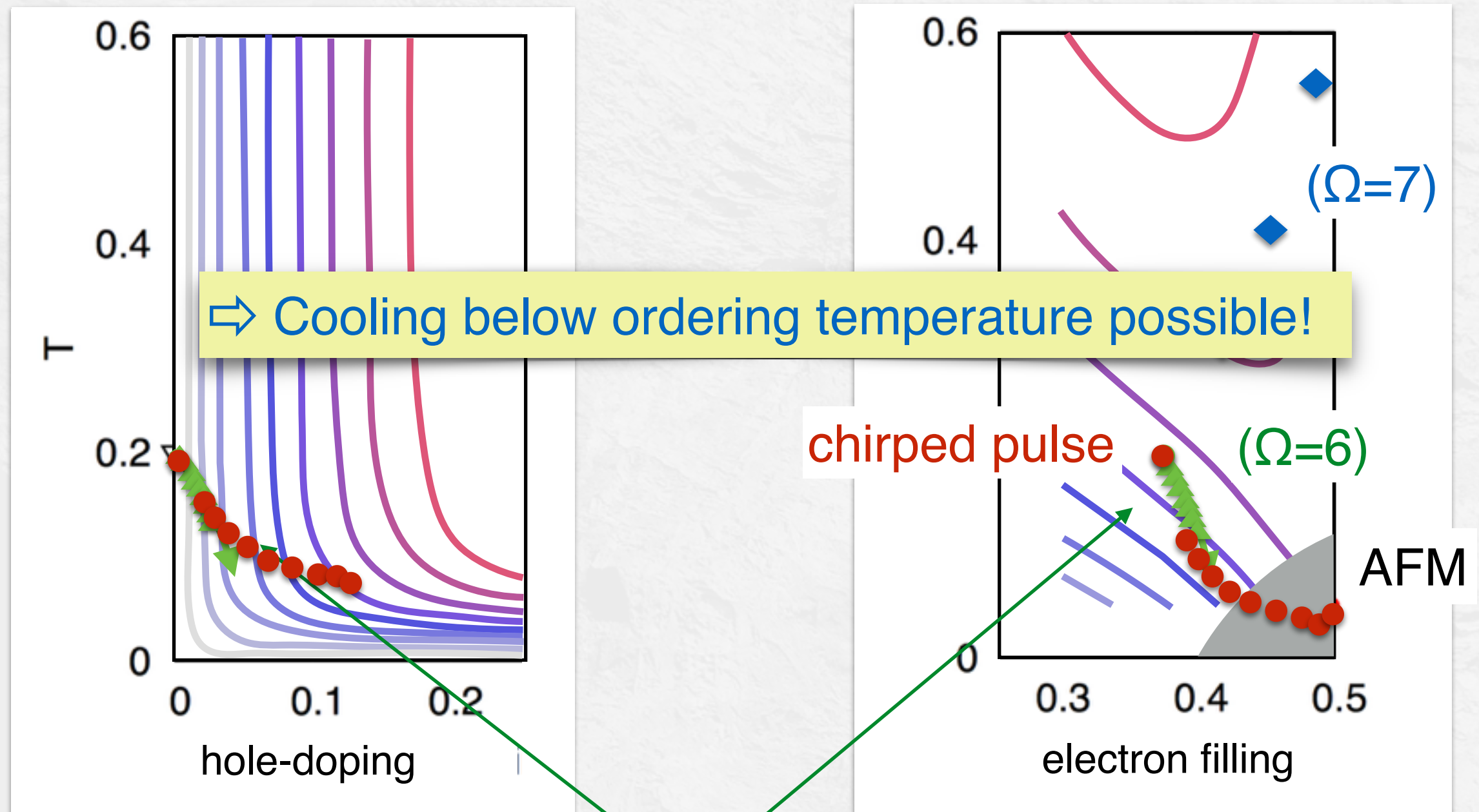
Charge transfer at constant entropy?



Effective Temperature after real (dynamic) charge transfer ($\Omega=6$)

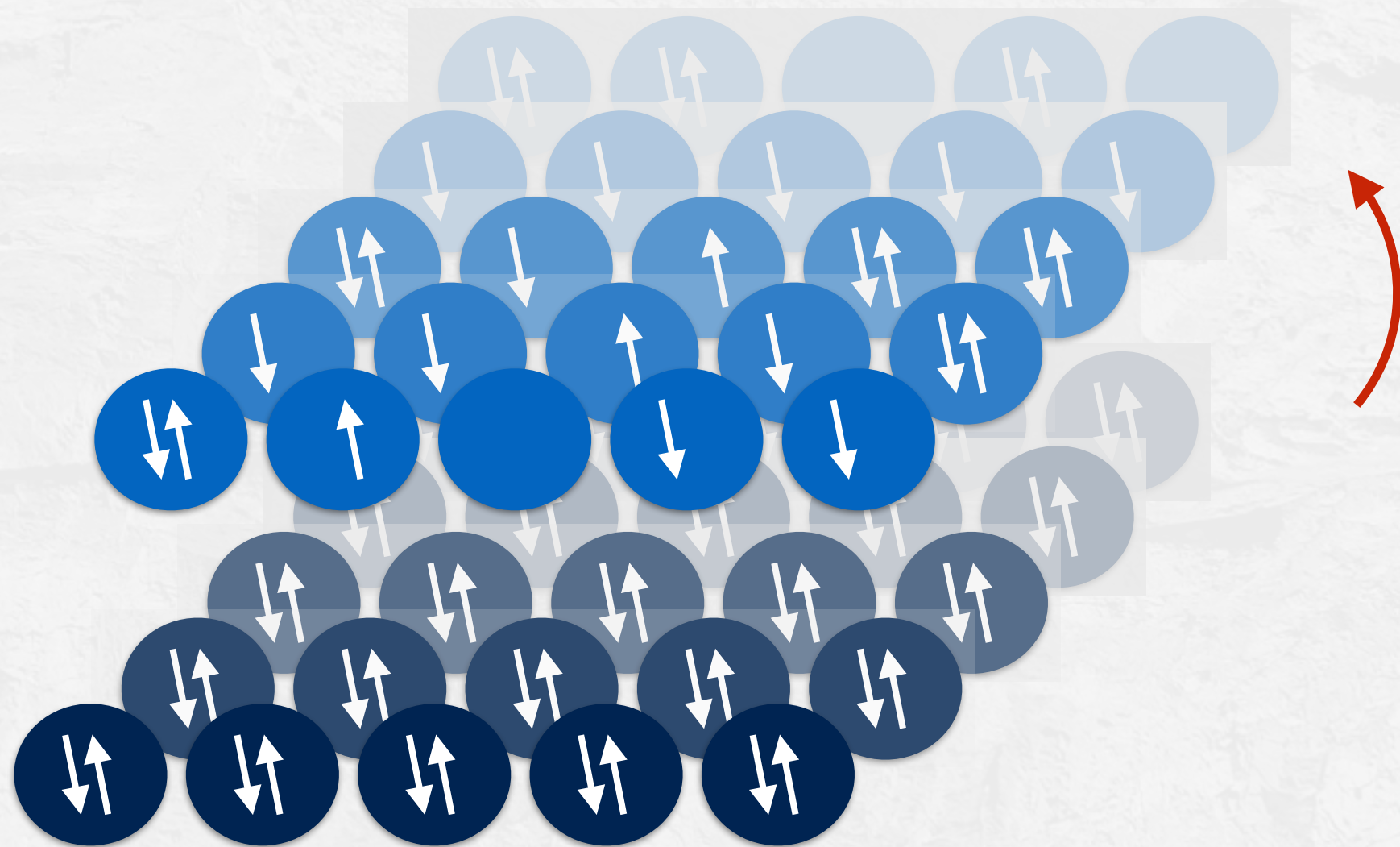
Entropic cooling

Charge transfer at constant entropy?



Entropic cooling

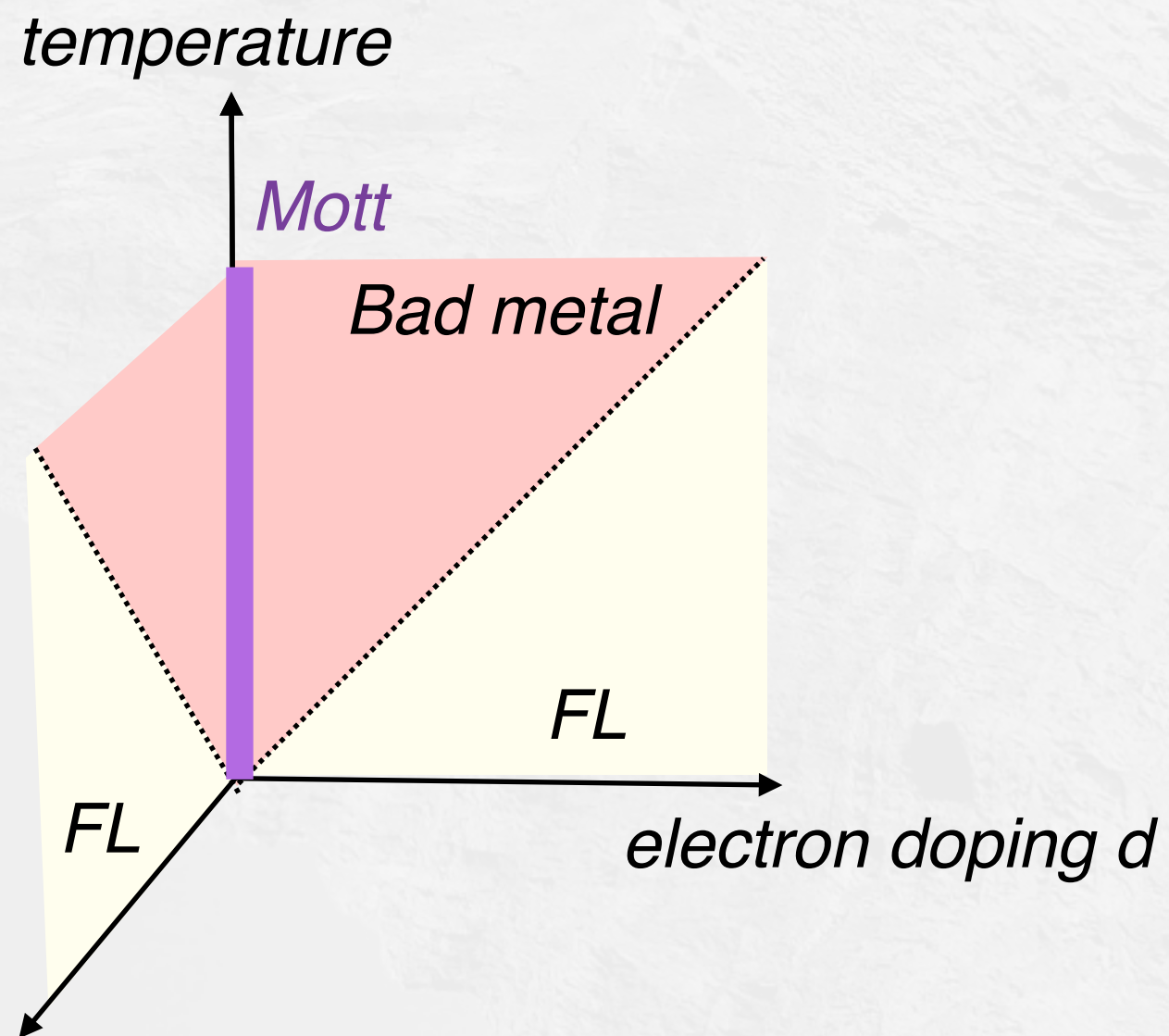
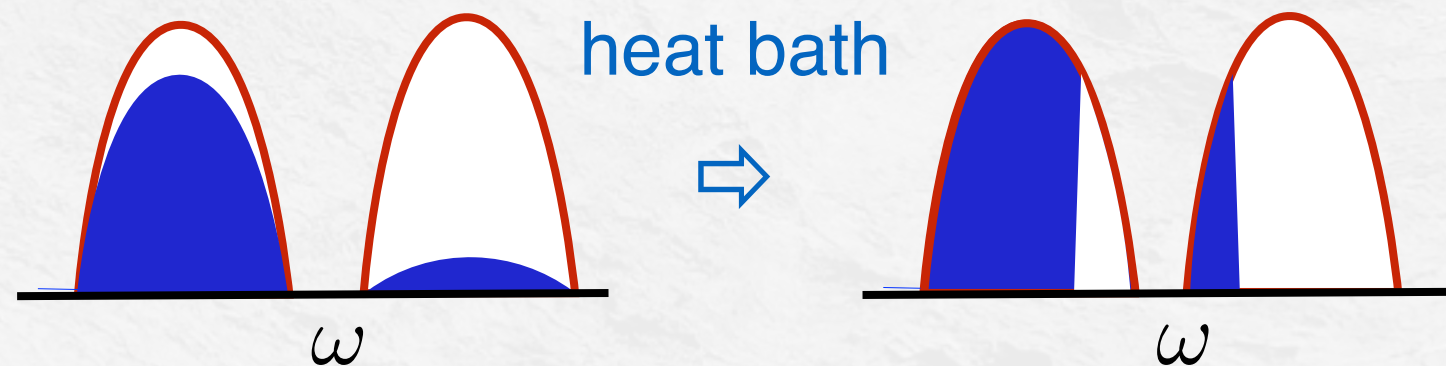
Realization as cooling protocol in cold atoms?



Theoretical “device” to prepare cold photo-doped states

Cooling after photo-doping?

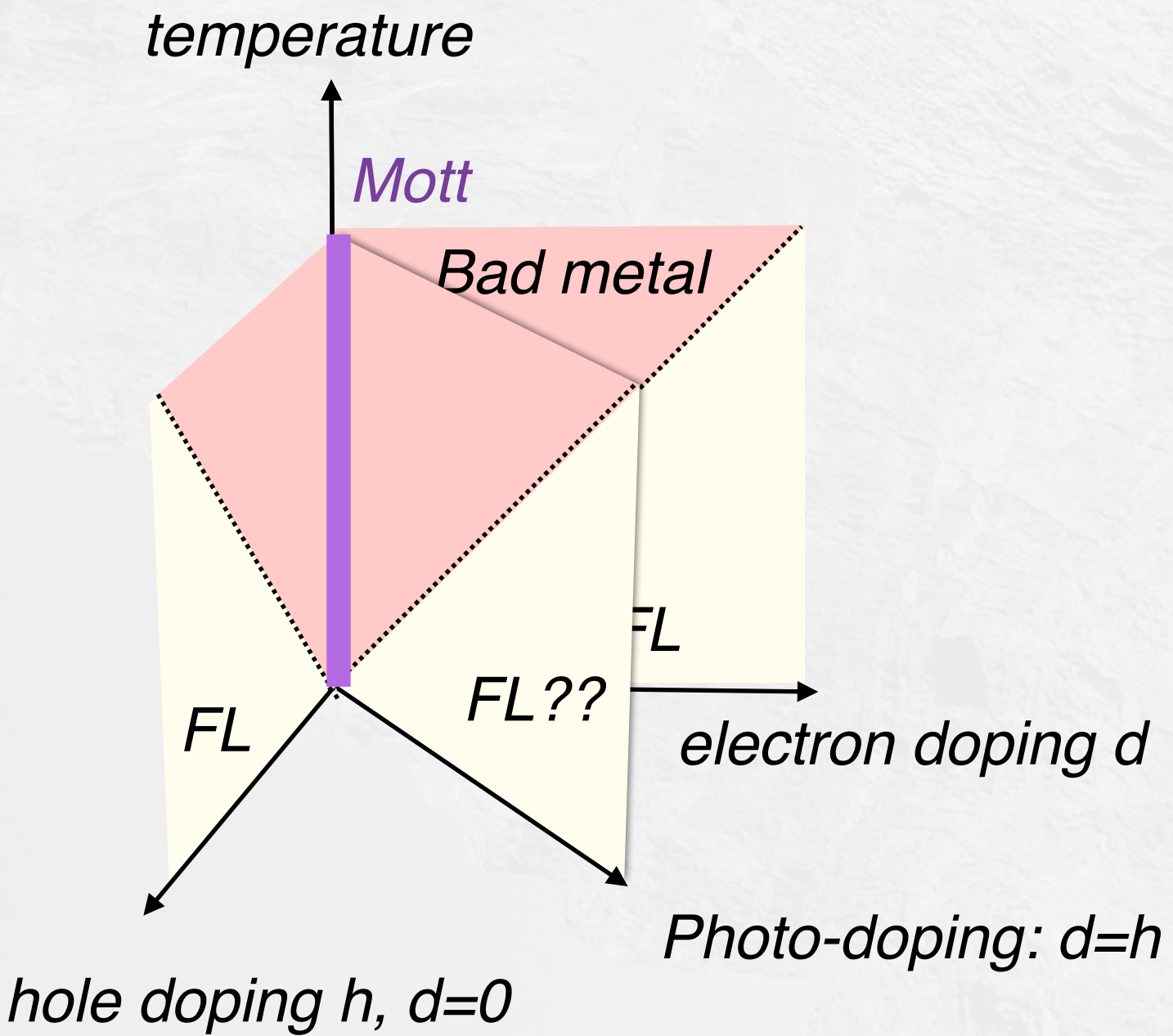
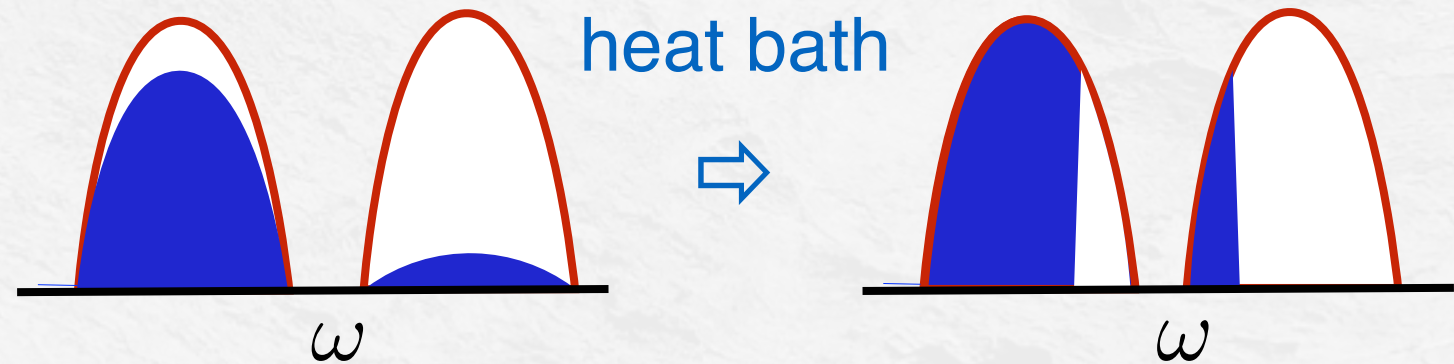
Cooling after photo-doping?



hole doping h , $d=0$

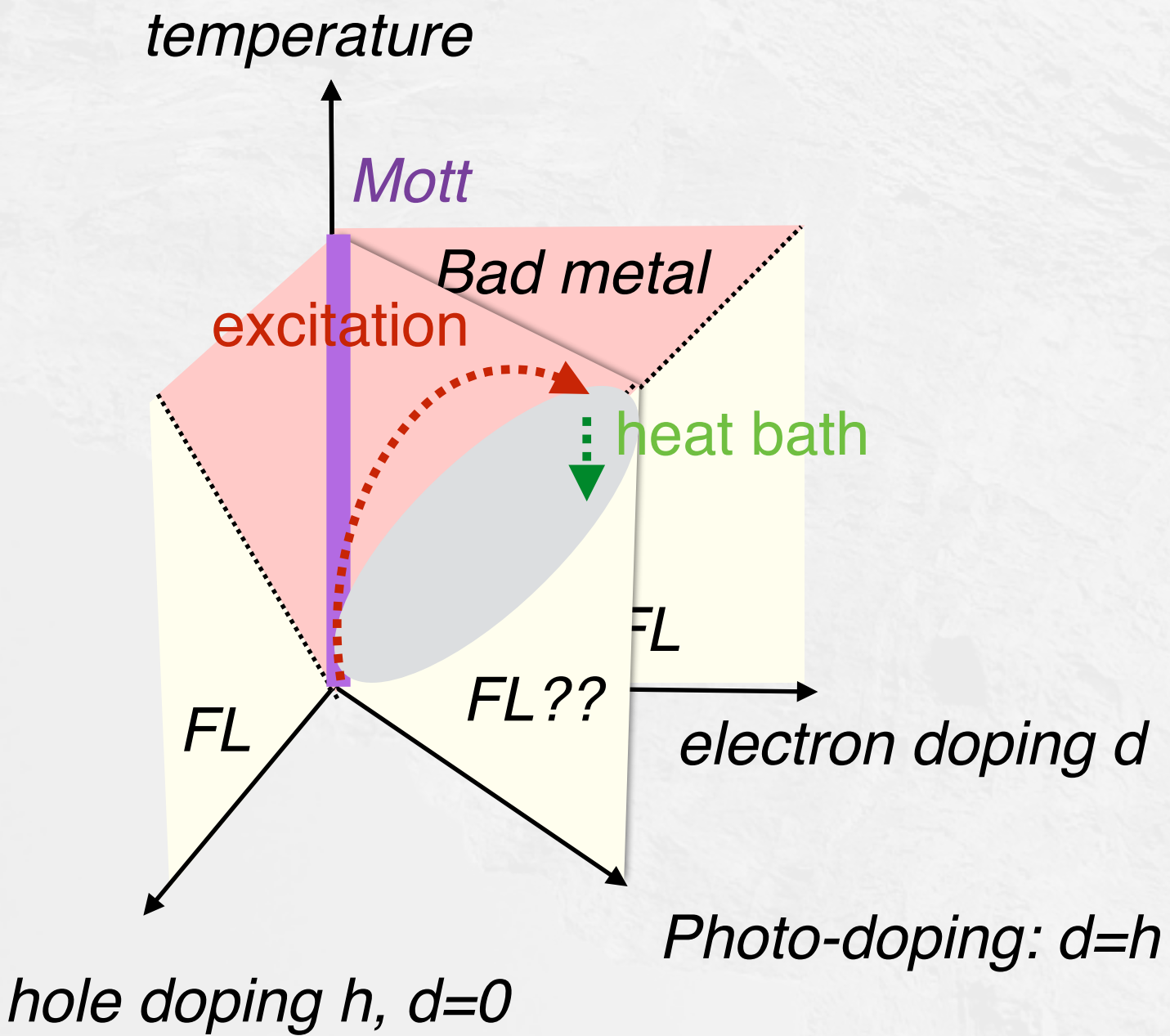
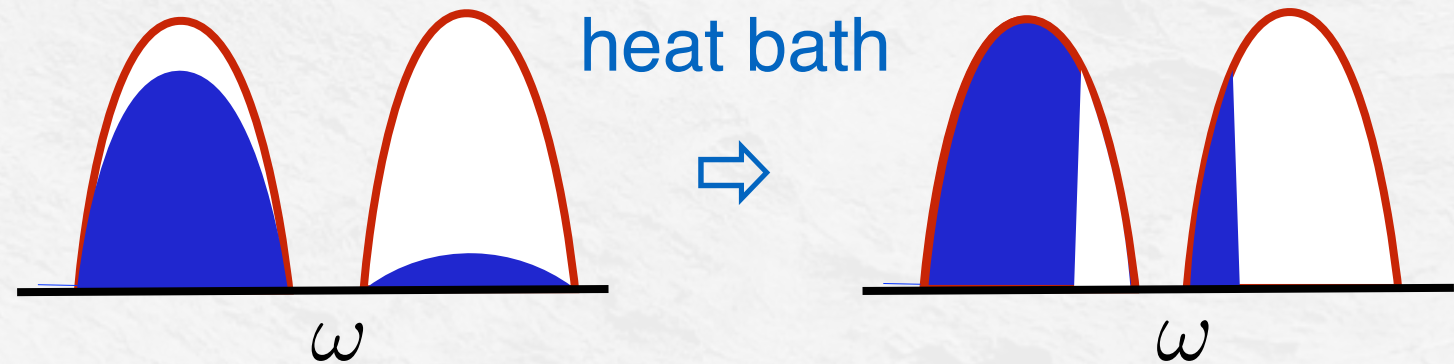
Cooling after photo-doping?

Cooling after photo-doping?



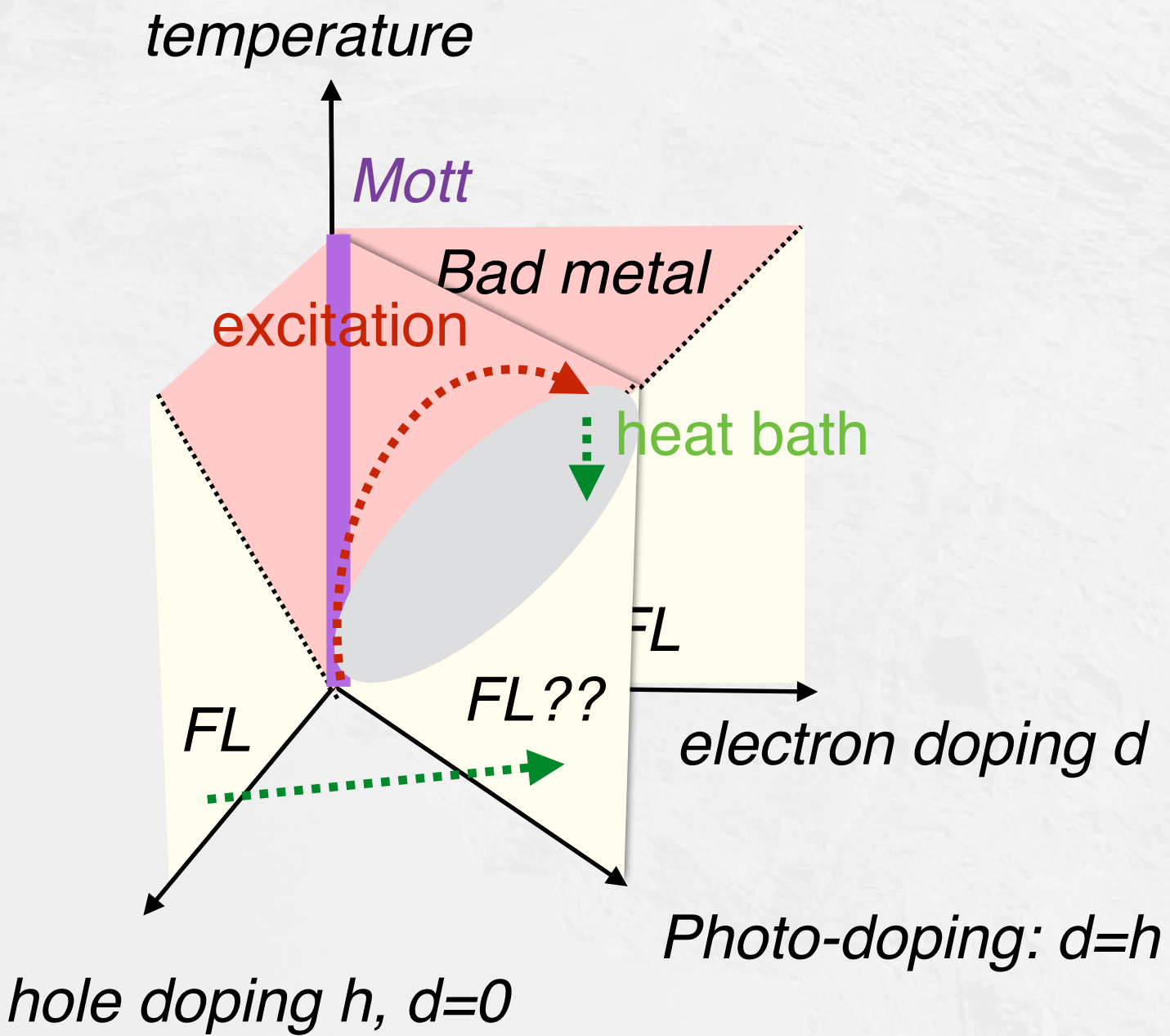
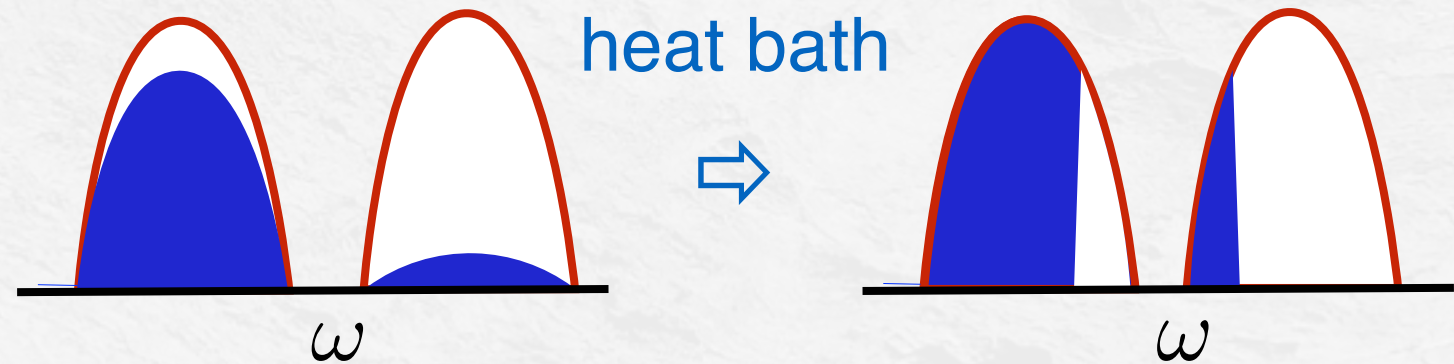
Cooling after photo-doping?

Cooling after photo-doping?



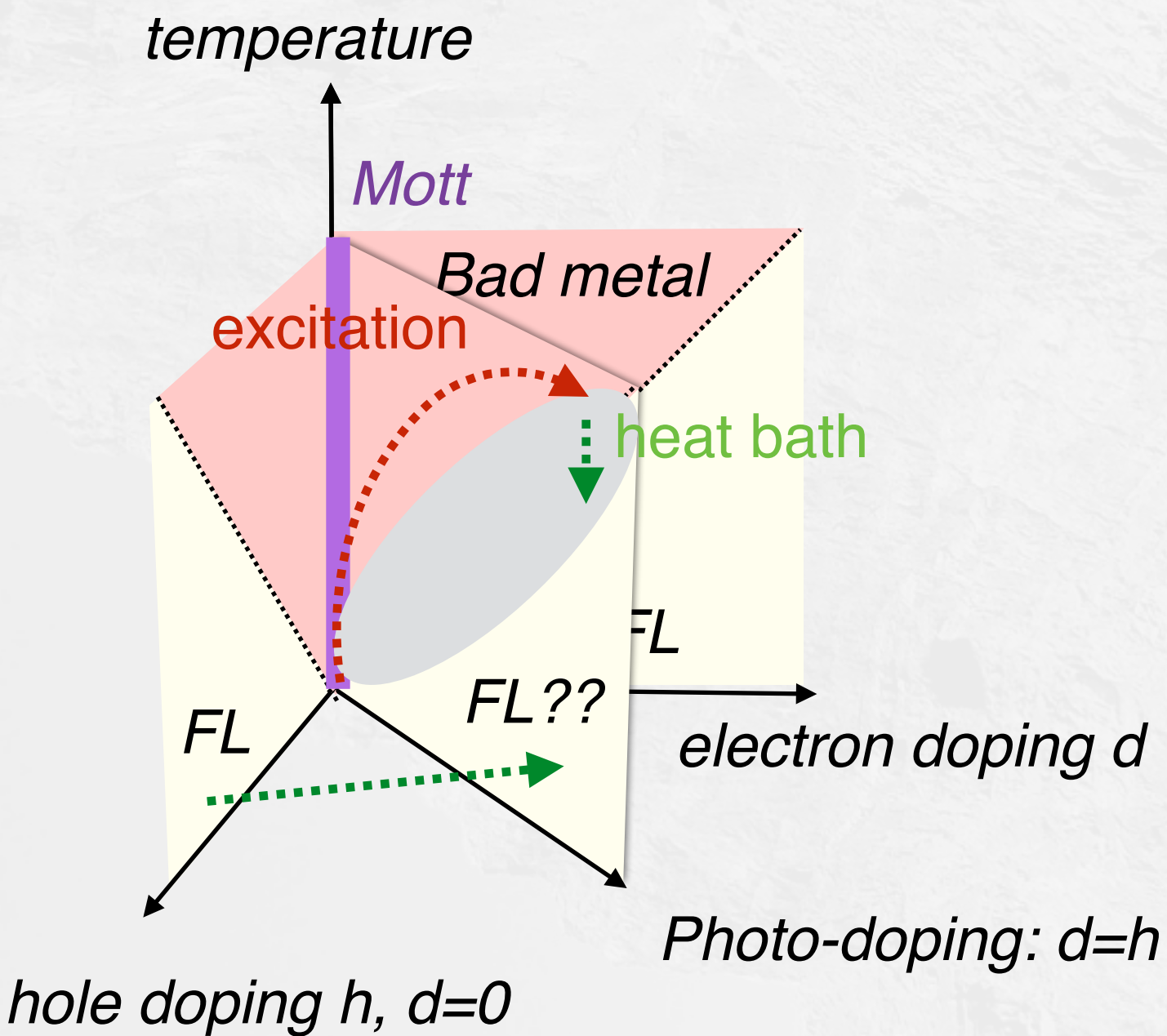
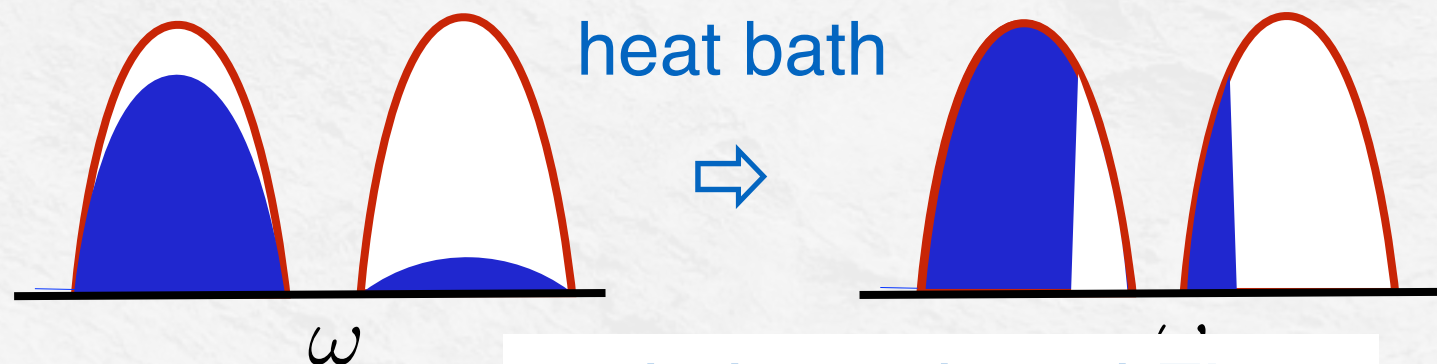
Cooling after photo-doping?

Cooling after photo-doping?

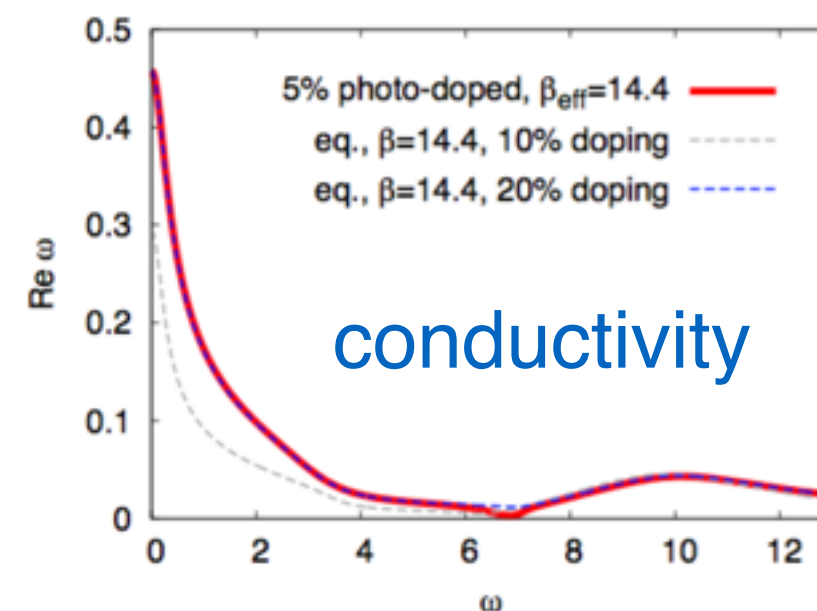
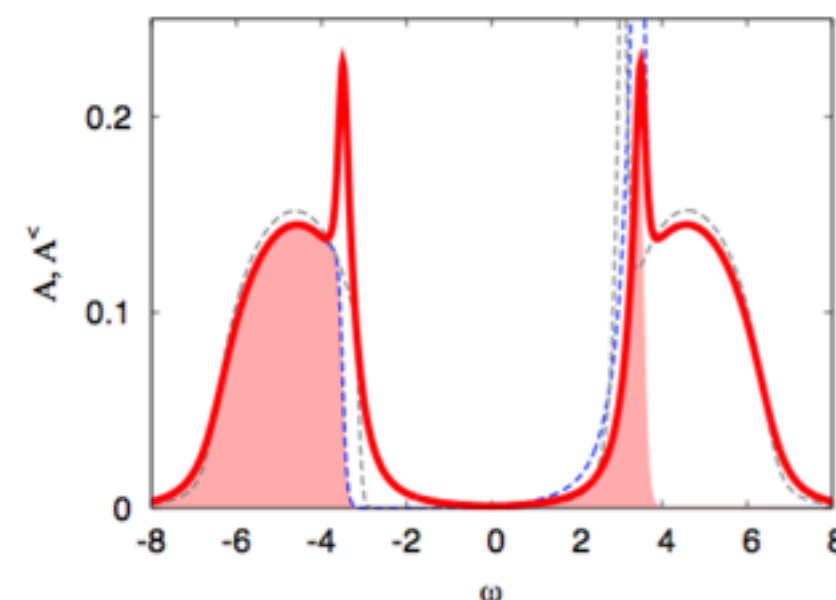


Cooling after photo-doping?

Cooling after photo-doping?



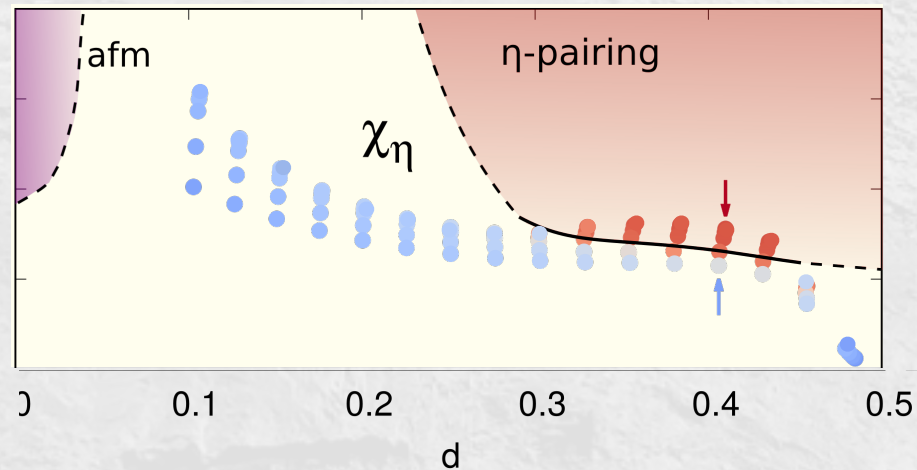
cold photo-doped FL



Summary & Acknowledgements

1) Phase diagram of photo-doped HM:

Jiajun Li, Denis Golez, Philipp Werner,
Martin Eckstein, arXiv:1908.08693



2) Cooling by photo-doping

Philipp Werner, Martin Eckstein,
Markus Müller, Gil Refael, arXiv:1904.00822