Friedrich-Alexander-Universität
Naturwissenschaftliche Fakultät

FAU

Inhomogeneous disordering
at a photoinduced charge density wave transition

Antonio Picano, College de France, Paris

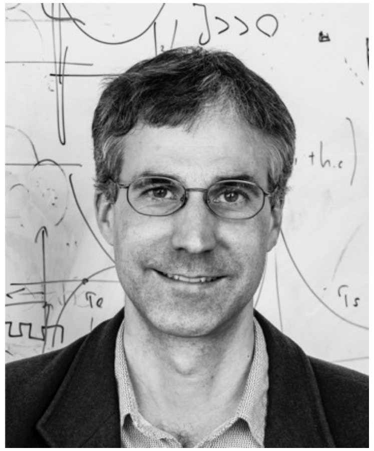
IMPACT 2024, Cargese, 27th August 2024

Acknowledgments

Stochastic semiclassical theory for electron-phonon interaction

Martin Eckstein
University of Hamburg

Francesco Grandi
RWTH Aachen University

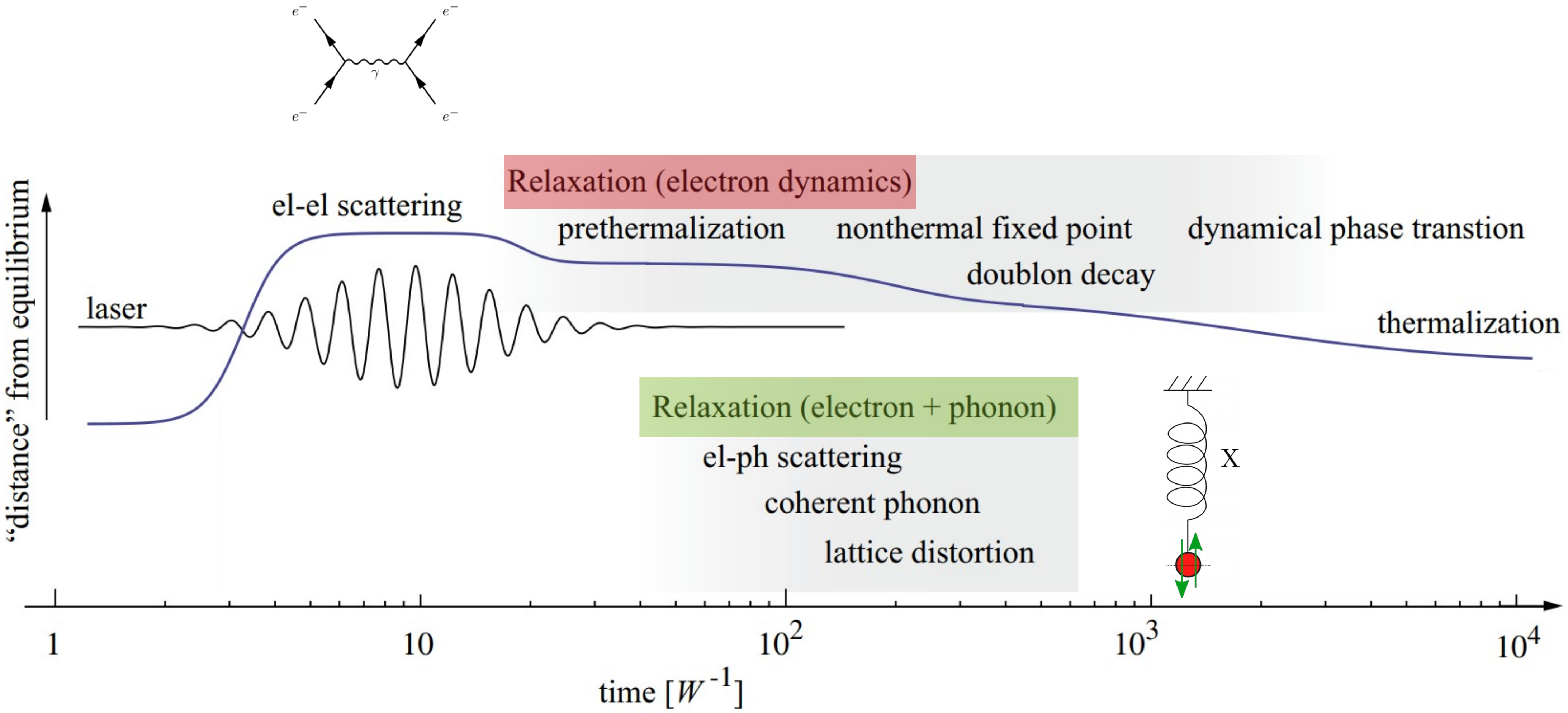


Philipp Werner
University of Fribourg

Marco Schiro
College de France

Non-equilibrium phenomena in condensed matter

The wide range of relevant time scales in the relaxation dynamics



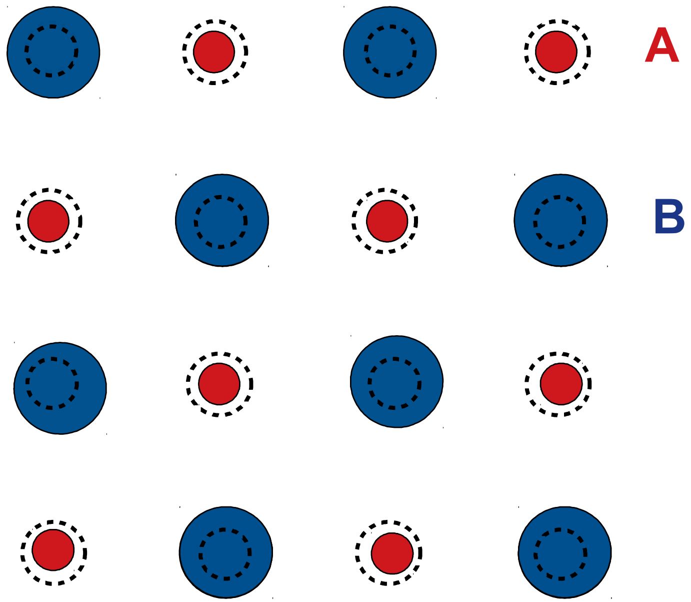
Re-adapted from Aoki, H. et al.: Rev. Mod. Phys. 86, 799 (2014)

Charge density wave order (CDW)

The representation taken into account in our model

A CDW is characterized by : 1) CDW modulation; 2) Periodic lattice distortion; 3) Energy gap

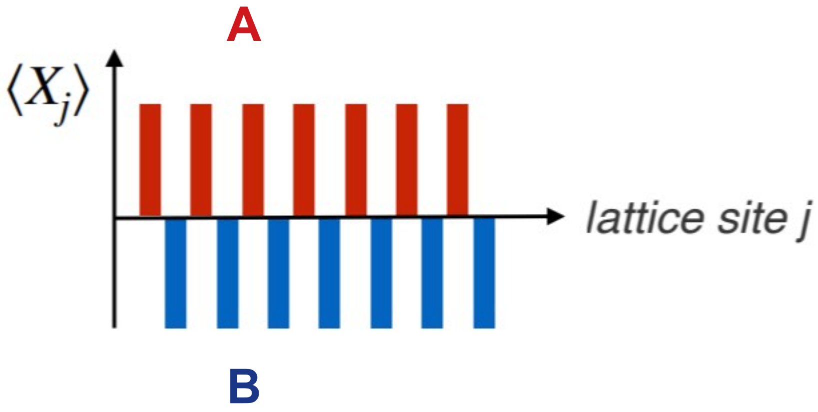
Electron density n



X

Phonon displacement

X

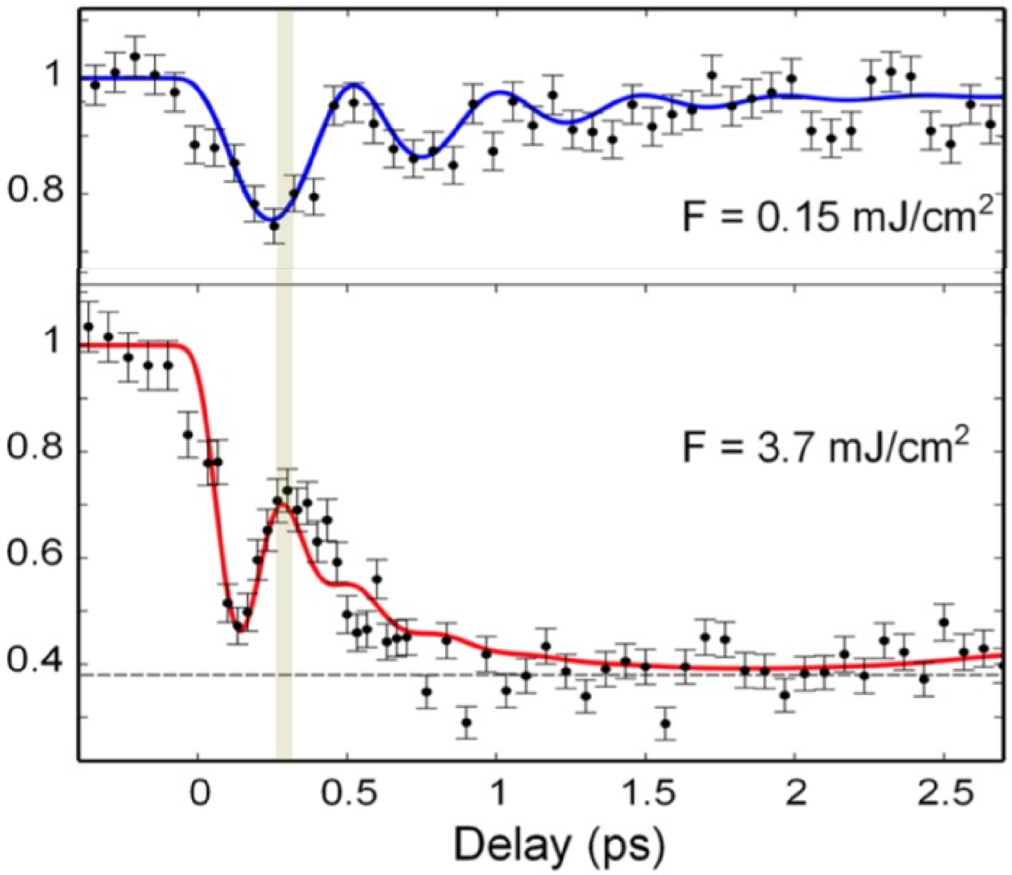


$$H_{el-ph} \propto X(n-1)$$

Coherent dynamics in $K_{0.3}MoO_3$

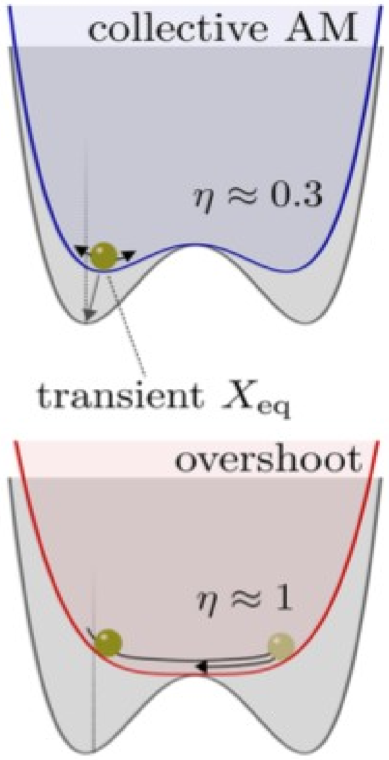
Well-described by time-dependent Ginzburg-Landau theory

CDW intensity (trXRD)



Fluence

Time-dependent GL theory

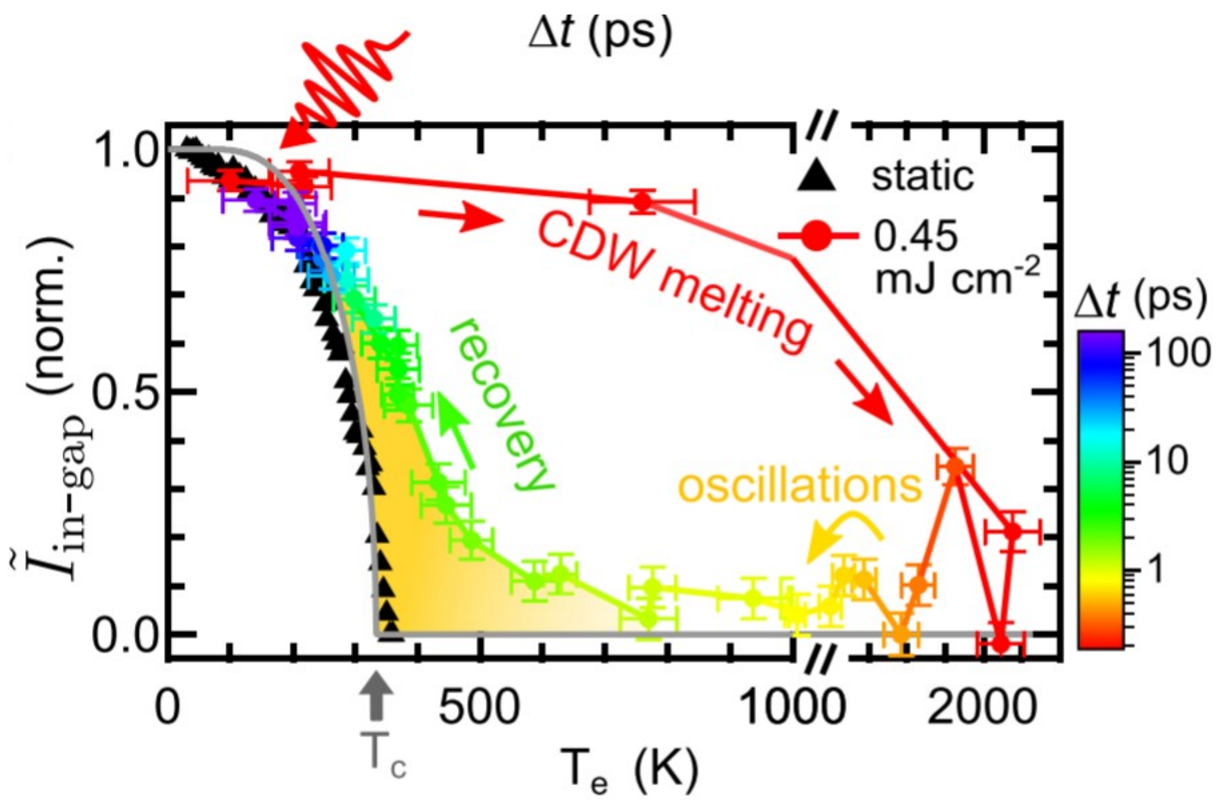


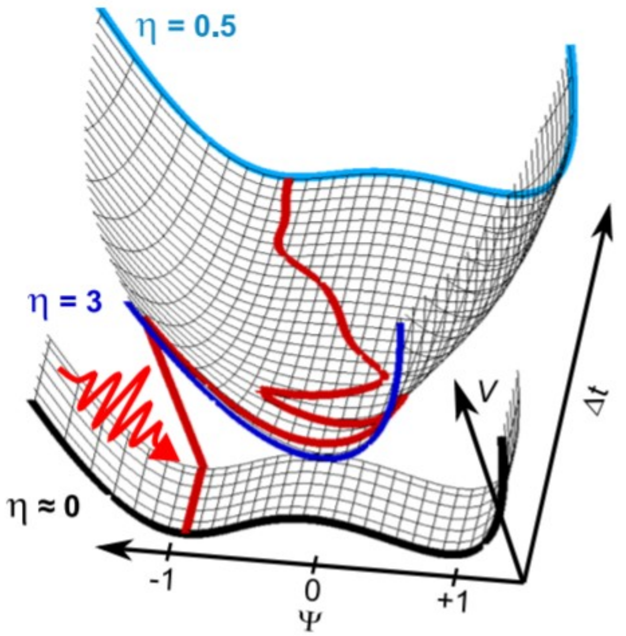
$$\omega_{CD}^{-2}\ddot{X} + \Gamma\dot{X} = -\frac{d}{dX}F[X, T_{eff}(t)]$$

Huber, T. et al.: PRL 113, 026401 (2014)

Non-thermal free-energy potentials

CDW dynamics in $TbTe_3$





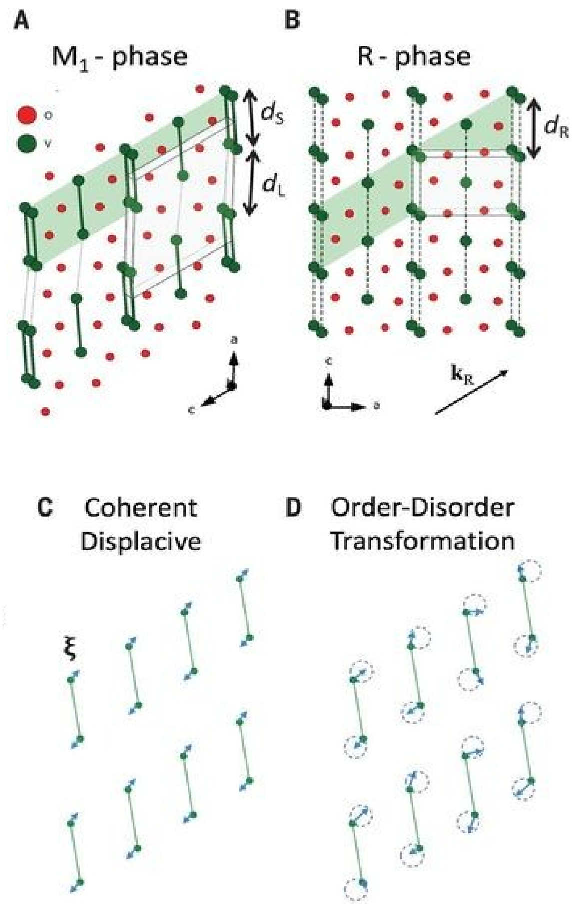
It seems that **in some cases the electrons <u>cannot</u> be described simply by an** effective temperature...

Maklar, J. et al.: Nature Comm. 12, 2499 (2021)

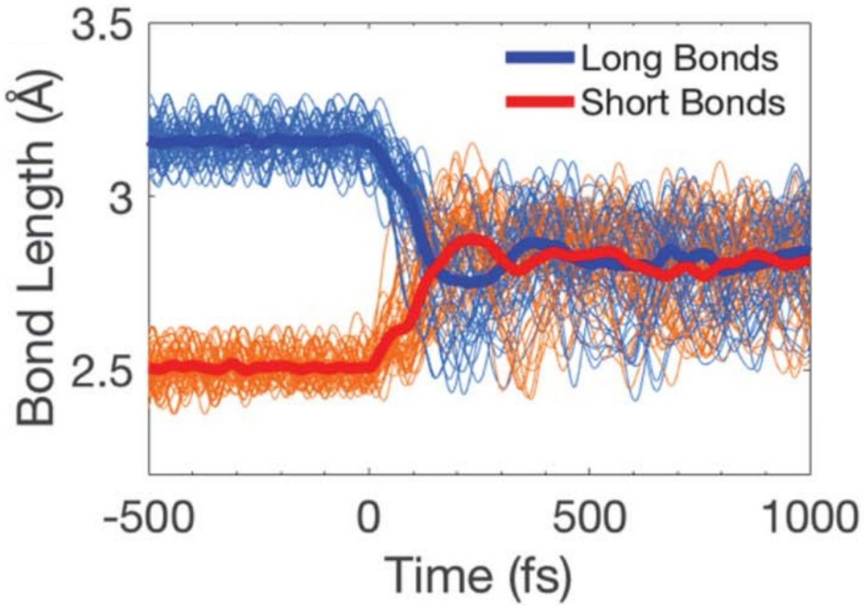
Coherent dynamics or ultrafast disordering?

The case for VO_2

Insulator → Metal: coherent or order-disorder?



Large-amplitude uncorrelated motion



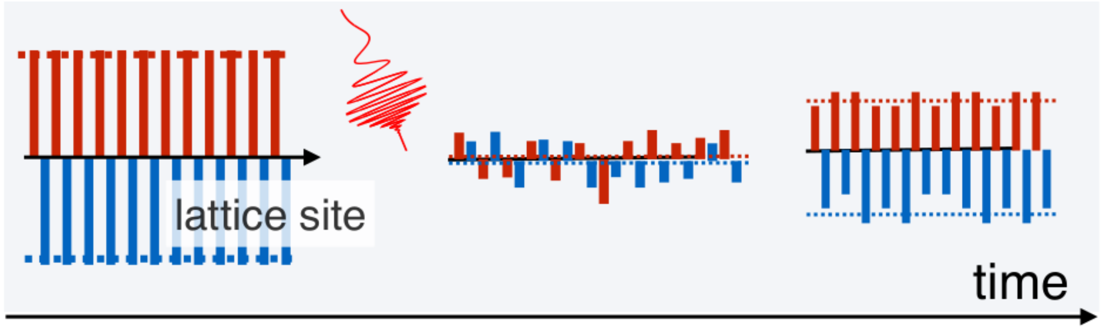
Wall, S. et al.: Science 362, 572 (2018)

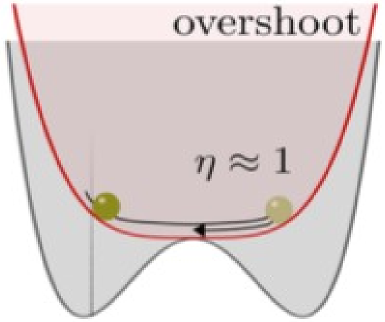
Ultrafast disordering?

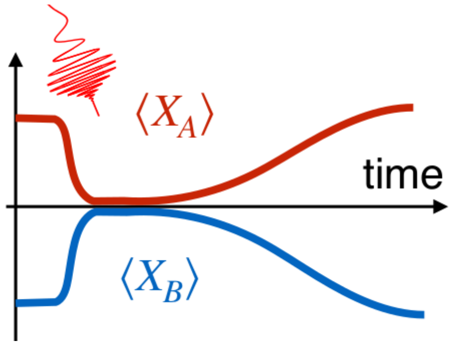
Conventional scenario or ultrafast disordering?

1) Conventianal scenario

Well described by **td-GL** + small Gaussian fluctuations







$$\omega_{CD}^{-2}\ddot{X} + \Gamma\dot{X} = -\frac{d}{dX}F[X, T_{eff}(t)]$$

Huber, T. et al.: PRL 113, 026401 (2014)

Ultrafast disordering?

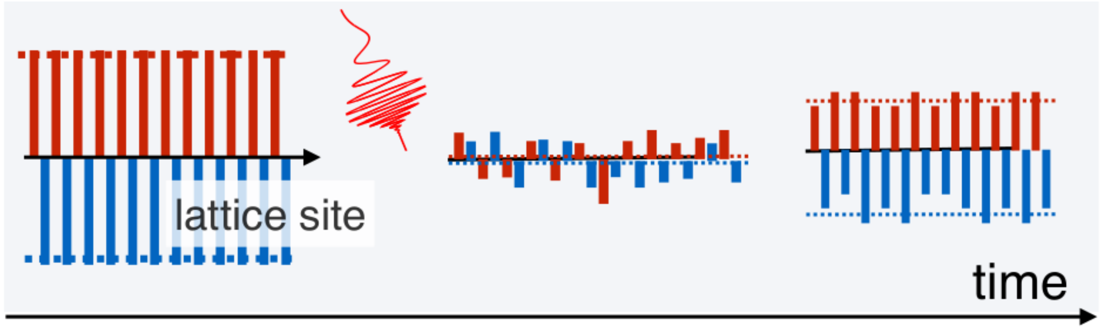
Conventional scenario or ultrafast disordering?

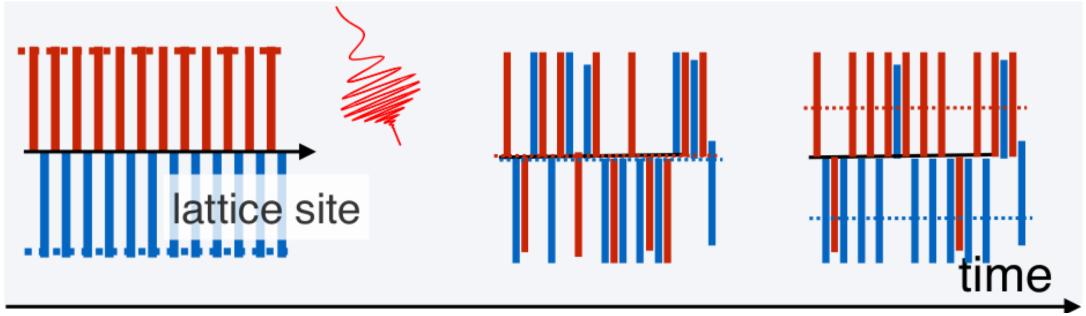
1) Conventianal scenario

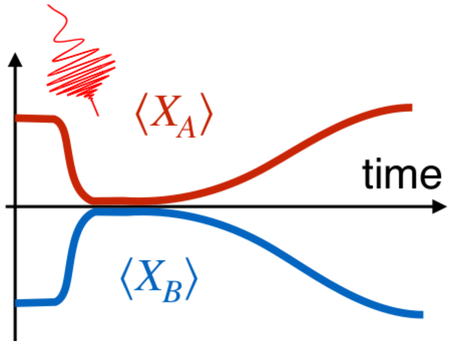
Well described by **td-GL** + small Gaussian fluctuations

2) Alternative scenario: **Ultrafast disordering**

Average order parameter not representative for individual sites







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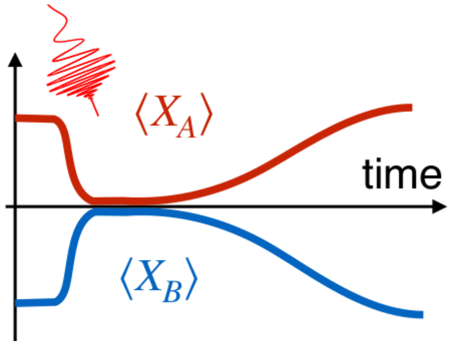


Ultrafast inhomogeneous disordering

Experimental indications

- Indications seen for VO_2 Wall et al., Science 362, 572 (2018)
  $La_{0.5}Sr_{1.5}MnO_4$ Salinas et al., Nature Comm. 13, 238 (2022)

- Indistinguishable in terms of average, but disorder may have profound effect on dynamics: Slow dynamics, metastable states, …

- Effects on the electronic structure: Incoherent spectral weight, …

Theoretical challenge: Joint evolution of inhomogeneous electronic structure and inhomogeneous order parameter

Theory of ultrafast disordering at a CDW transition
(a microscopic perspective)

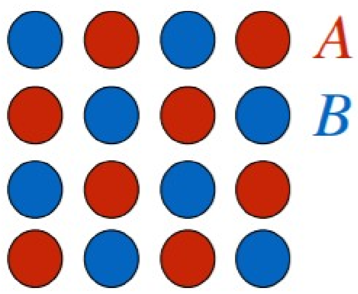
The Holstein model

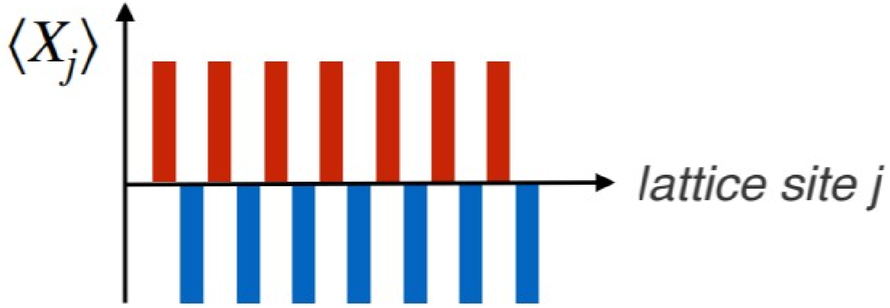
The lattice order parameter

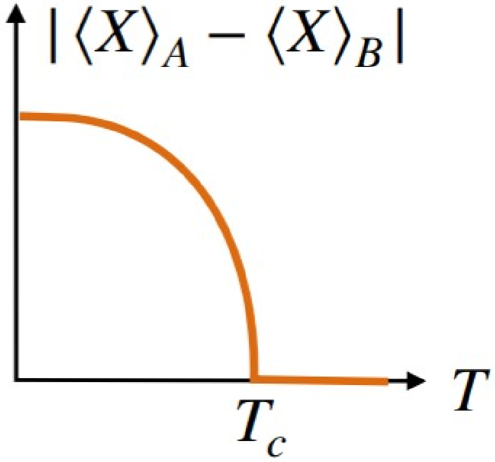
$$H = -J \sum_{\langle i,j \rangle,\sigma} c^\dagger_{i,\sigma} c_{j,\sigma} + \sum_j \sqrt{2} g (n_j - 1) X_j + \sum_j \frac{\Omega}{2} (X_j^2 + P_j^2)$$

Electron-phonon interaction
(responsible for CDW order)

⇨ CDW order on bipartite lattice





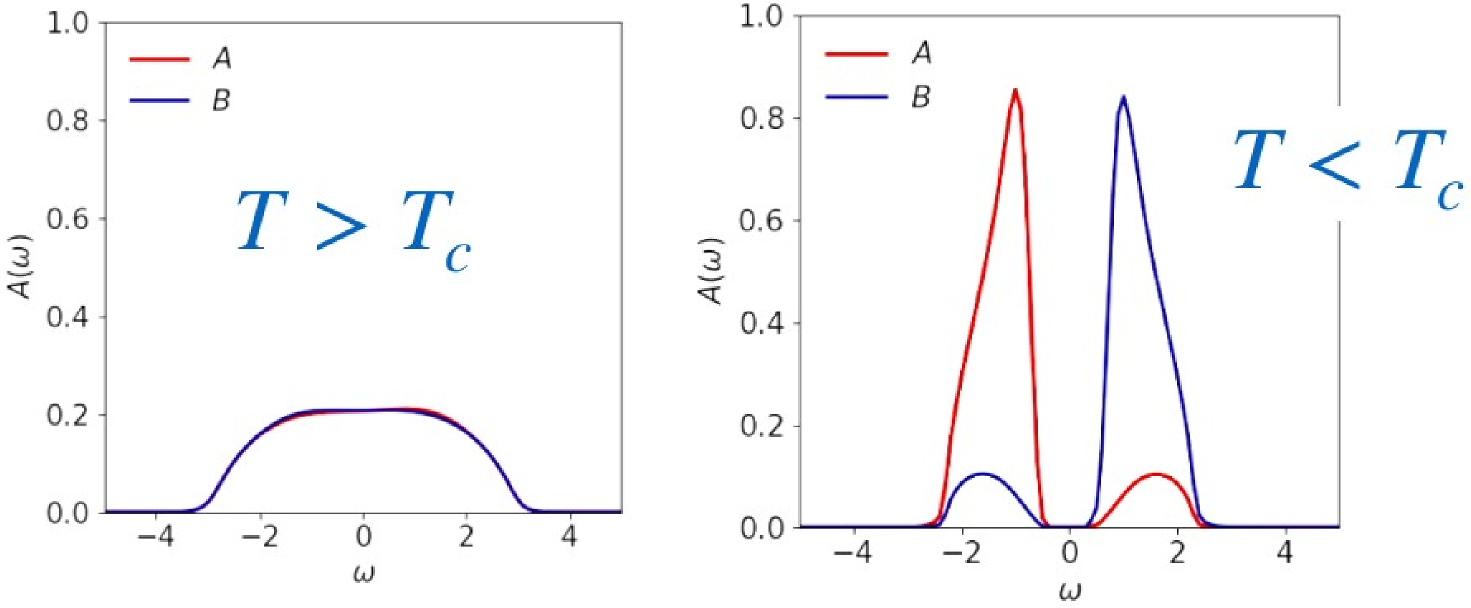


The Holstein model

The opening of the gap

$$H = -J \sum_{\langle i,j \rangle,\sigma} c^\dagger_{i,\sigma} c_{j,\sigma} + \sum_j \sqrt{2} g (n_j - 1) X_j + \sum_j \frac{\Omega}{2} (X_j^2 + P_j^2)$$

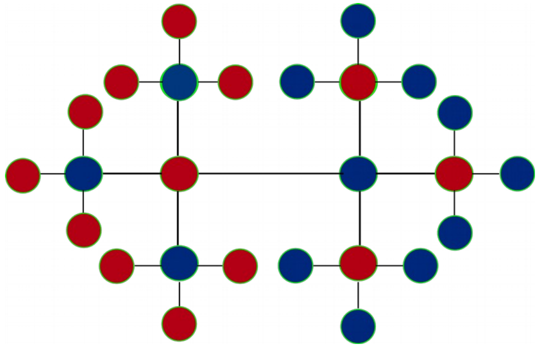
Electron-phonon interaction
(responsible for CDW order)



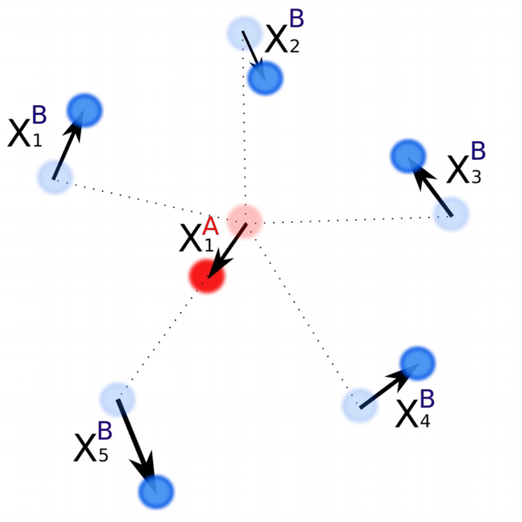
Statistical dynamical mean-field theory

A generalization of the approach to non-equilibrium

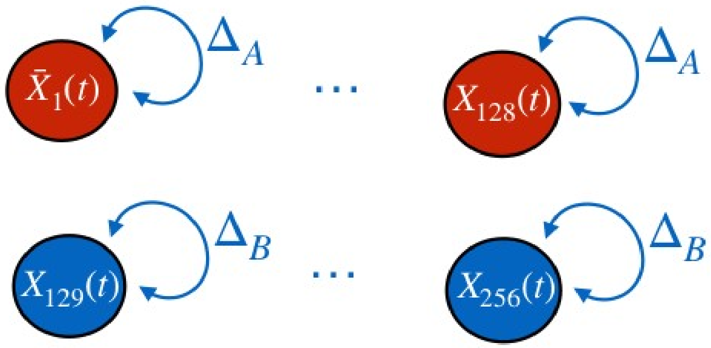
Bipartite Bethe lattice



Each of the N sites has its own displacement X_j



One has to solve an impurity problem for each of the N sites



$$\Delta_{1,A} = |J|^2 \langle G_j \rangle_{j \in B}$$
$$\Delta_{1,B} = |J|^2 \langle G_j \rangle_{j \in A}$$

$$N \to \infty$$

$$\Delta_A = |J|^2 \langle G \rangle_{j \in B}$$
$$\Delta_B = |J|^2 \langle G \rangle_{j \in A}$$

Phonons: Stochastic semiclassical theory

Equations of motion for the phonons

$$\ddot{X}_j = -\Omega^2 X_j - g\sqrt{2\Omega}(\langle n_j \rangle_{\text{cl}} - 1) - \gamma_j \Omega \dot{X}_j + \sqrt{\Omega}\xi_j$$

Elastic force — Mean-field force on phonons — Damping and noise from electrons

The fluctuations of the electron density, $\chi_{j,\text{cl}}(t,t') = -i\langle T_{\mathcal{C}} n_j(t) n_j(t') \rangle$, affect the phonon dynamics:

$$\gamma_j(t) = -2g^2 \partial_\omega \Im \chi^R_{j,\text{cl}}(t,\omega)|_{\omega=0}$$

Damping

$$\langle \xi_j(t) \rangle = 0$$

$$\langle \xi_j(t) \xi_{j'}(t') \rangle = K_j(t) \delta_{j,j'} \delta(t,t')$$

$$K_j(t) = -g^2 \Im \chi^K_{j,\text{cl}}(t,\omega)|_{\omega=0}$$

White **noise** approx.: $\tau_e \ll \frac{1}{\Omega}$

Picano, A., et al.: Phys. Rev. B 108, 035115 (2023)

A truly coupled electron-phonon dynamics

The Quantum Boltzmann equation (QBE) for the electronic problem

$$\begin{cases} \partial_t F_j(\omega,t) &= I_\omega[F_j(\omega,t), \mathcal{A}_j(\omega,t)] \\ \mathcal{A}_j(\omega,t) &= \mathcal{A}_\omega^{\mathrm{NESS}}[F_j(\omega,t)] \\ \ddot{X}_j(t) &= -\Omega_j^2 X_j(t) - g\sqrt{2\Omega}(\langle n_j(t)\rangle_{\mathrm{cl}} - 1) - \Omega\gamma_j \dot{X}_j(t) + \sqrt{\Omega}\xi_j(t) \end{cases}$$

Electronic distribution function

Electronic spectrum

} QBE

Phonon displacement

QBE for the electronic distribution function and the electronic spectrum

+

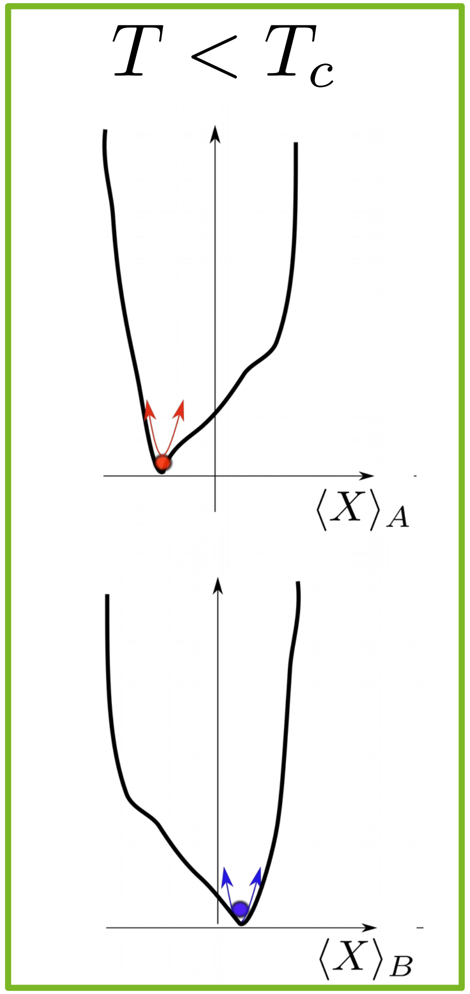
stochastic semiclassical equations for the phonon displacement

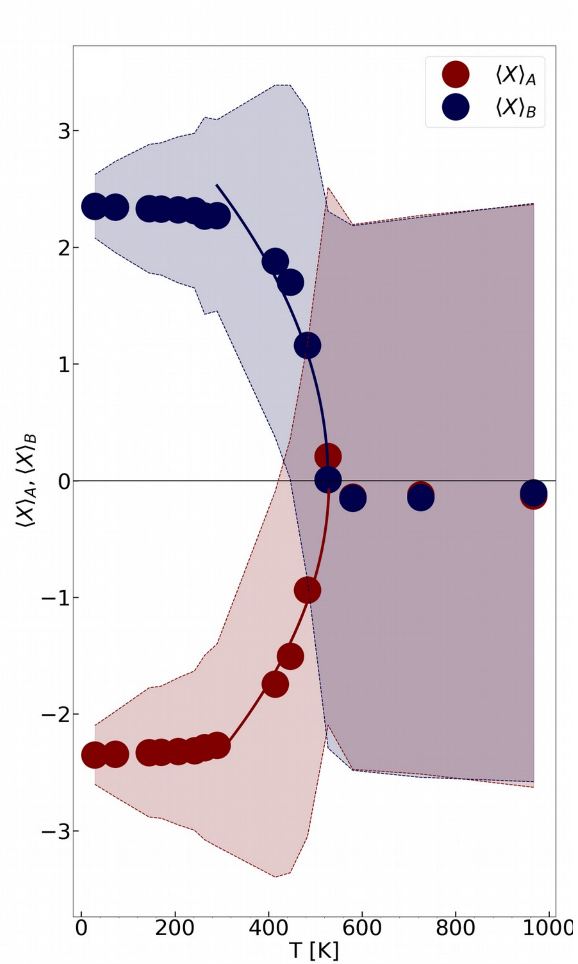
Picano, A. et al.: Phys. Rev. B 104, 085108 (2021)

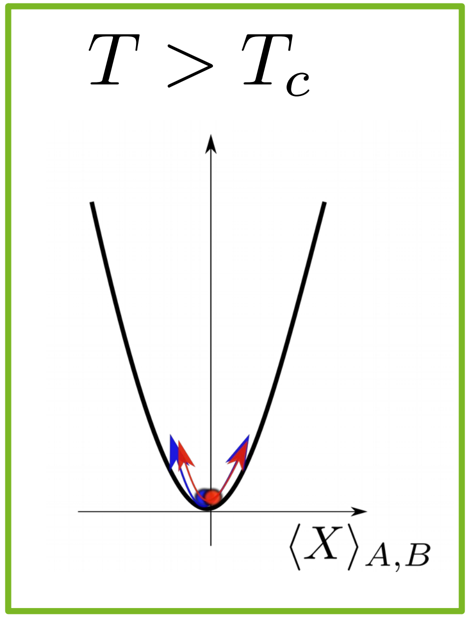
Picano, A., et al.: Phys. Rev. B 108, 035115 (2023)

Semiclassical solution: equilibrium

Second-order phase transition



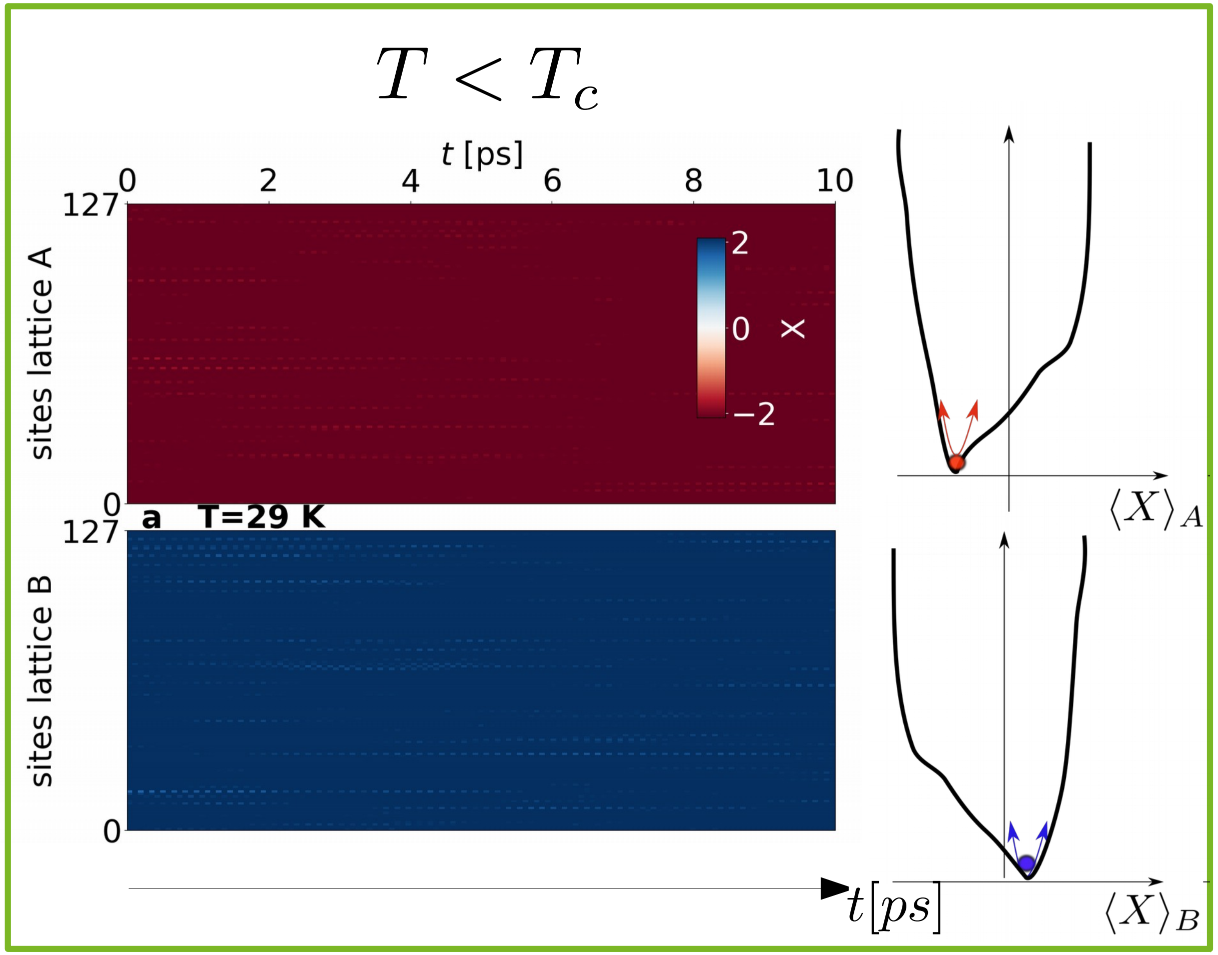


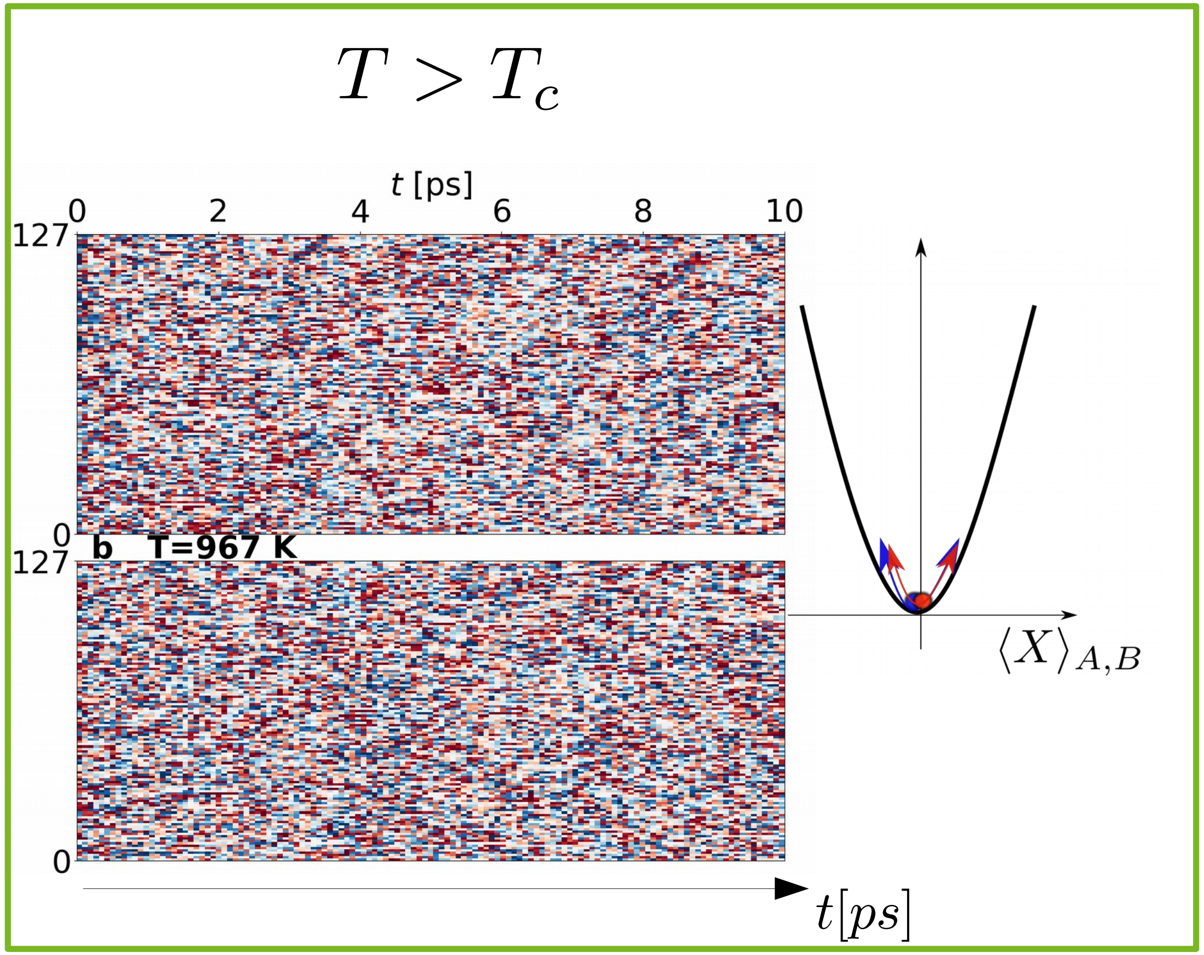


Picano, A. et al. : Phys. Rev. B 107, 245112 (2023)

Semiclassical solution: equilibrium

The dynamics of the phonon trajectories at equilibrium

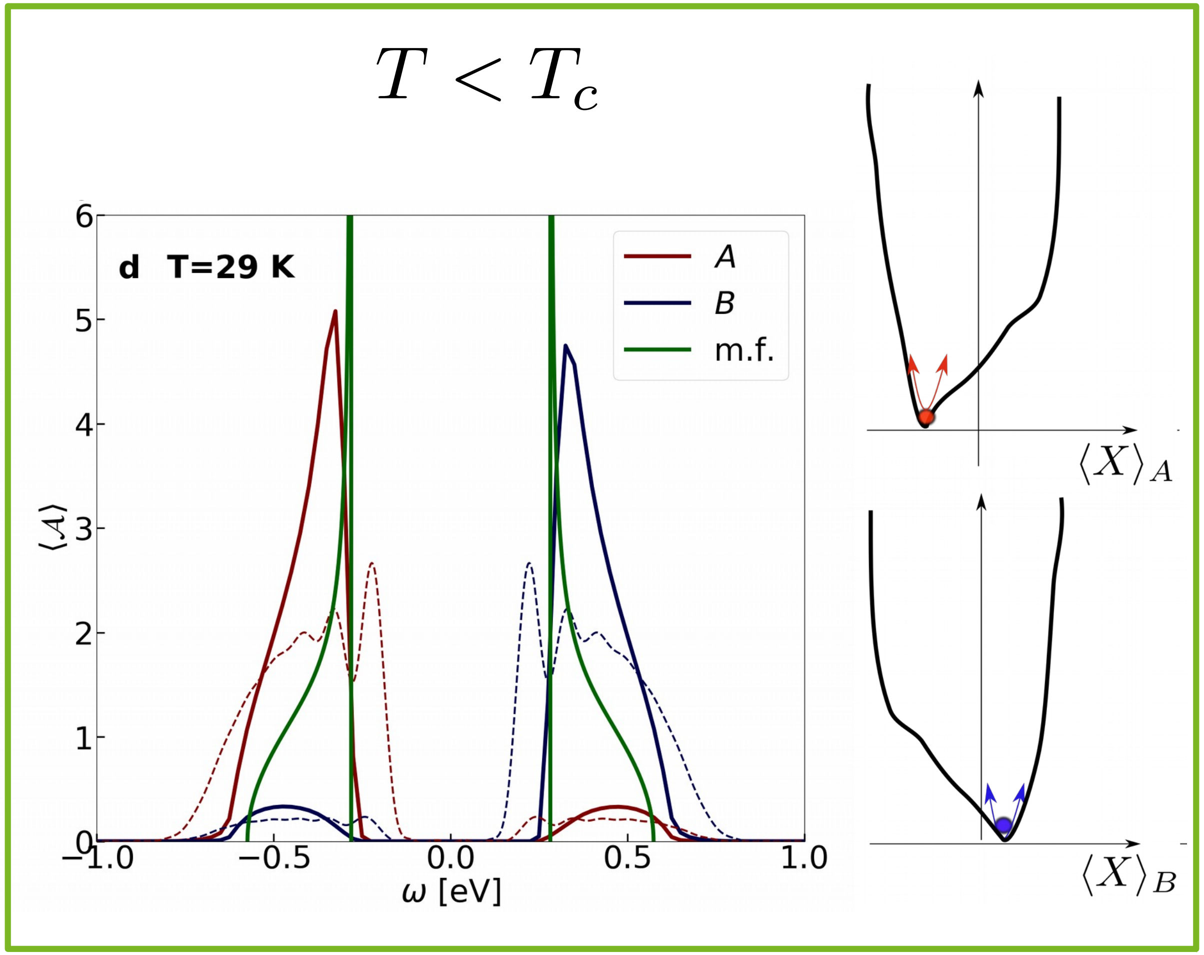


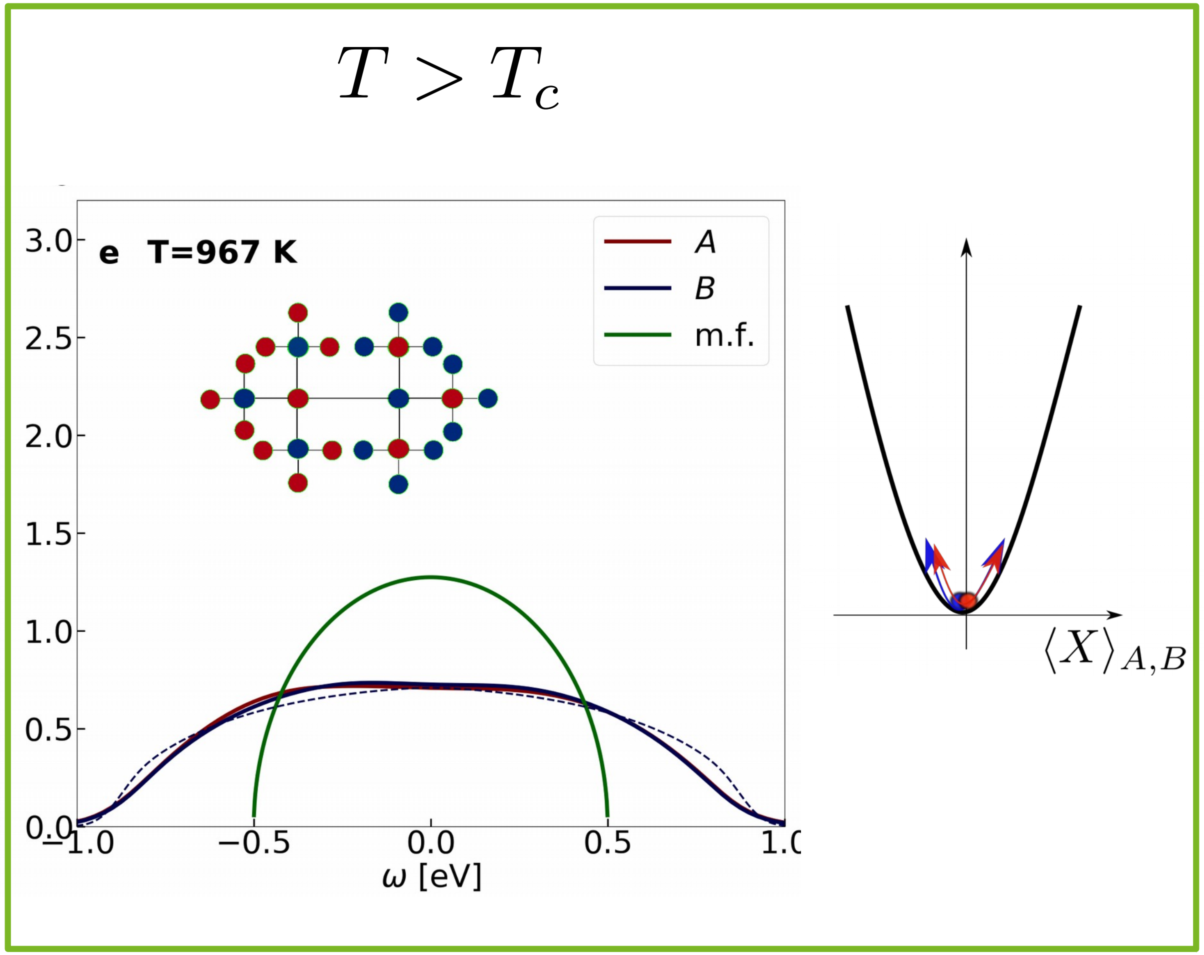


Picano, A. et al. : Phys. Rev. B 107, 245112 (2023)

Semiclassical solution: equilibrium

Opening of the **gap in the electronic spectra** below Tc





Mean-field solution

Perturbative DMFT solution : **spectral broadening** due to phonon fluctuations

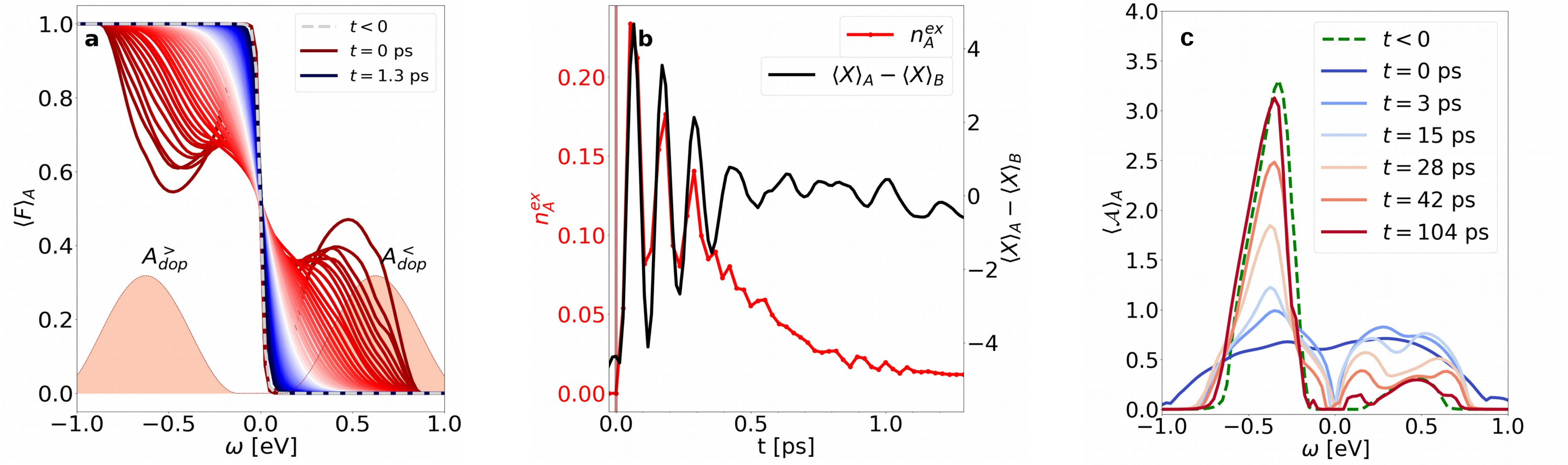
Semiclassical solution: dynamics

Short-time evolution after photo-excitation

Excitation protocol: population transfer from valence band to conduction band

Electron dynamics: open system, including dissipative heath bath

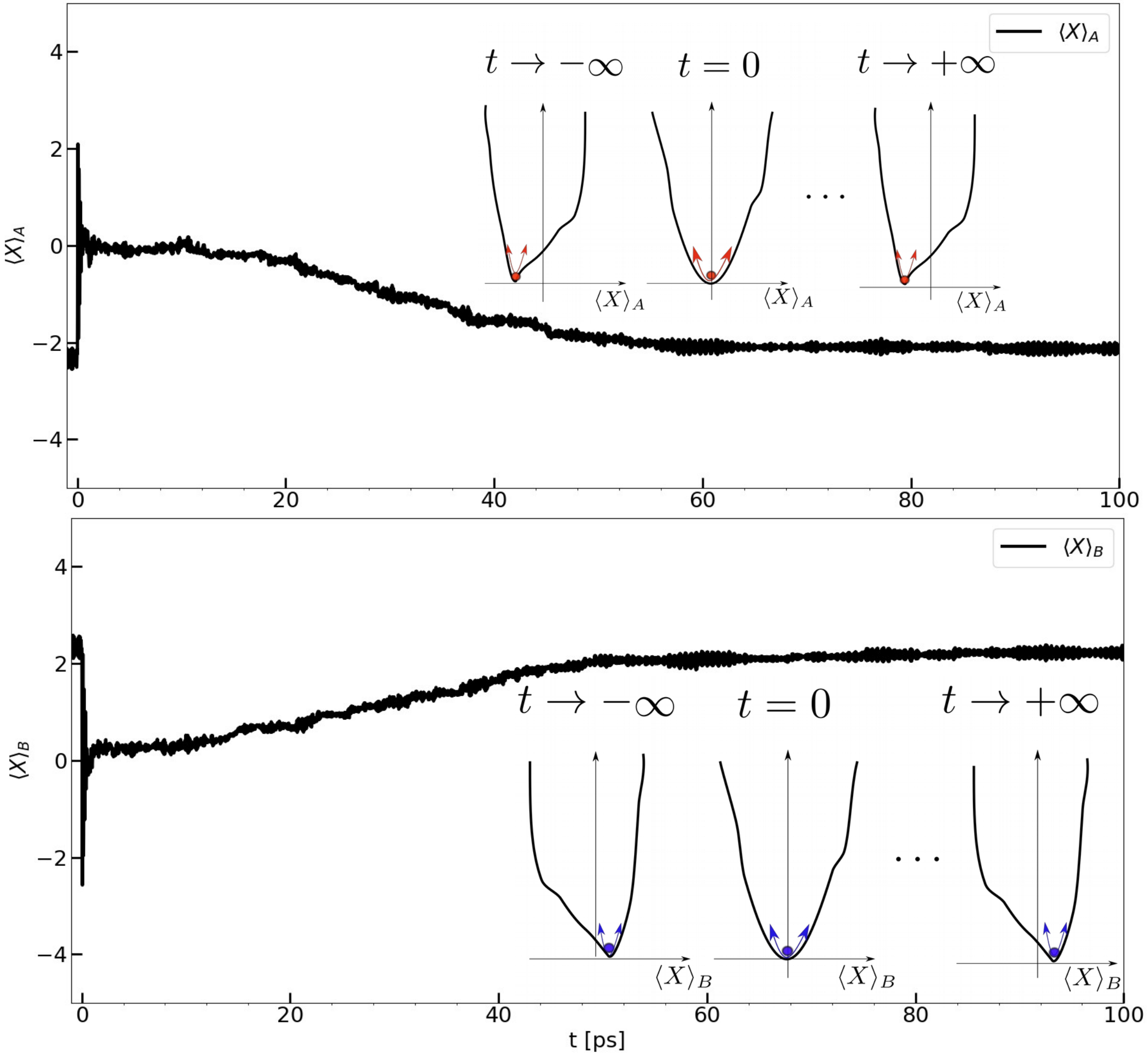
Electronic distribution function | Electronic excitation density | Incoherent spectral weight

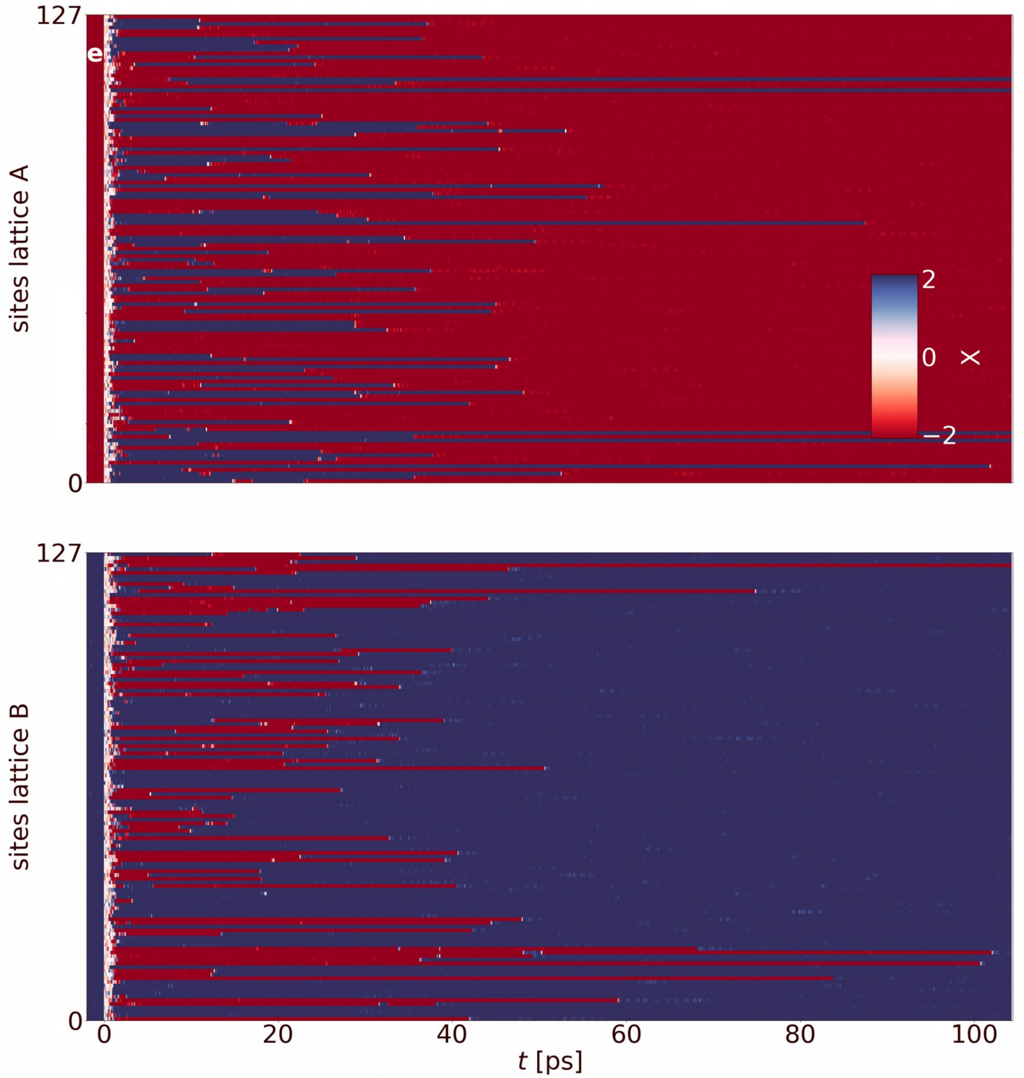


Picano, A. et al. : Phys. Rev. B 107, 245112 (2023)

Semiclassical solution: dynamics

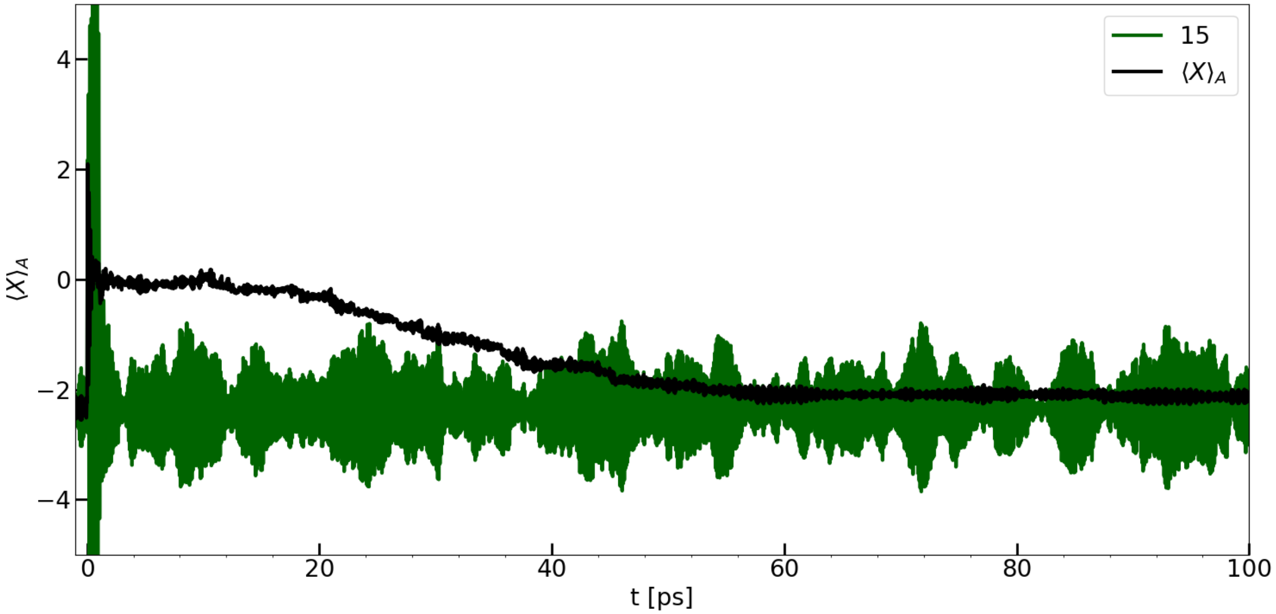
Long-time evolution: the average displacement and the single trajectories

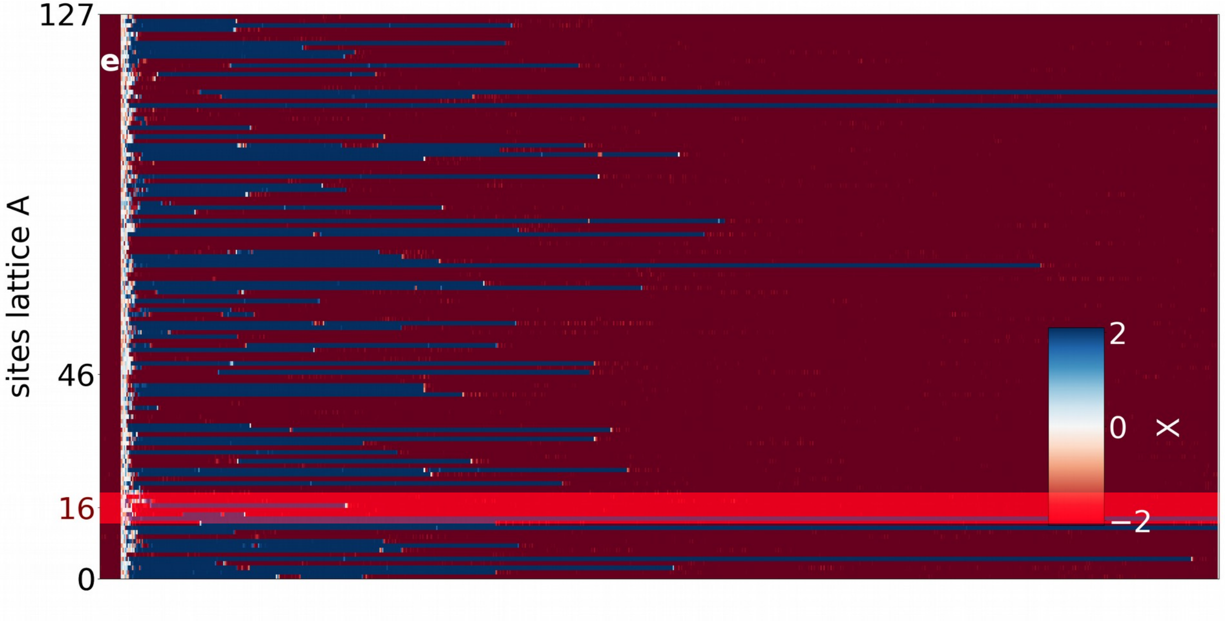




Semiclassical solution: dynamics

Long-time evolution: single trajectories not always representative of the average displacement



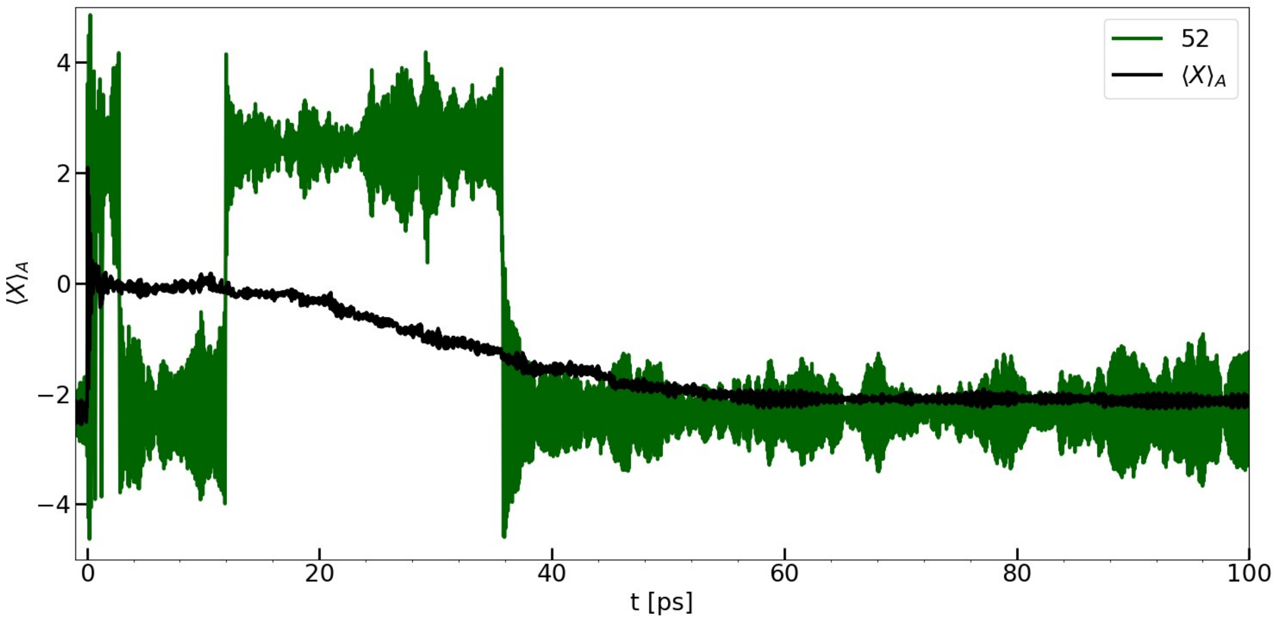


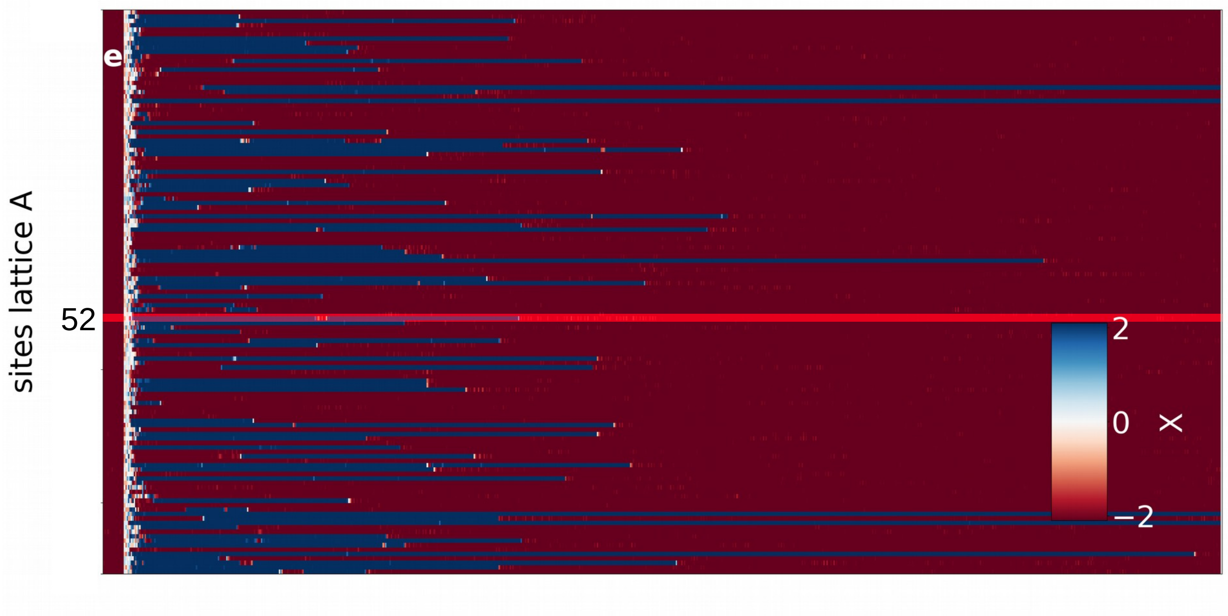
Trajectory 15 jumps to the global minimum well before the average does

Picano, A. et al. : Phys. Rev. B 107, 245112 (2023)

Semiclassical solution: dynamics

Long-time evolution: single trajectories not always representative of the average displacement



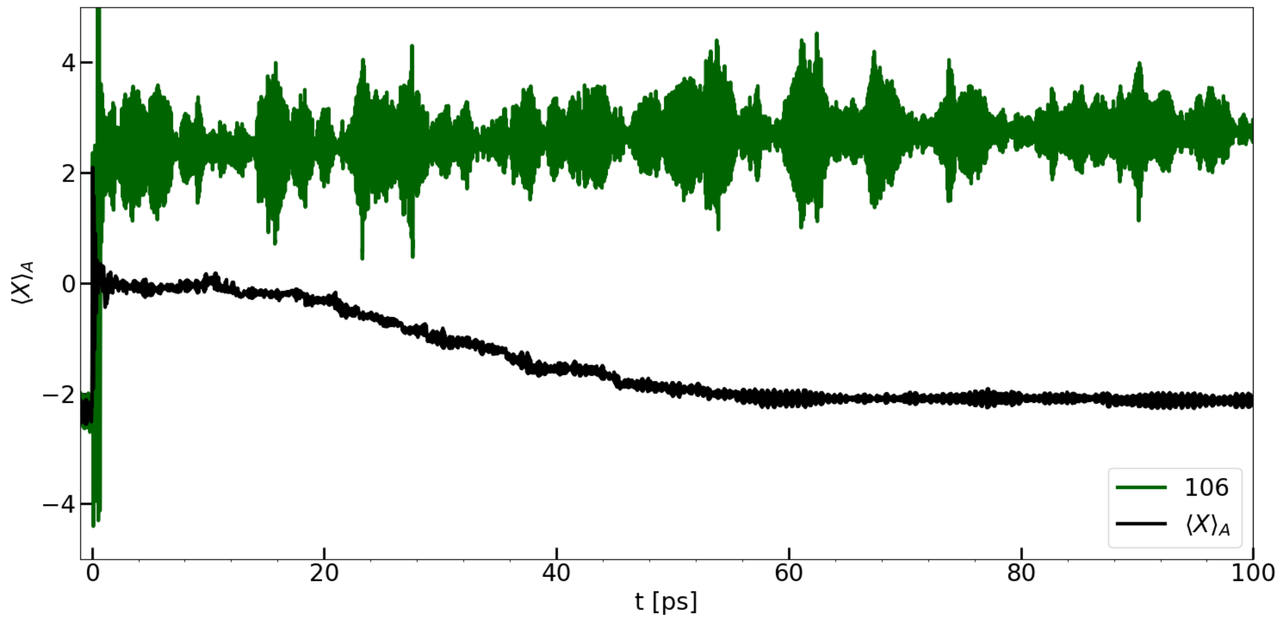


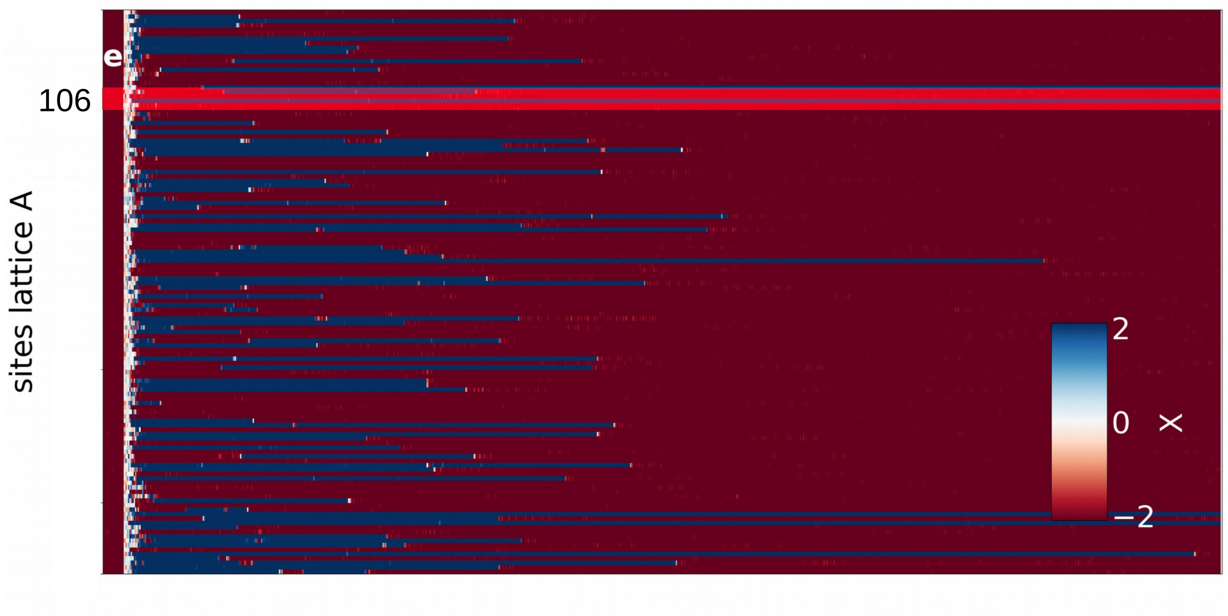
Trajectory 52 jumps back and forth from the metastable to the global minimum

Picano, A. et al. : Phys. Rev. B 107, 245112 (2023)

Semiclassical solution: dynamics

Long-time evolution: single trajectories not always representative of the average displacement





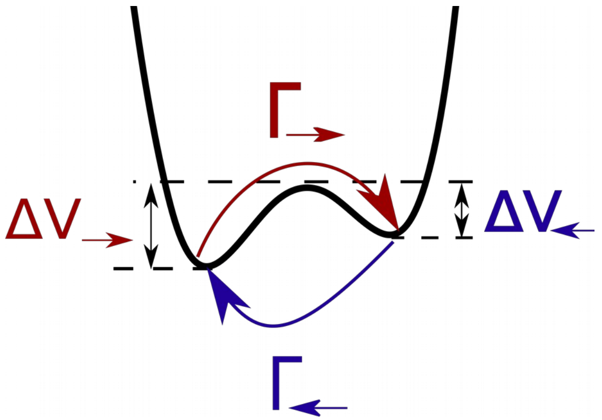
Trajectory 106 in the end of the simulation is still stacked in the metastable minimum

Picano, A. et al. : Phys. Rev. B 107, 245112 (2023)

Semiclassical solution: dynamics

Reconstruction of the double-well potential

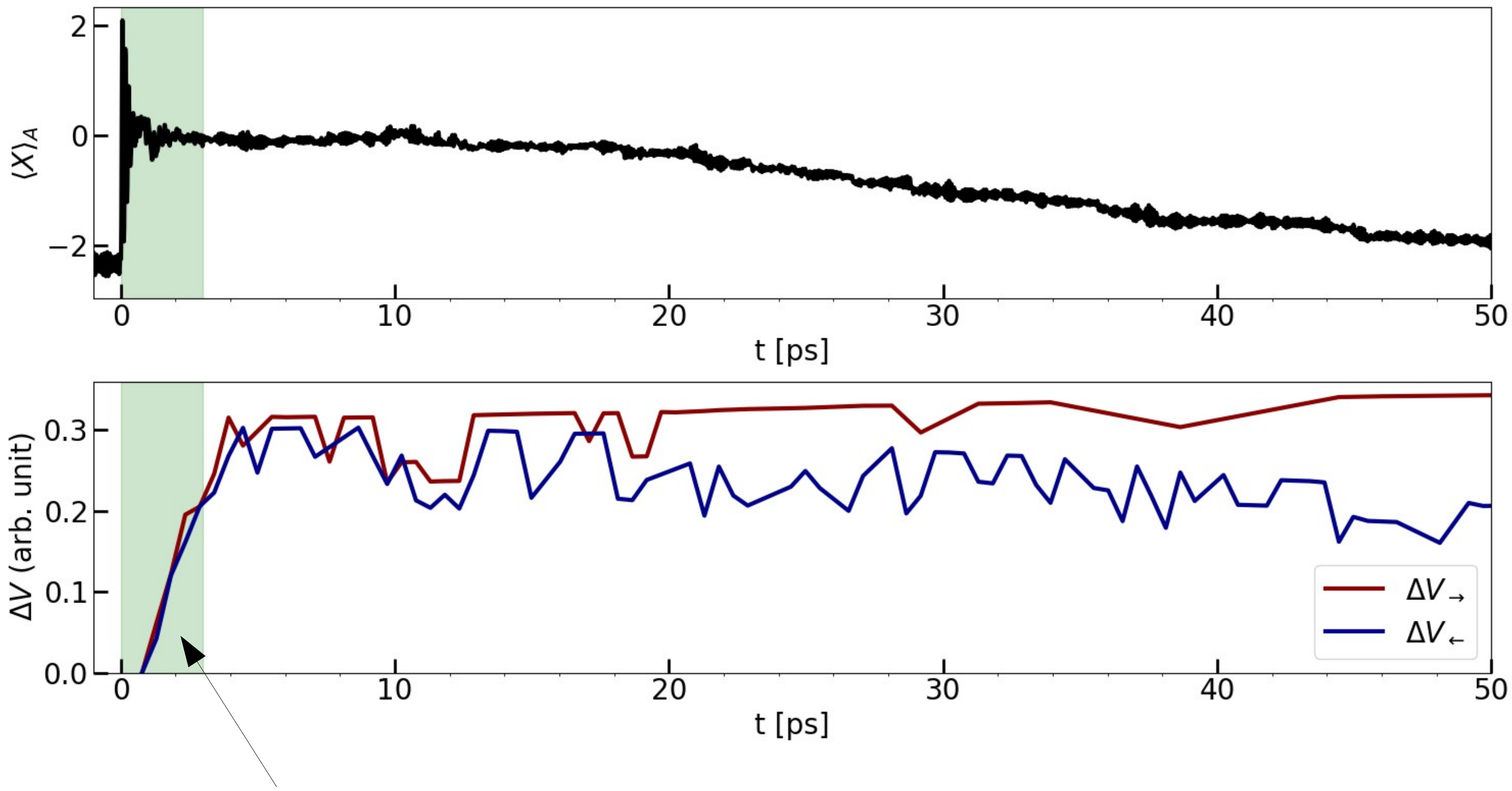
Local X_j moves in an effective asymmetric double-well potential:



Arrhenius transition rates:

$$\Gamma_\leftarrow = \Gamma_0 e^{-\Delta V_\leftarrow / T_e}$$

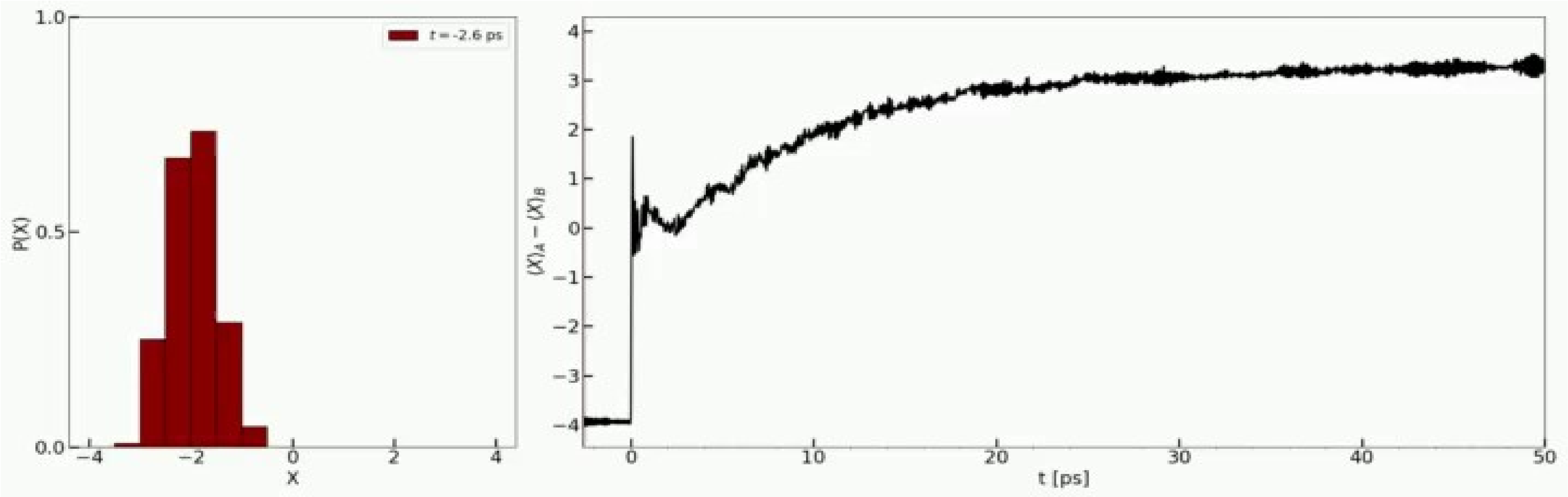
$$\Gamma_\rightarrow = \Gamma_0 e^{-\Delta V_\rightarrow / T_e}$$



Build-up of the potential barriers before symmetry breaking

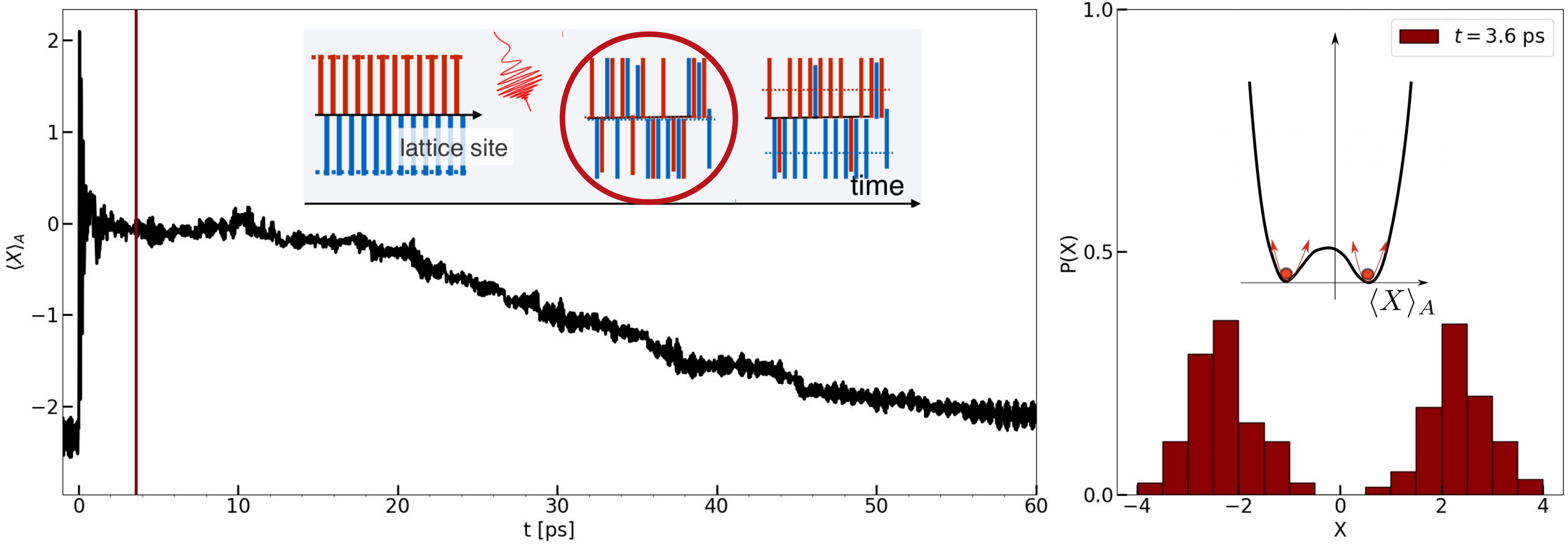
Semiclassical solution: dynamics

Long-time evolution: **ultrafast inhomogeneous disorder**



Inhomogeneous disordered state

Build-up of the potential barriers before symmetry breaking



Picano, A. et al. : Phys. Rev. B 107, 245112 (2023)

Inhomogeneous disordering

Summary

01 **Ultrafast** generation of **disorder** after simulated photoexcitation

02 Theory does not rely on the assumption of the existence of a **non-equilibrium potential**

03 Individual trajectories not always representative of the dynamics of the average order parameter

04 **Incoherent** electronic **spectral weight** after photoexcitation

Outlook: extension to more realistic models (e.g., two-band Mott insulators, VO_2, ...)

Thank you for your attention

Conclusions and Outlook

01 Full thermalization dynamics of a Slater AFM in the non-thermal critical region

Controlled truncation of memory integrals in KBEs

02 Inhomogeneous disordering at a photo-induced charge density wave transition

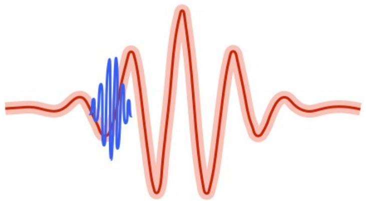
Electron dynamics : non-perturbative Quantum Boltzmann equation

Phonon dynamics : stochastic semiclassical theory for electron-phonon coupled systems

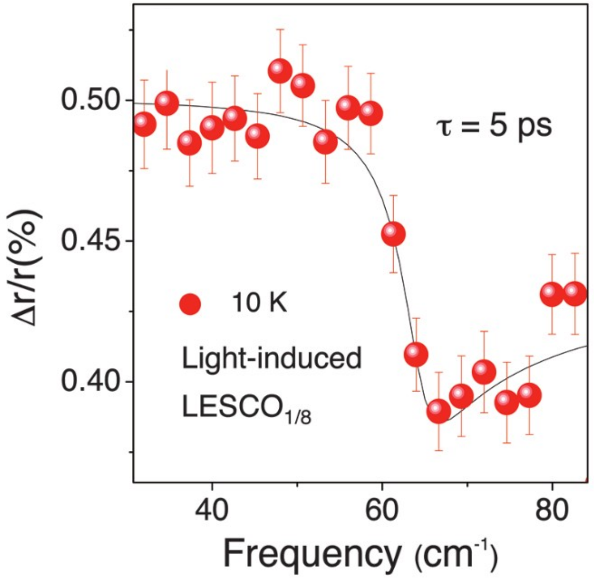
03 Outlook: extension to more realistic models (e.g., two-band Mott insulators, VO_2, ...)

Non-equilibrium phenomena in condensed matter

Pump-probe experiments

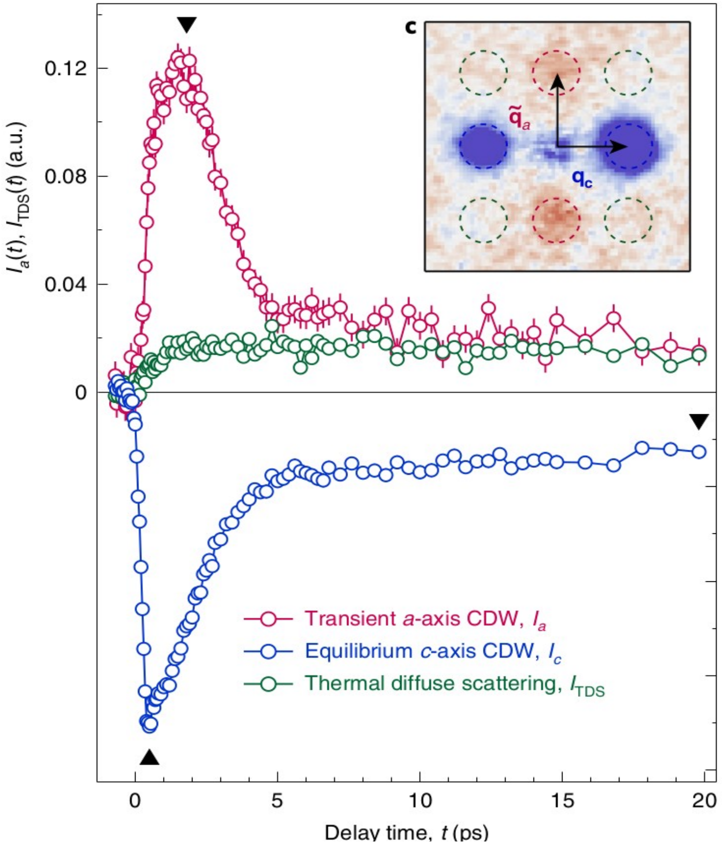


Light-induced
superconductivity



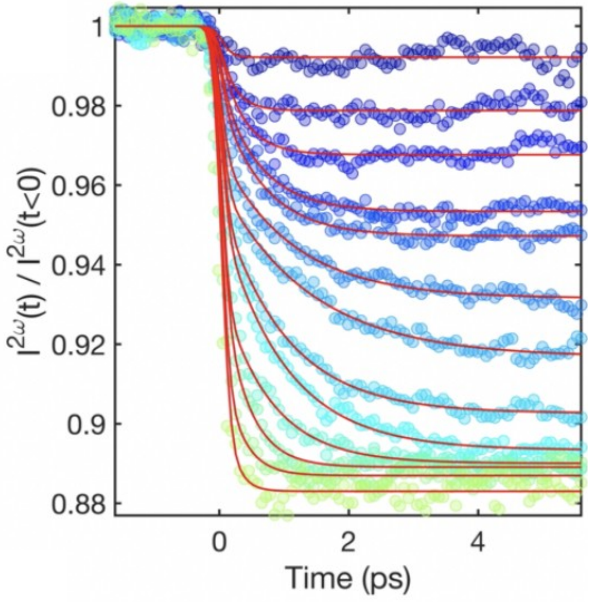
Fausti, D. et al.,
Science 331, 189-191 (2011)

Light-induced
charge density wave in $LaTe_3$



Kogar, A. et al., Nat. Phys. 16, 159-163 (2019)

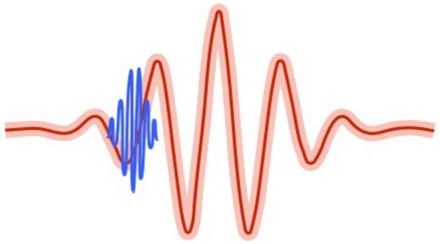
Light-induced
Insulator → metal transition

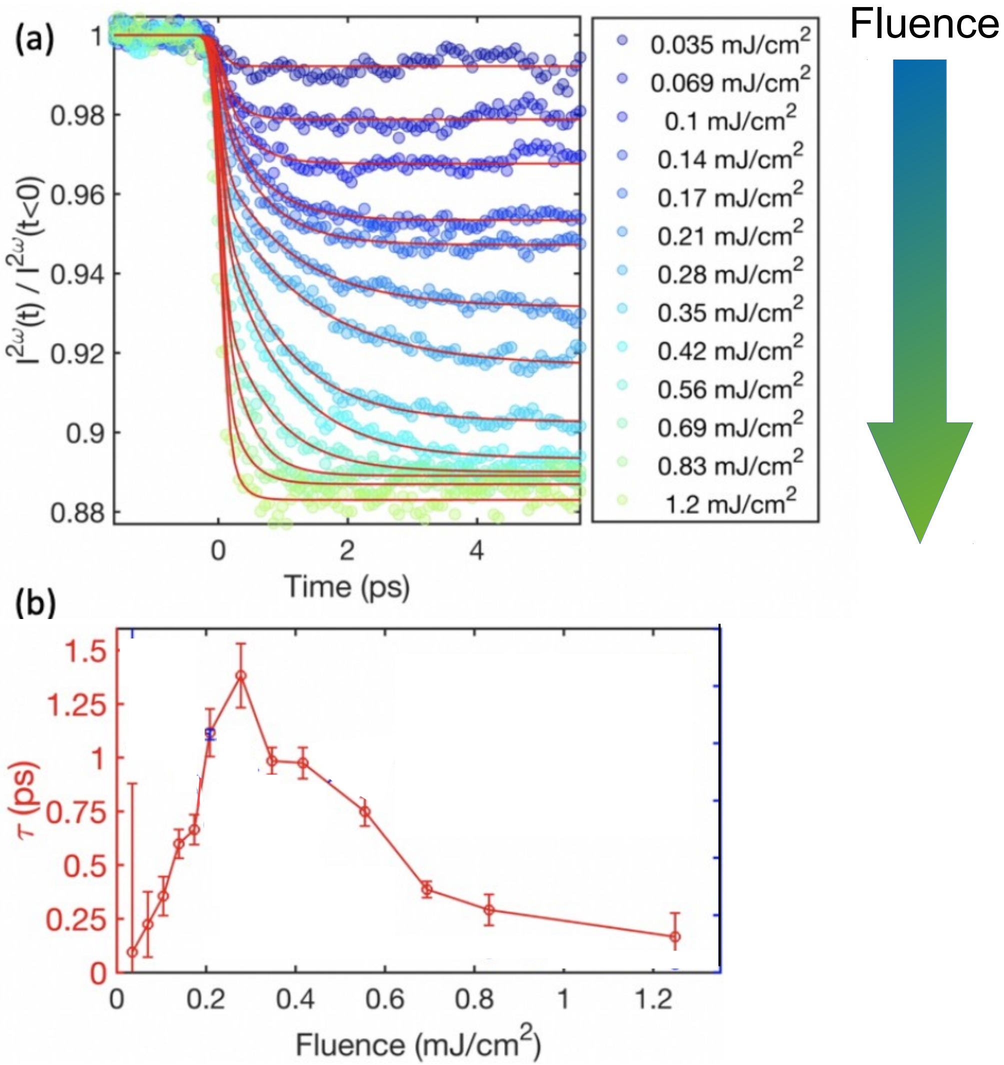


Carbin, T. et al., (2022)

Non-equilibrium phenomena in condensed matter

Light-induced AFM insulator → PM metal transition in $Ca_3Ru_2O_7$





Carbin, T. et al., under review (2022)

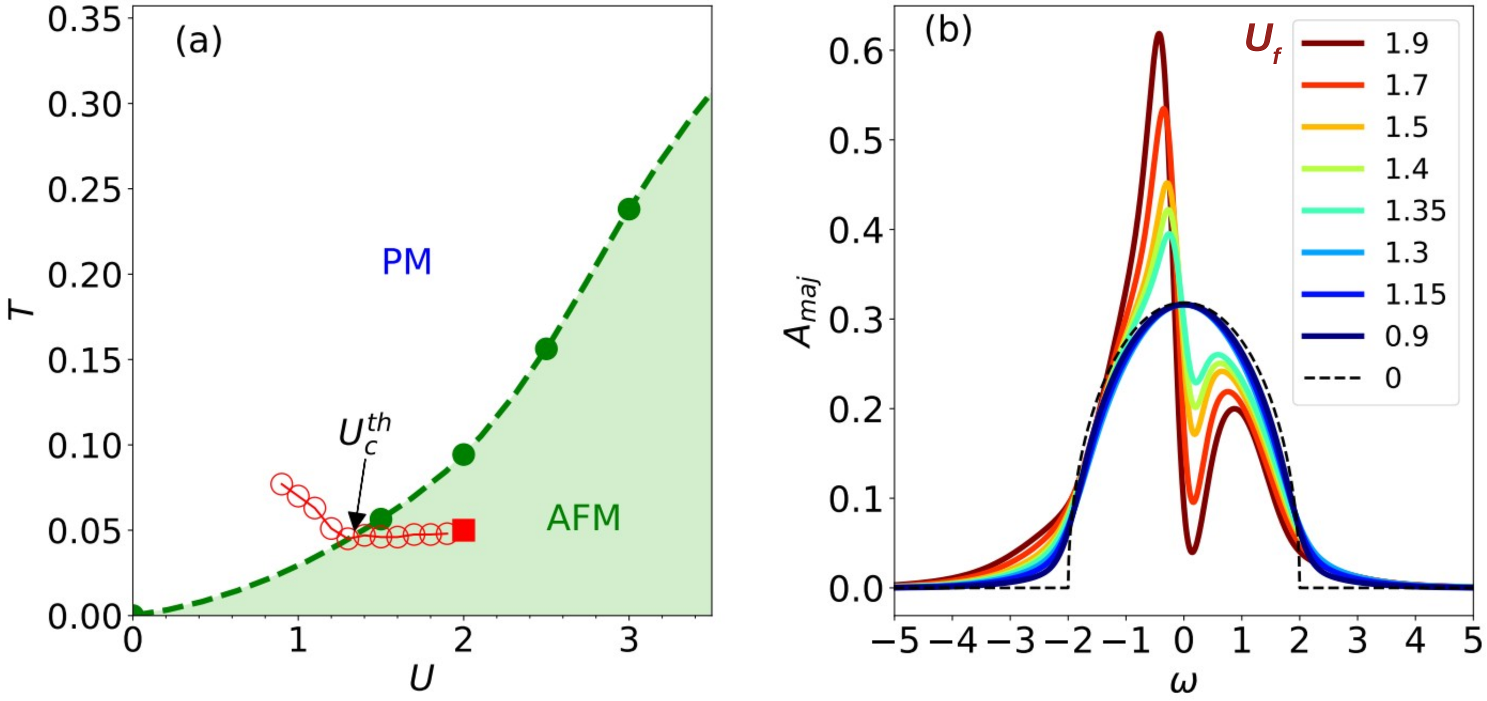
*Our goal: to simulate the **long-time** thermalization dynamics of correlated electrons and electron-lattice systems with **ultrafast** resolution*

Dynamical phase transition in a Slater antiferromagnet

Half-filled Hubbard model

Equilibrium phase diagram

$$H = -t_h \sum_{\langle i,j \rangle, \sigma} c^\dagger_{i\sigma} c_{j\sigma} + U(t) \sum_j (n_{j\uparrow} - \tfrac{1}{2})(n_{j\downarrow} - \tfrac{1}{2})$$

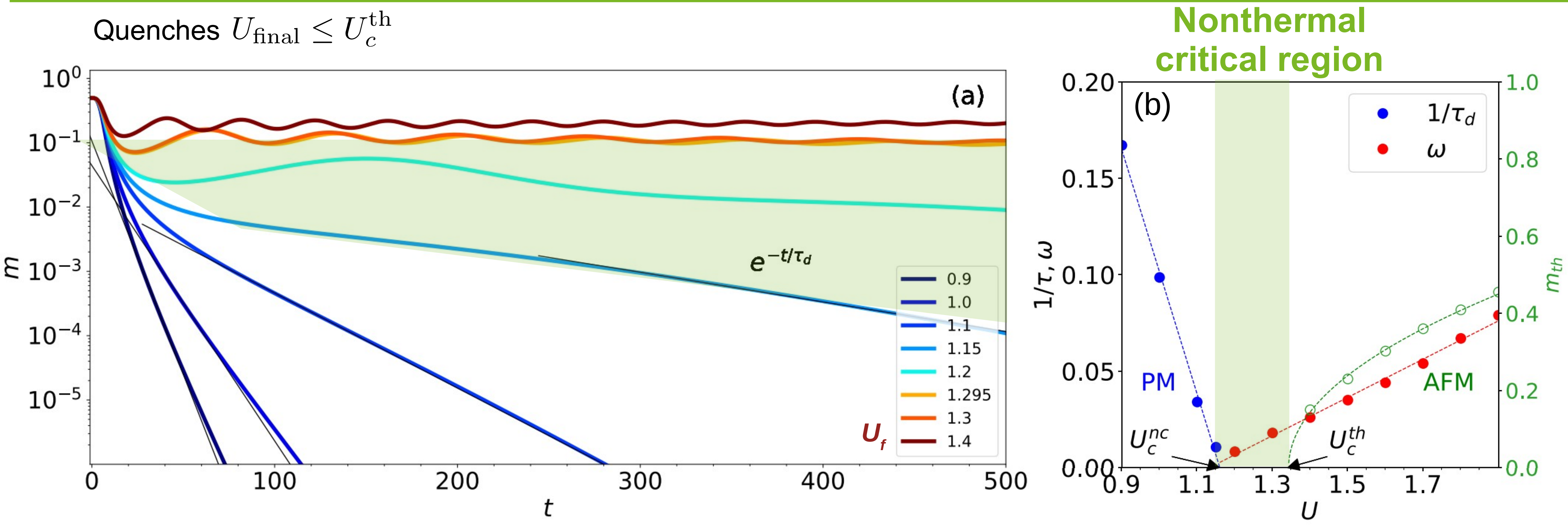


Picano, A. et al.: Phys. Rev. B 103, 165118 (2021)

Short-time dynamics

Nonthermal critical region: separates regimes of ordered and disordered prethermal states

Quenches $U_{\text{final}} \leq U_c^{\text{th}}$



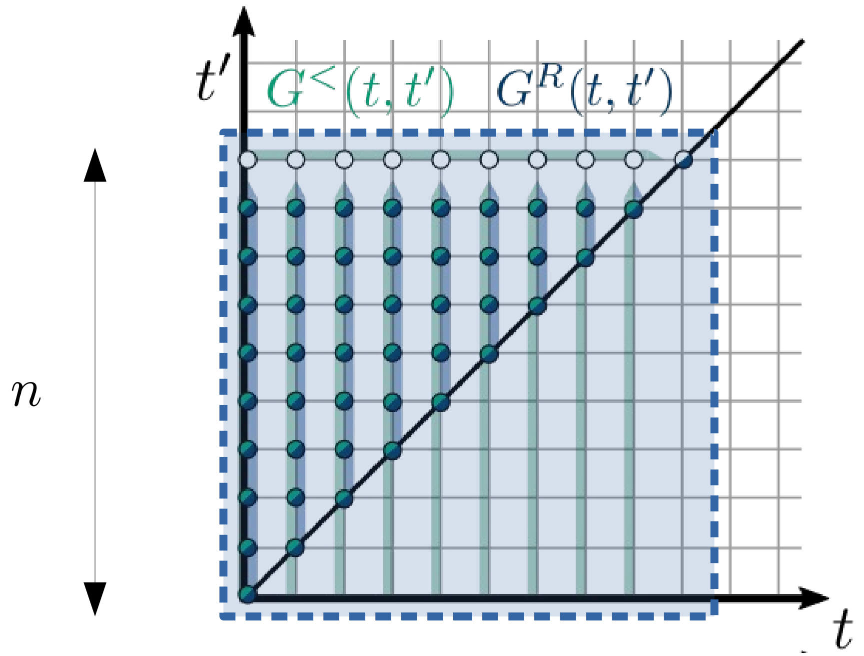
Staggered magnetization $$m = \frac{1}{2}(n_{A,\uparrow} - n_{B,\uparrow}) = \frac{1}{2}(n_{B,\downarrow} - n_{A,\downarrow})$$

Picano A. et al. : Phys. Rev. B 103, 165118 (2021)

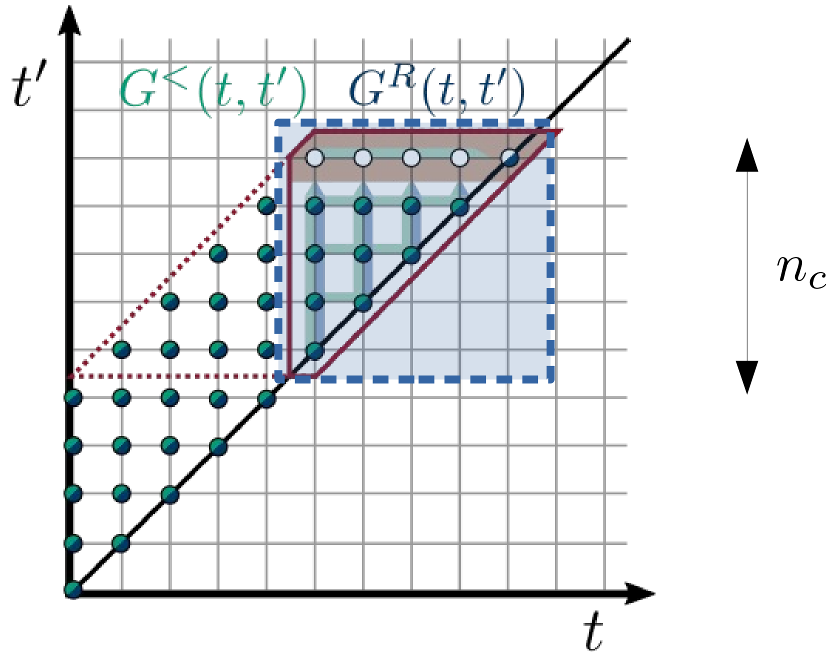
Memory truncated Kadanoff-Baym equations

Bypassing the limiting memory bottleneck of the full evaluation scheme

Memory untruncated scheme



Memory **truncated** scheme
(If Σ decays sufficiently fast in time)



Computational effort $\sim \mathcal{O}(n^3)$ $\sim \mathcal{O}(n n_c^2)$

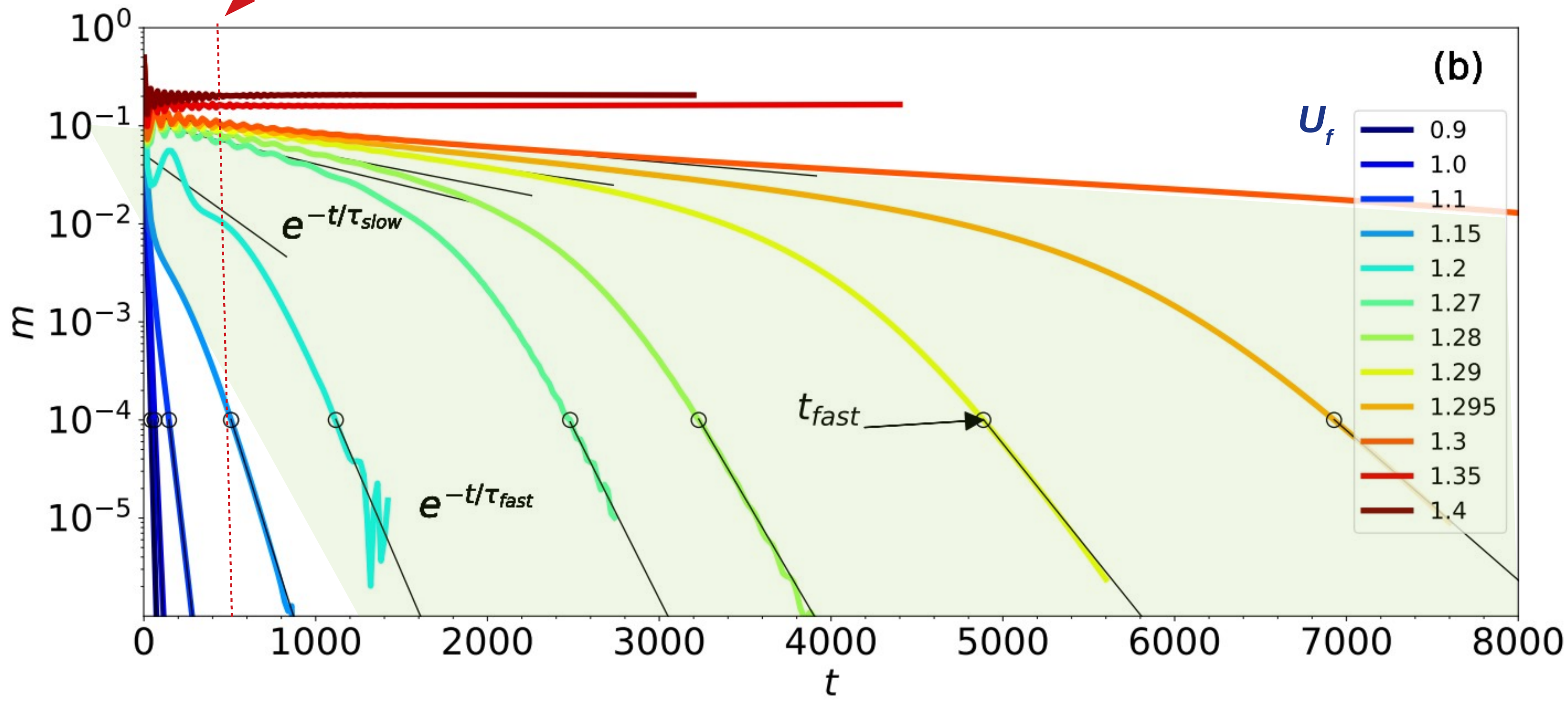
Memory occupation $\sim \mathcal{O}(n^2)$ $\sim \mathcal{O}(n_c^2)$

Stahl C., Dasari N., Li J., Picano A. et al.: Phys. Rev. B 105, 115146 (2022)

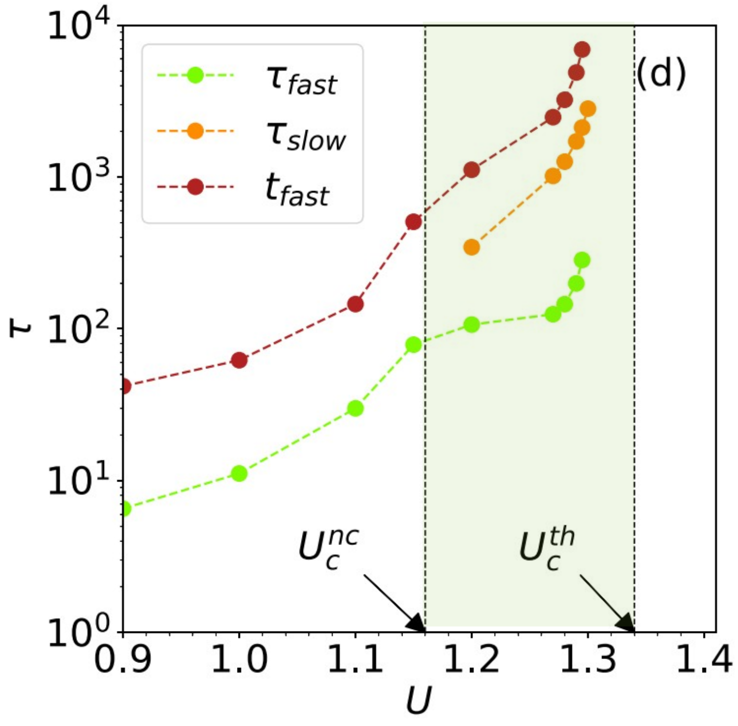
Long-time dynamics

Accelerated gap collapse

Two order of magnitudes longer in time!



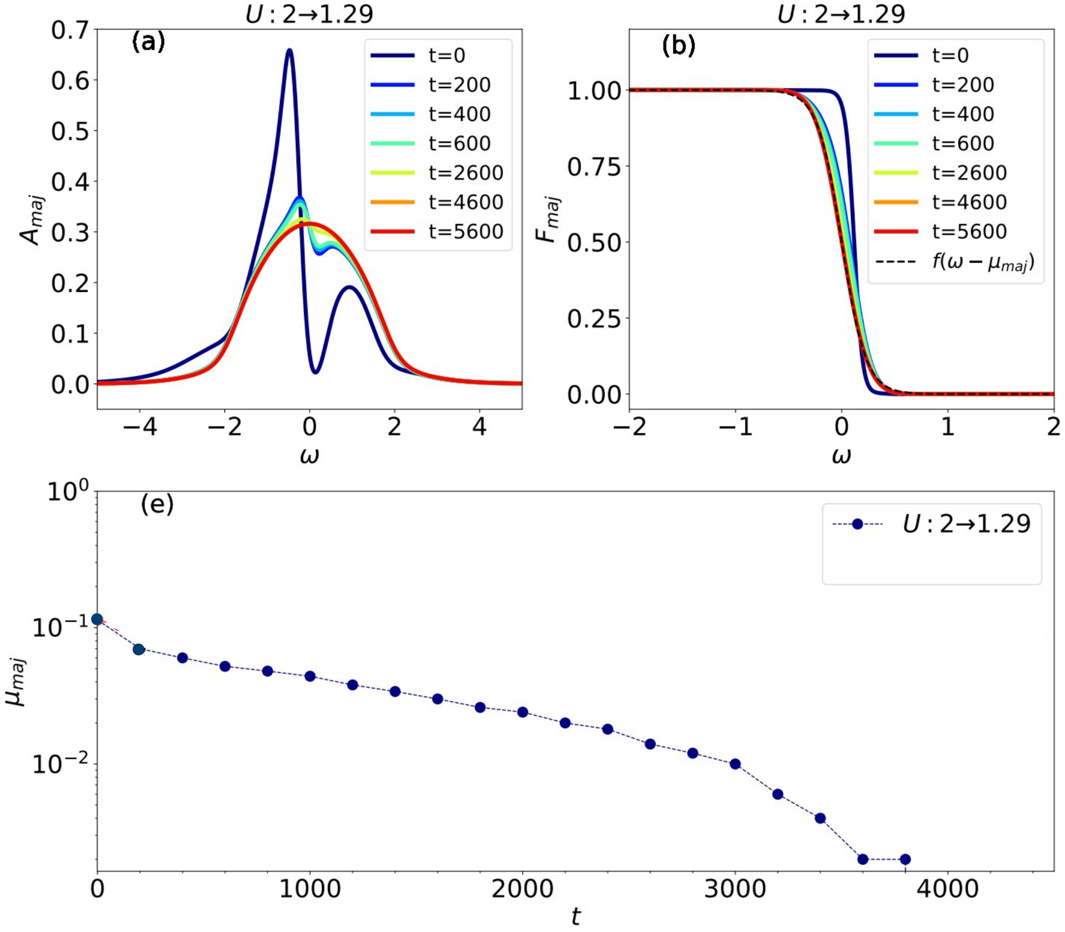
Nonthermal critical region



Picano, A. et al.: Phys. Rev. B 103, 165118 (2021)

Prethermal state

Can be understood in terms of nearly conserved quantities like the individual band populations



$$F_{\mathrm{maj}}(\omega) = f(\omega - \mu_{\mathrm{maj}}, T_{\mathrm{maj}})$$

Picano, A. et al.: Phys. Rev. B 103, 165118 (2021)

Summary

By means of the controlled truncation of memory integrals in KBEs...

01 Simulation of the slow melting of a prethermal symmetry broken state after an interaction quench in the Hubbard model

02 Highly nonlinear gap closing dynamics: a slow evolution of the order parameter quickly transits into a faster gap collapse after the gap has fallen below a threshold

03 The initial slow evolution of the order parameter is captured by a mean-field theory while the gap collapse is not

Inhomogeneous disordering in a charge density wave transition

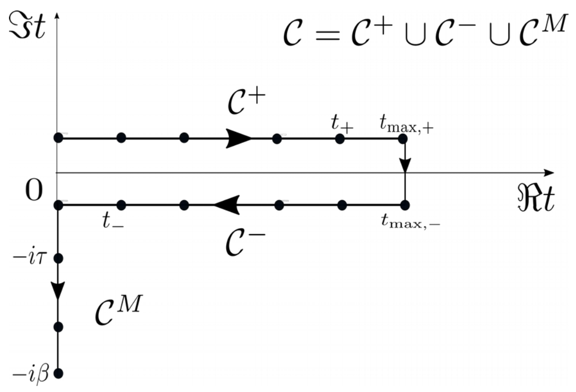
Thank you for your attention

Memory-truncated Kadanoff-Baym equations

Evaluation of memory integrals

Kadanoff-Baym equations

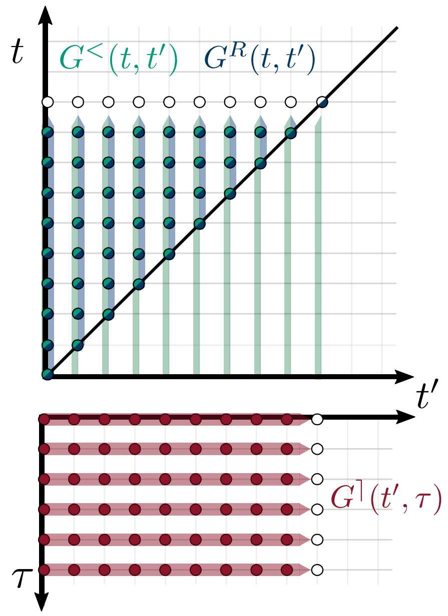
$$i\partial_t G(t,t') - \int_{\mathcal{C}} d\bar{t} \;\; [\Sigma + \Delta](t,\bar{t})G(\bar{t},t') = \delta_{\mathcal{C}}(t,t')$$



For equidistant grid with *n* time-steps:

Computational effort $\sim \mathcal{O}(n^3)$

Memory occupation $\sim \mathcal{O}(n^2)$



Evaluation of memory integrals

Kadanoff-Baym equations

KBEs for the single Keldysh components:

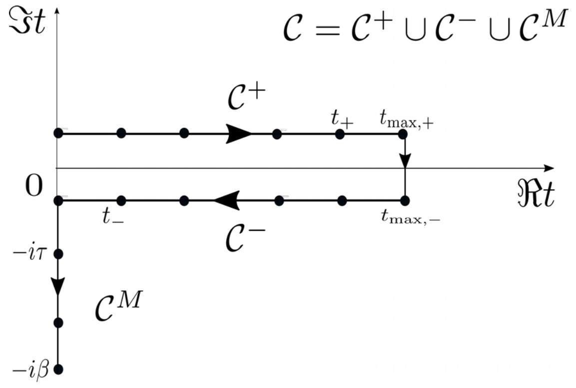
$$[i\partial_t - \epsilon(t)]G^R(t,t') - \int_{t'}^{t} d\bar{t}\ \Sigma^R(t,\bar{t})G^R(\bar{t},t') = \delta(t-t') \qquad (1)$$

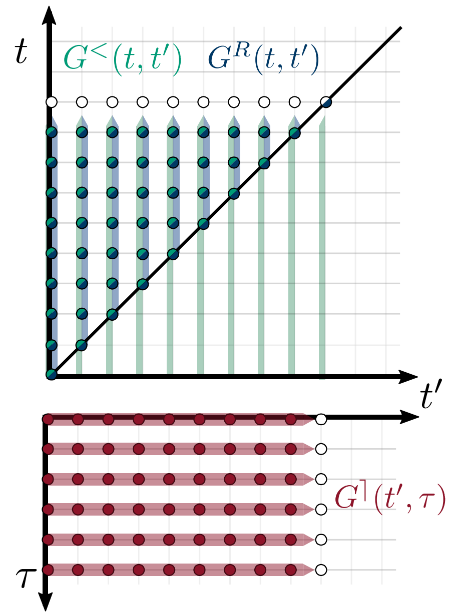
$$[i\partial_t - \epsilon(t)]G^{\rceil}(t,\tau) - \int_{0}^{t} d\bar{t}\ \Sigma^R(t,\bar{t})G^{\rceil}(\bar{t},\tau) = \int_0^{\beta} d\tau'\ \Sigma^{\rceil}(t,\tau')G^M(\tau'-\tau) \qquad (2)$$

$$[i\partial_t - \epsilon(t)]G^<(t,t') - \int_{0}^{t} d\bar{t}\ \Sigma^R(t,\bar{t})G^<(\bar{t},t')$$

$$= \int_0^{t'} d\bar{t}\ \Sigma^<(t,\bar{t})G^R(t',\bar{t})^\dagger \pm i \int_0^{\beta} d\tau\ \Sigma^{\rceil}(t,\tau')G^{\rceil}(t',\beta-\tau')^\dagger, \qquad (3)$$

$$[-\partial_\tau - \epsilon(0^-)]G^M(\tau) - \int_0^{\beta} d\bar{\tau}\ \Sigma^M(\tau-\bar{\tau})G^M(\bar{\tau}) = \delta(\tau) \qquad (4)$$





Computational effort $\sim \mathcal{O}(n^3)$

Memory occupation $\sim \mathcal{O}(n^2)$

Evaluation of memory integrals

Truncated Kadanoff-Baym equations

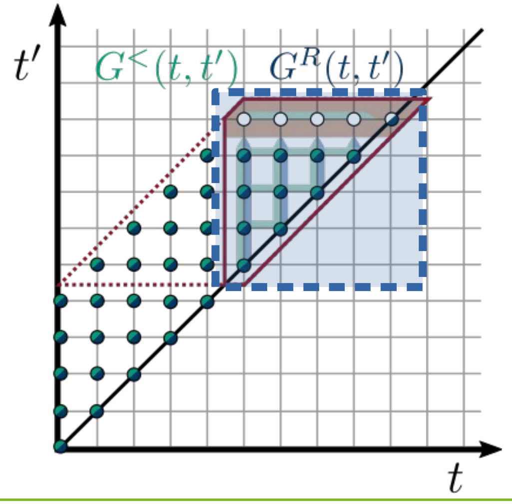
If the self-energy decays to zero, for large enough time one can impose:

$$\begin{array}{lll} \Sigma^R(t,t') = \Sigma^<(t,t') = 0 & \text{for} & t-t' > t_c, \\ \Sigma^\rceil(t,\tau) = 0 & & t > t_c \end{array}$$

The self-consistency scheme therefore reduces to:

$$[i\partial_t - \epsilon(t)]G^R(t,t-s) - \int_{t-s}^{t} d\bar{t}\; \Sigma^R(t,\bar{t})G^R(\bar{t},t-s) = \delta(s)$$

$$[i\partial_t - \epsilon(t)]G^<(t,t-s) - \int_{t-t_c}^{t} d\bar{t}\; \Sigma^R(t,\bar{t})G^<(\bar{t},t-s) = \int_{t-t_c}^{t'} d\bar{t}\Sigma^<(t,\bar{t})G^R(t-s,\bar{t})^\dagger$$



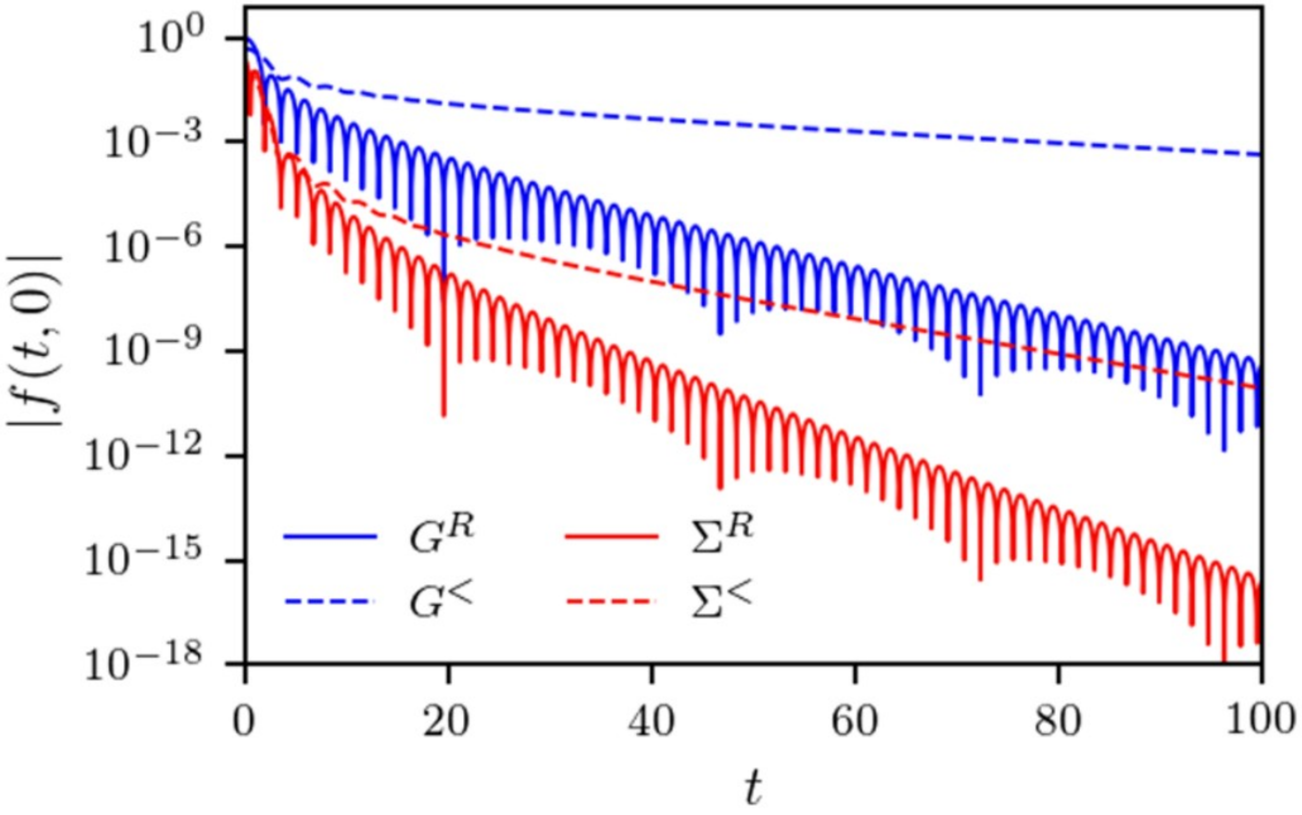
Computational effort $\sim \mathcal{O}(nn_c^2)$

Memory occupation $\sim \mathcal{O}(n_c^2)$

Half-filled Hubbard model on infinite Bethe lattice

Decay of the two-times correlation functions

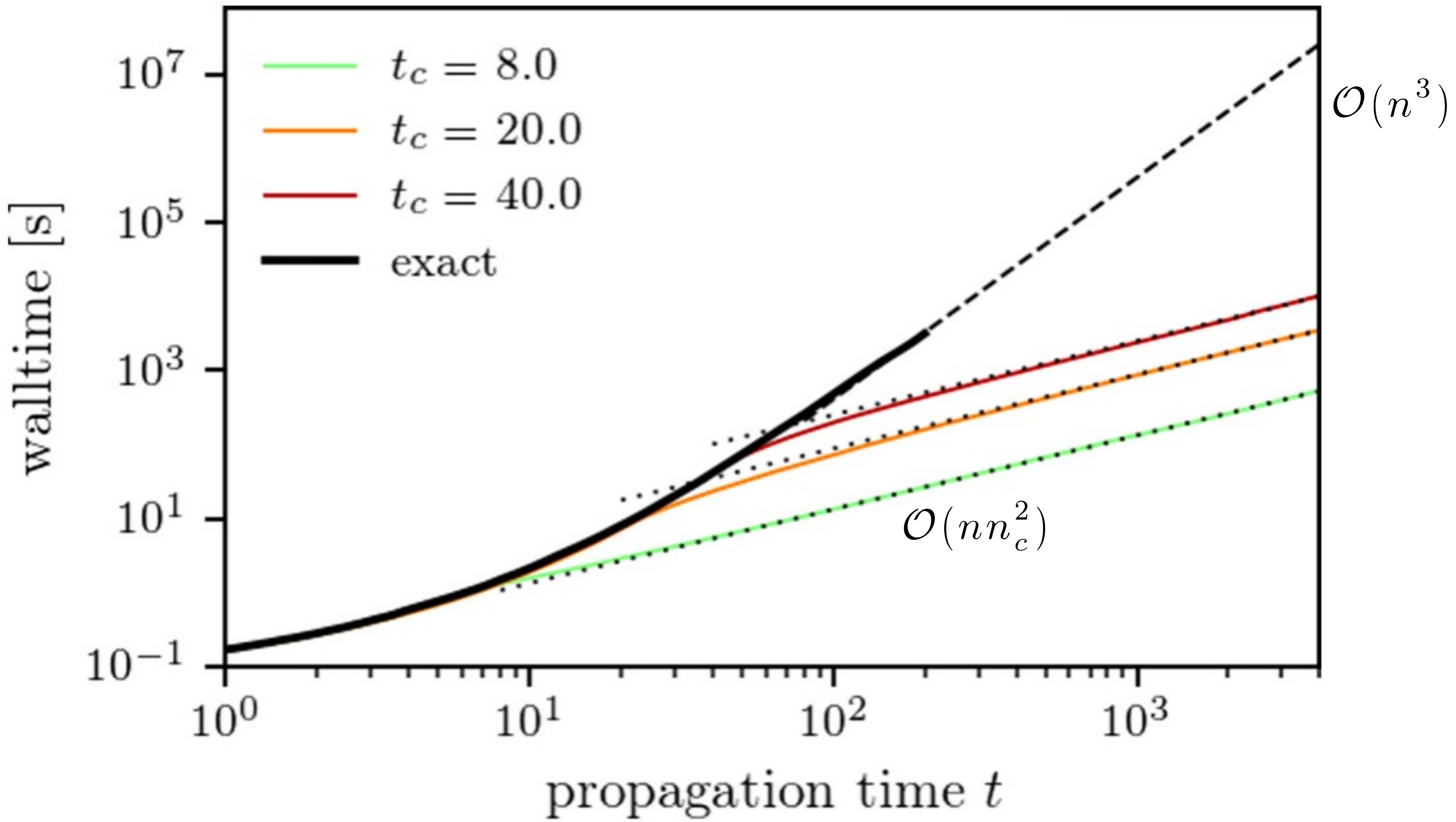
$$H = -t_h \sum_{\langle i,j \rangle, \sigma} c^\dagger_{i\sigma} c_{j\sigma} + U(t) \sum_j (n_{j\uparrow} - \tfrac{1}{2})(n_{j\downarrow} - \tfrac{1}{2})$$



Stahl C., Dasari N., Li J., Picano A. et al.: Phys. Rev. B 105, 115146 (2022)

Total simulation time

Non-truncated vs truncated KBEs

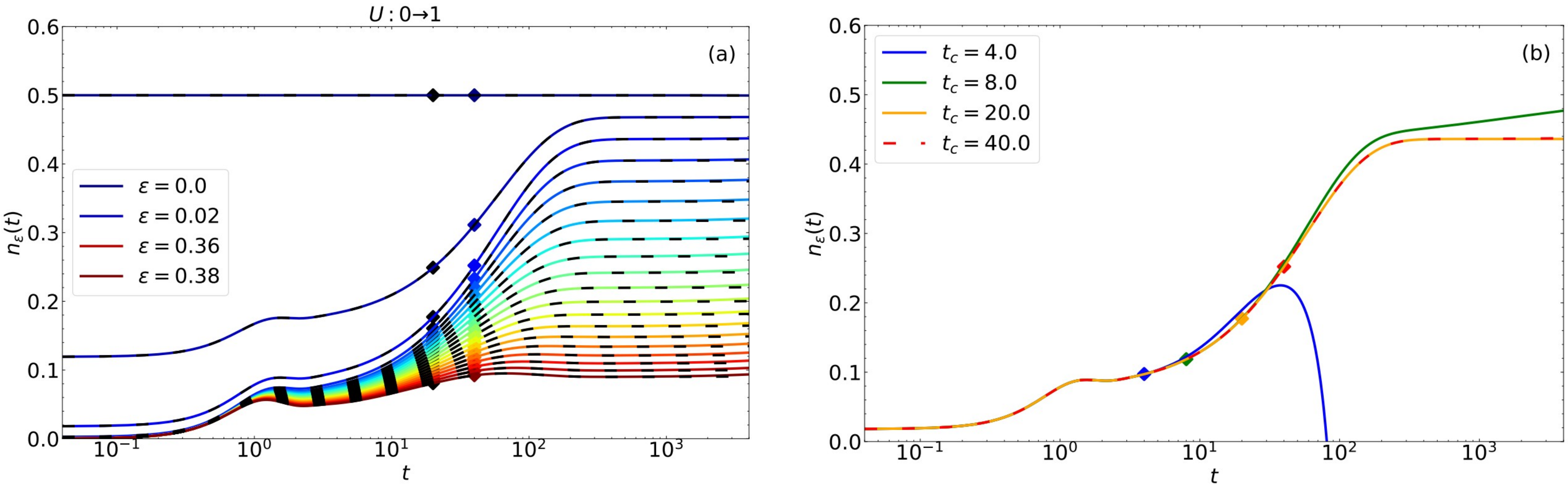


Stahl C., Dasari N., Li J., Picano A. et al.: Phys. Rev. B 105, 115146 (2022)

Half-filled Hubbard model on infinite Bethe lattice

The fast prethermalization and the slow thermalization

$$H = -t_h \sum_{\langle i,j \rangle,\sigma} c^\dagger_{i\sigma} c_{j\sigma} + U(t) \sum_j (n_{j\uparrow} - \tfrac{1}{2})(n_{j\downarrow} - \tfrac{1}{2})$$

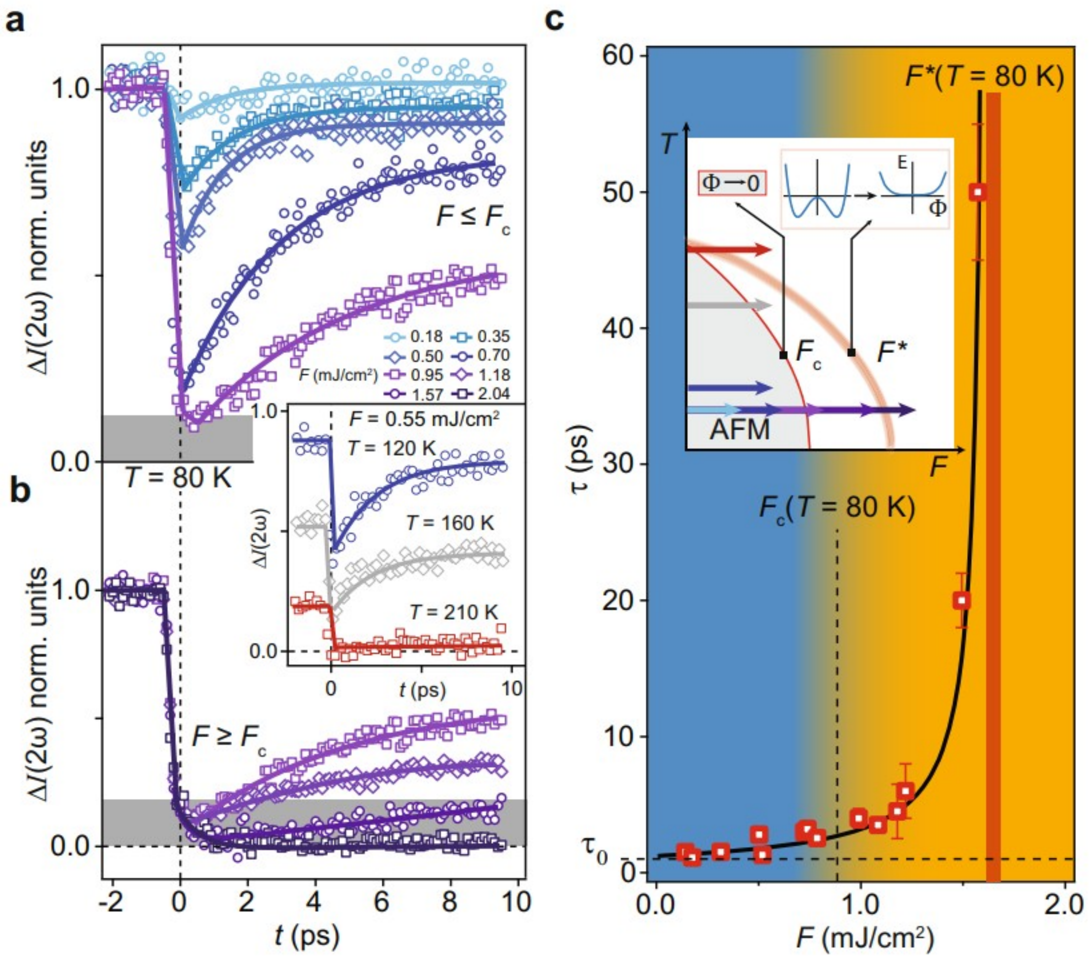


Stahl C., Dasari N., Li J., Picano A. et al.: Phys. Rev. B 105, 115146 (2022)

Accelerated gap collapse

Decoupling of static and dynamic criticality

Strongly driven AFM Mott insulator



De la Torre, A. et al. : Communications physics 5, 35 (2022)

Half-filled Hubbard model on infinite bethe lattice

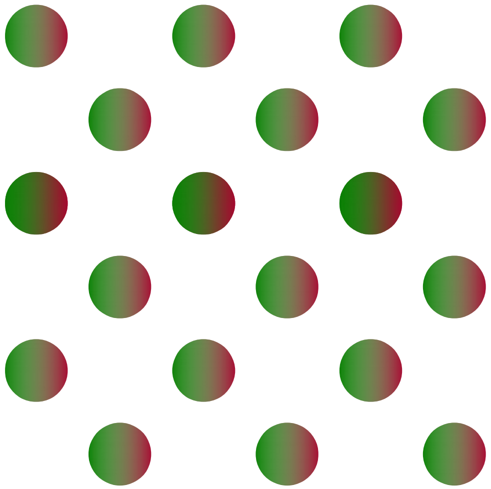
Equilibrium phase diagram

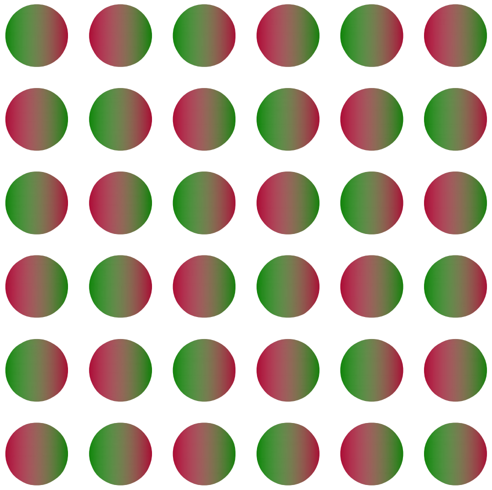
$$H = -t_h \sum_{\langle i,j \rangle,\sigma} c^\dagger_{i\sigma} c_{j\sigma} + U(t) \sum_j (n_{j\uparrow} - \tfrac{1}{2})(n_{j\downarrow} - \tfrac{1}{2})$$

AFM order at low T

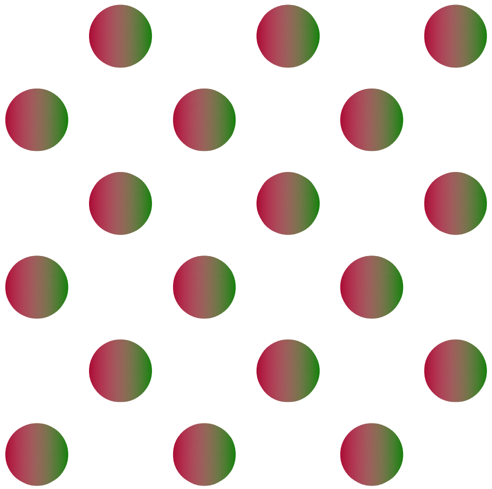
$$n_{j,\uparrow} \neq n_{j,\downarrow}$$

Sublattice A : majority of spin $\uparrow$





Sublattice B : majority of spin $\downarrow$



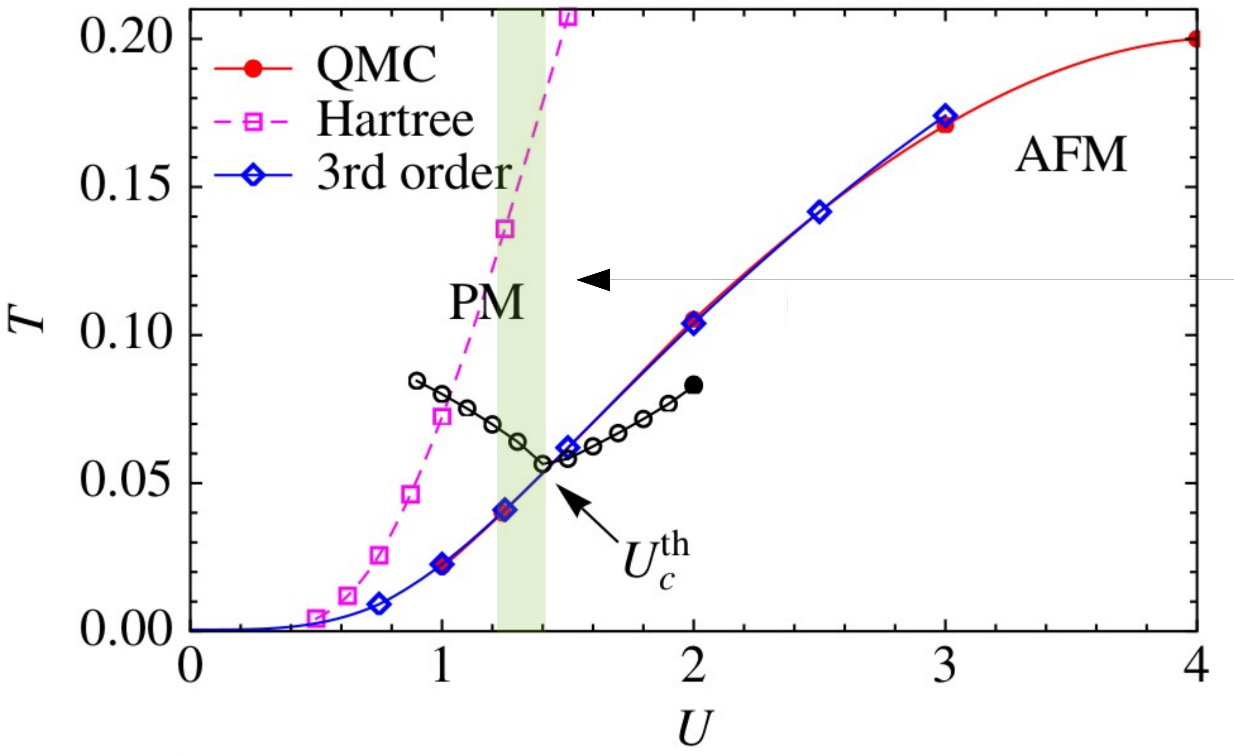
Staggered magnetization

$$m = \frac{1}{2}(n_{A,\uparrow} - n_{B,\uparrow}) = \frac{1}{2}(n_{B,\downarrow} - n_{A,\downarrow})$$

Half-filled Hubbard model on infinite bethe lattice

Equilibrium phase diagram

$$H = -t_h \sum_{\langle i,j \rangle,\sigma} c^\dagger_{i\sigma} c_{j\sigma} + U(t) \sum_j (n_{j\uparrow} - \tfrac{1}{2})(n_{j\downarrow} - \tfrac{1}{2})$$

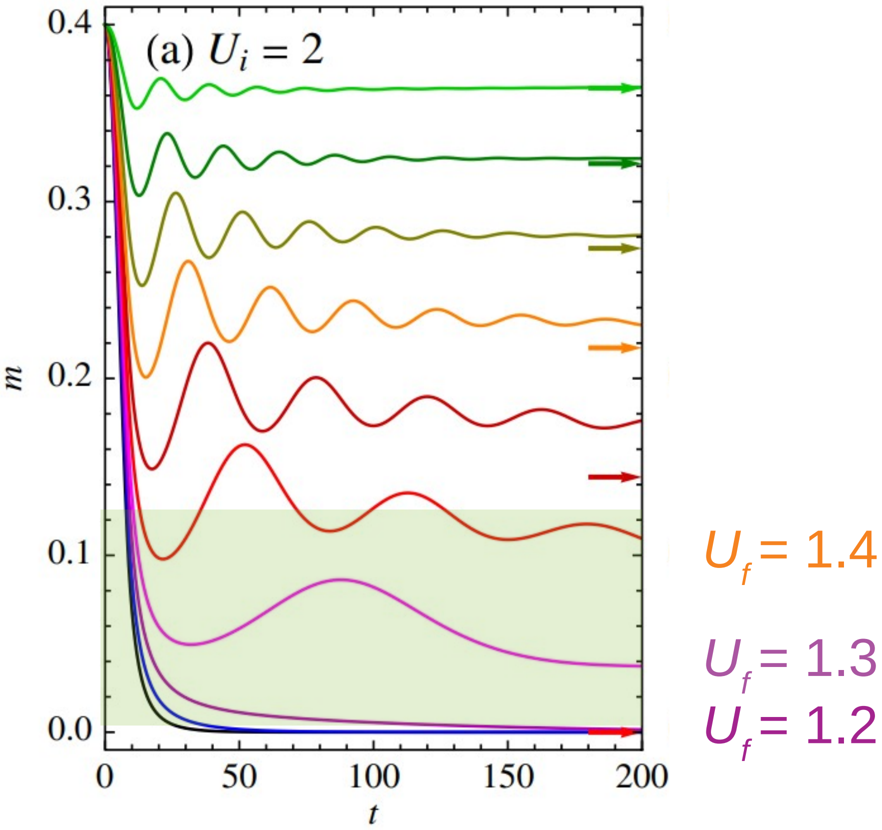


Nonthermal critical region

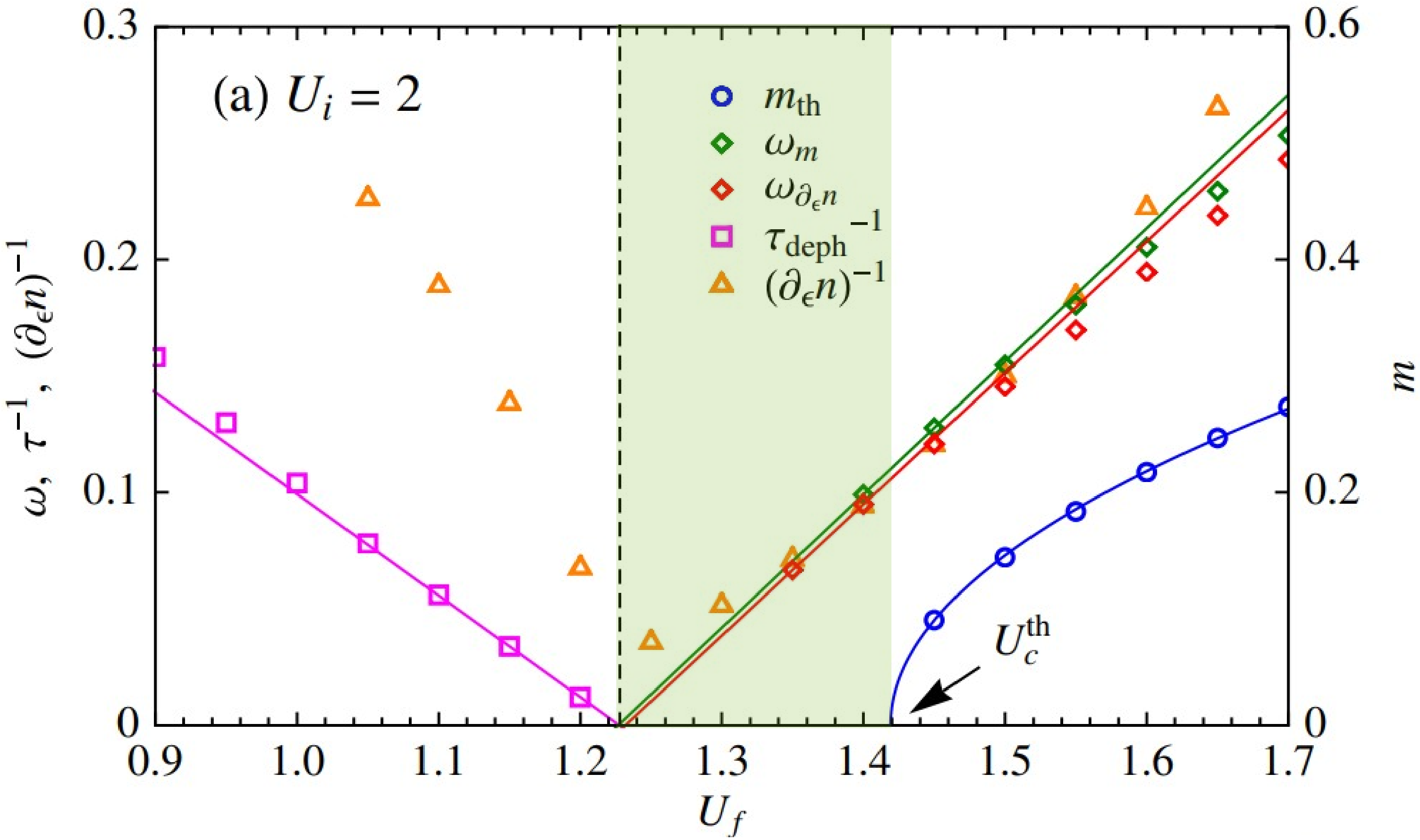
Tsuji, N. et al.: Phys. Rev. Lett. 110, 136404 (2013)

Half-filled Hubbard model on infinite bethe lattice

Nonthermal critical region



Nonthermal critical region



Tsuji, N. et al.: Phys. Rev. Lett. 110, 136404 (2013)

DMFT self-consistency

Bipartite lattice

A given time-profile for *U* is imposed:

$$U(t) = U_{\text{init}} + (U_{\text{final}} - U_{\text{init}}) \sin^2\left(\frac{t}{t_{\text{quench}}}\frac{\pi}{2}\right), 0 \leq t \leq t_{\text{quench}}$$

DMFT self-consistency :

$$m(t) = \frac{1}{2}[n_{A,\uparrow}(t) - n_{B,\uparrow}(t)] = \frac{1}{2}[n_{B,\downarrow}(t) - n_{A,\downarrow}(t)]$$

$$[i\partial_t + \mu(t) - \xi_\alpha \sigma m(t) U(t)] G_{\alpha,\sigma}(t,t') - \int_{\mathcal{C}} d\bar{t}\, [\Delta_{\alpha,\sigma}(t,\bar{t}) + \Sigma_{\alpha,\sigma}(t,\bar{t})] G_{\alpha,\sigma}(\bar{t},t') = \delta(t,t')$$

$$\Delta_{\alpha,\sigma} = t_h^2 G_{\alpha,\bar{\sigma}}$$

$$\Sigma_{\alpha,\sigma} = U(t)U(t')G_{\alpha,\sigma}(t,t')G_{\alpha,\bar{\sigma}}(t',t)G_{\alpha,\bar{\sigma}}(t,t') \qquad \alpha \in \{A,B\} \qquad \sigma \in \{\uparrow,\downarrow\}$$

DMFT self-consistency

k-dependent case

A given time-profile for *U* is imposed:

$$U(t) = U_{\text{init}} + (U_{\text{final}} - U_{\text{init}}) \sin^2\left(\frac{t}{t_{\text{quench}}} \frac{\pi}{2}\right), 0 \leq t \leq t_{\text{quench}}$$

DMFT self-consistency :

$$m(t) = \frac{1}{2}[n_{A,\uparrow}(t) - n_{B,\uparrow}(t)] = \frac{1}{2}[n_{B,\downarrow}(t) - n_{A,\downarrow}(t)]$$

$$[i\partial_t + \mu - \hat{h}_\epsilon(t)]\hat{G}_{\epsilon,\sigma}(t,t') - \int_{\mathcal{C}} d\bar{t}\, \hat{\Sigma}_\sigma(t,\bar{t})\hat{G}_{\epsilon,\sigma}(\bar{t},t') = \delta(t,t')$$

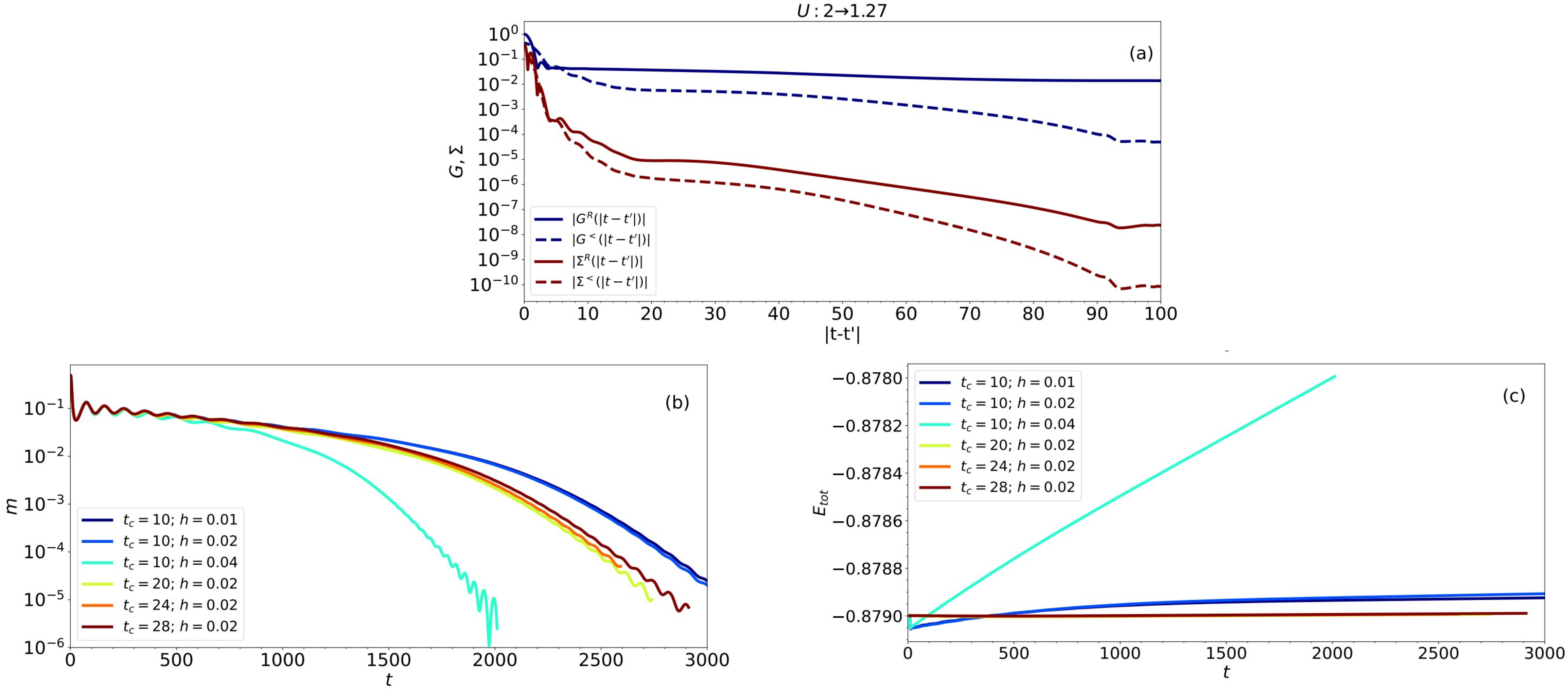
$$\hat{h}_\epsilon(t) = -Um(t)\hat{\tau}_z + \epsilon\hat{\tau}_x$$

$$\hat{\Sigma}_\sigma = \text{diag}(\Sigma_{A\sigma}, \Sigma_{B\sigma})$$

$$\Sigma_{\alpha,\sigma} = U(t)U(t')G_{\alpha,\sigma}(t,t')G_{\alpha,\bar{\sigma}}(t',t)G_{\alpha,\bar{\sigma}}(t,t') \qquad \alpha \in \{A,B\} \qquad \sigma \in \{\uparrow,\downarrow\}$$

The decay of the correlation functions

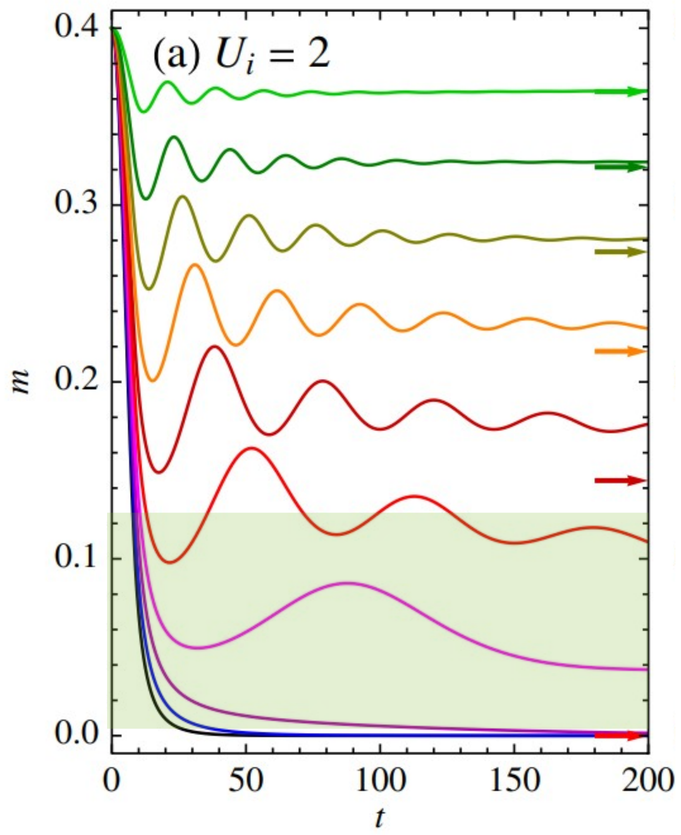
The energy conservation



Picano, A. et al.: Phys. Rev. B 103, 165118 (2021)

Decay of the staggered magnetization

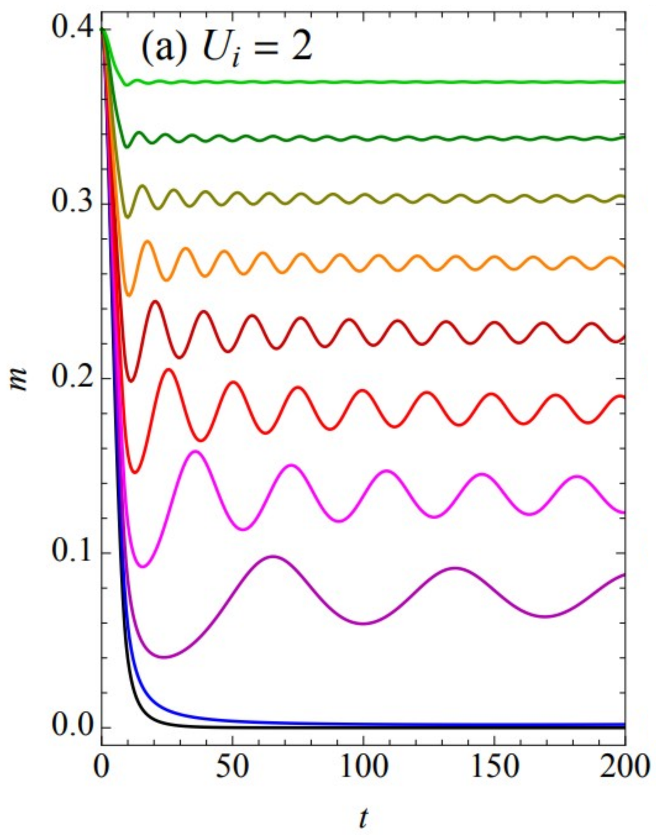
Third-order weak coupling expansion vs mean-field theory



$U_f = 1.4$

$U_f = 1.3$

$U_f = 1.2$



$U_f = 1.4$

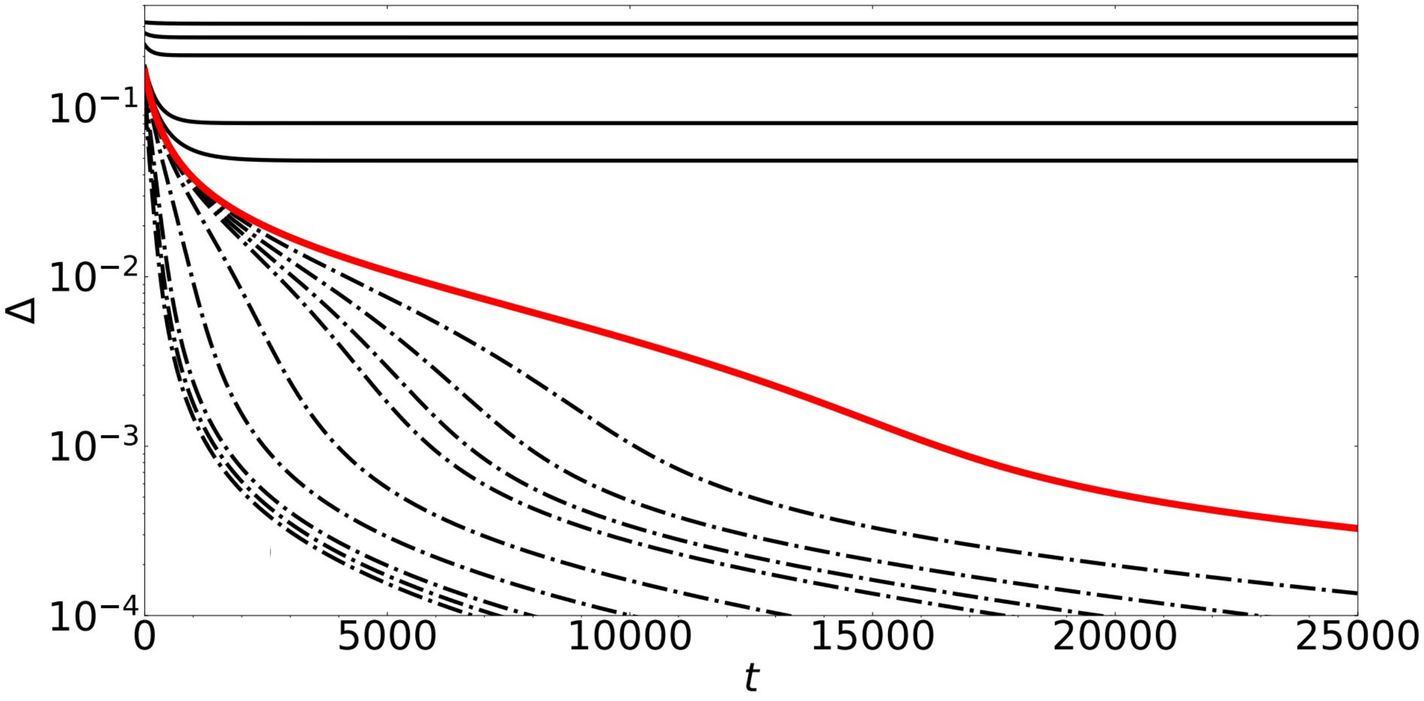
$U_f = 1.3$

$U_f = 1.2$

Tsuji, N. et al.: Phys. Rev. Lett. 110, 136404 (2013)

Mean-field solution

Does not predict the accelerated gap collapse



Picano, A. et al.: Phys. Rev. B 103, 165118 (2021)

Quantum Boltzmann equation

Quasi-Particle Quantum Boltzmann Equation

Time-evolution of the quasi-particle distribution function

$$\partial_t f + v_r \cdot \nabla_r f + v_k \cdot \nabla_k f = \big(\partial_t f\big)_{coll}$$

Common assumptions:

1) The existence of quasi-particles sharply peaked at energies $\omega = \epsilon_k$:

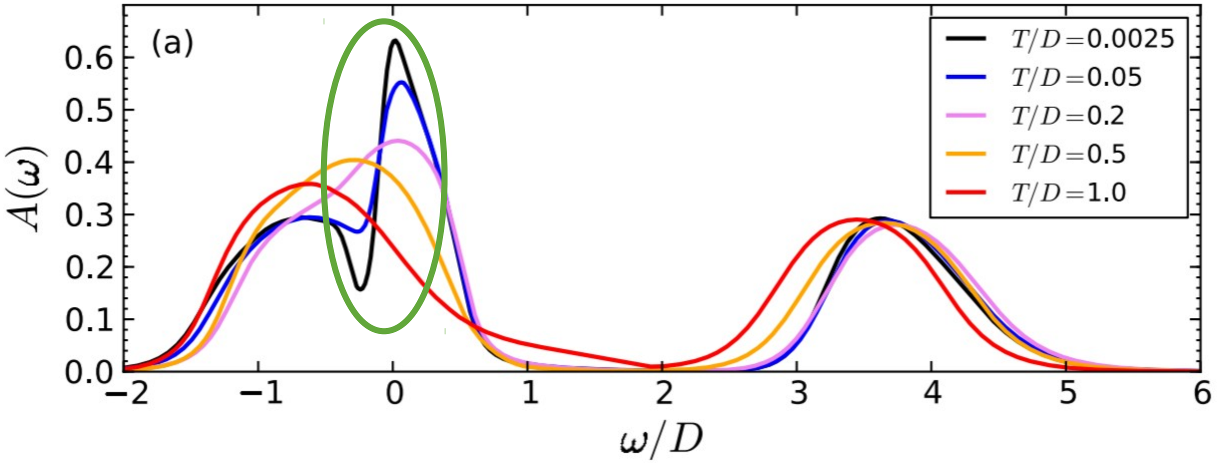
$$f(k) = \frac{1}{e^{\beta(\epsilon_k - \mu)} + 1}$$

2) Perturbative approximation in the evaluation of the collision term (e.g., Fermi golden rule)

Quasi-Particle Quantum Boltzmann Equation

The limits of the two assumptions

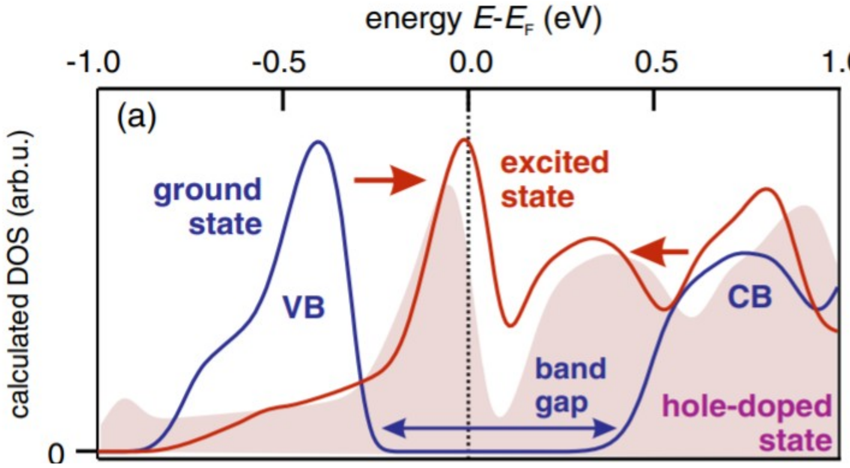
1) The existence of quasi-particles is not always justified



As temperature increases, the QP can be barely resolved

Phys. Rev. Lett. 110, 086401 (2013)

2) The DOS can depend strongy on the distribution



Phys. Rev. Lett. 113, 216401 (2014)

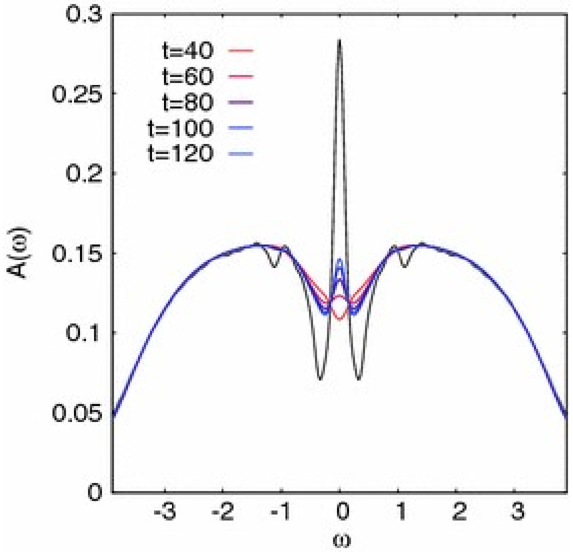
Separation of timescales

The main assunption of QBE

$$\delta t \gg 1/\delta\omega$$

δt sets the scale for the time evolution of the system

$\delta\omega$ is given by the line-width of relevant spectral features



M. Eckstein (2018), Springer Series in Solid-State Sciences

Non-perturbative QBE

Non-equilibrium steady-state DMFT loop

$F_{\mathbf{k}}(t,\omega)$ →

1.

$$G^R_{\boldsymbol{k}}(t,\omega) = [\omega + i0^+ + \mu - h_{\boldsymbol{k}}(t) - \Sigma^R(t,\omega)]^{-1}$$
$$G^<_{\boldsymbol{k}}(t,\omega) = F_{\boldsymbol{k}}(t,\omega)[G^A_{\boldsymbol{k}}(t,\omega) - G^R_{\boldsymbol{k}}(t,\omega)]$$

2.

$$\bar{G}^{R,<}(\omega) = \sum_{\boldsymbol{k}} G^{R,<}_{\boldsymbol{k}}(t,\omega).$$

3. NESS impurity problem:

$$\mathcal{G}^R_0(\omega) = [\bar{G}^R(\omega)^{-1} + \bar{\Sigma}^R(\omega)]^{-1},$$
$$\Delta^<(\omega) = \bar{G}^R(\omega)^{-1}\bar{G}^<(\omega)\bar{G}^A(\omega)^{-1} - \bar{\Sigma}^<(\omega)$$
$$\mathcal{G}^<_0(\omega) = \mathcal{G}^R_0(\omega)\Delta^<\mathcal{G}^A_0(\omega),$$

where $\bar{\Sigma}^{R,<}(\omega) = \Sigma^{R,<}(t,\omega)$.

4. Calculate new $\Sigma[\mathcal{G}]$ by using IPT.

5. Iterate Step 1. to 4. until convergence.

→ $\Sigma(t,\omega), \mathcal{A}_{\mathbf{k}}(t,\omega)$

↓

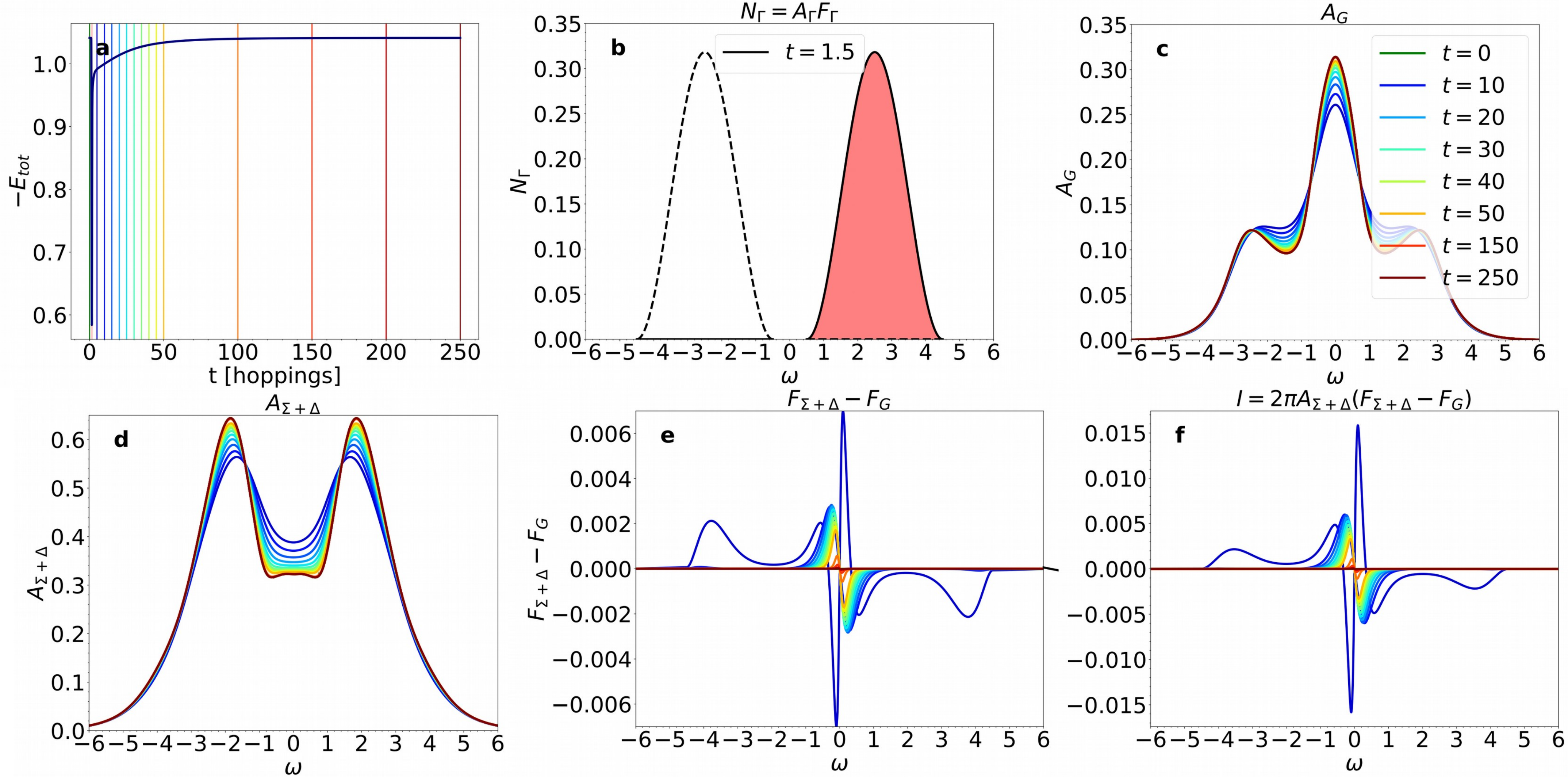
$$\partial_t F_{\mathbf{k}}(t,\omega) = I_{\mathbf{k}}(\Sigma, \mathcal{A}_k, F_k)$$

→ $F_{\mathbf{k}}(t+h,\omega)$

Non-equilibrium steady-state DMFT loop

The scattering integral

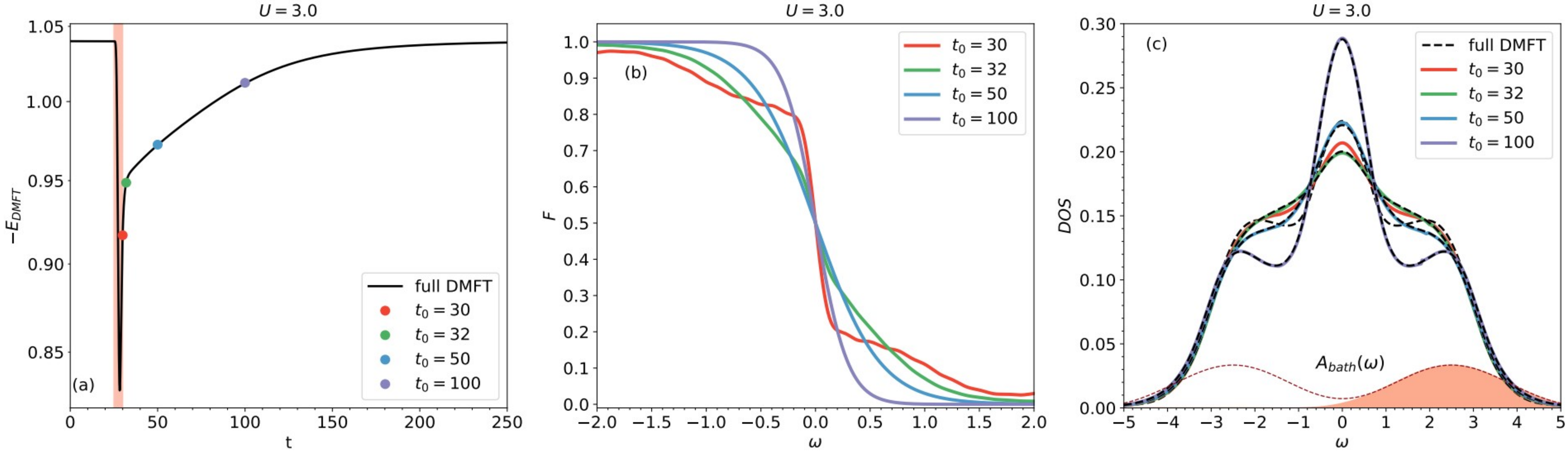
The distribution function of the self-consistent bath



The auxiliary steady-state representation of the spectra

Comparing the spectrum from NESS with the full DMFT one

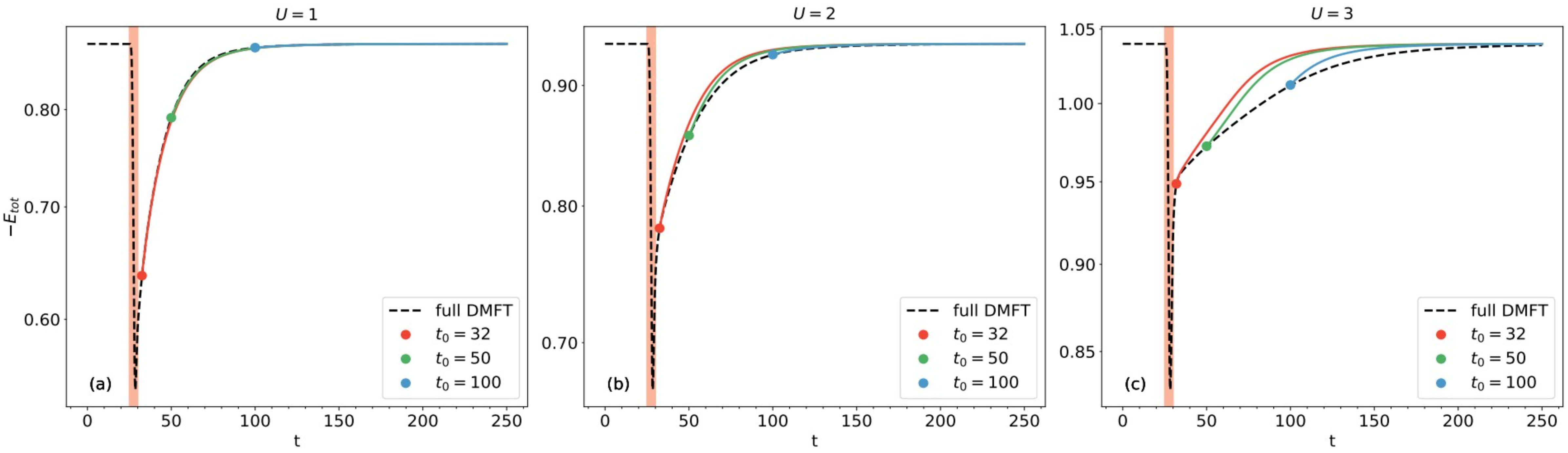
$$\mathcal{A}(\omega, t_0) \overset{?}{=} \mathcal{A}_\omega^{\text{NESS}}[F(t_0)]$$



The DOS can be very accurately obtained as a steady-state functional of the distribution function F

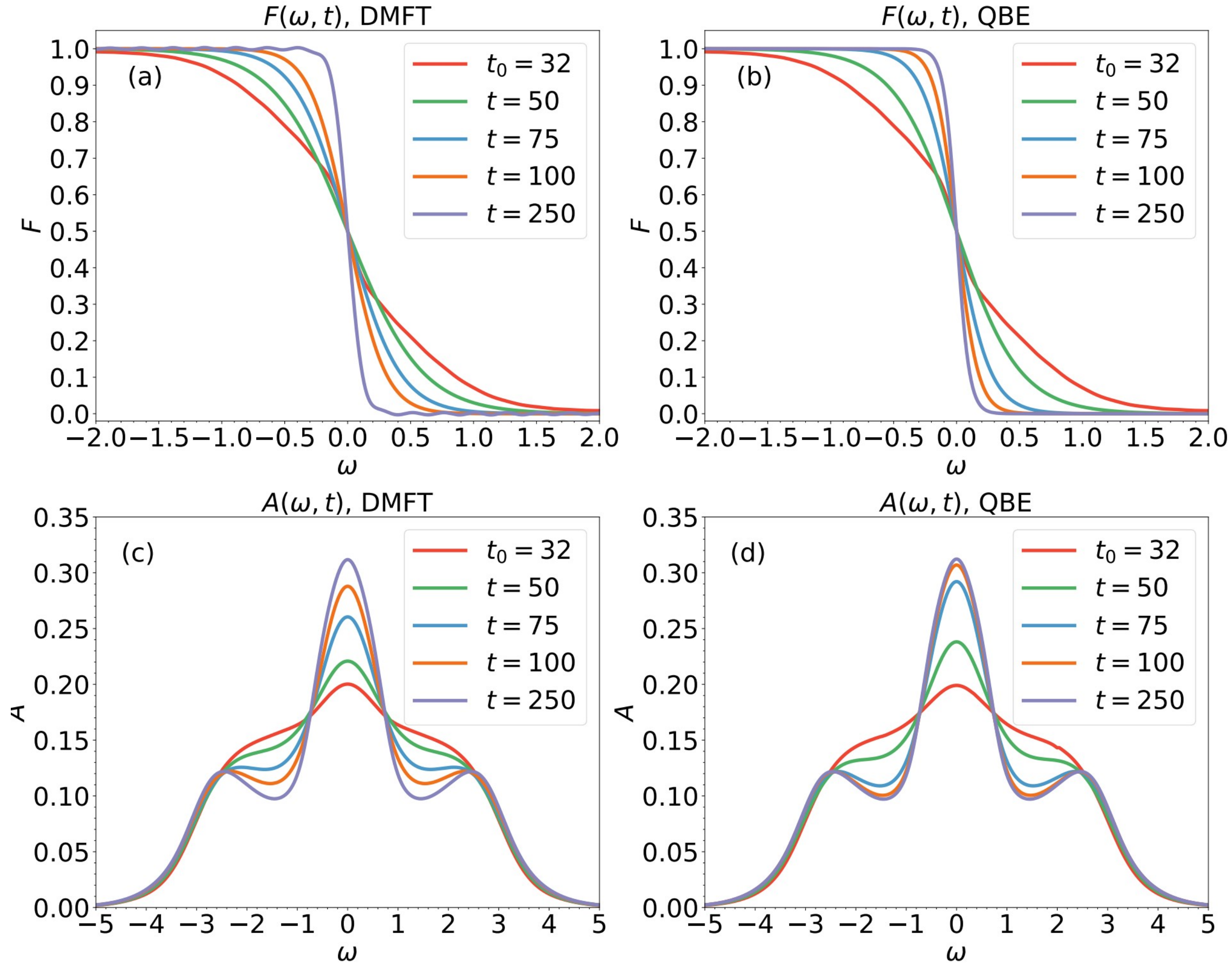
Time evolution of the total energy

Comparing the relaxation dynamics from QBE with the one from full DMFT



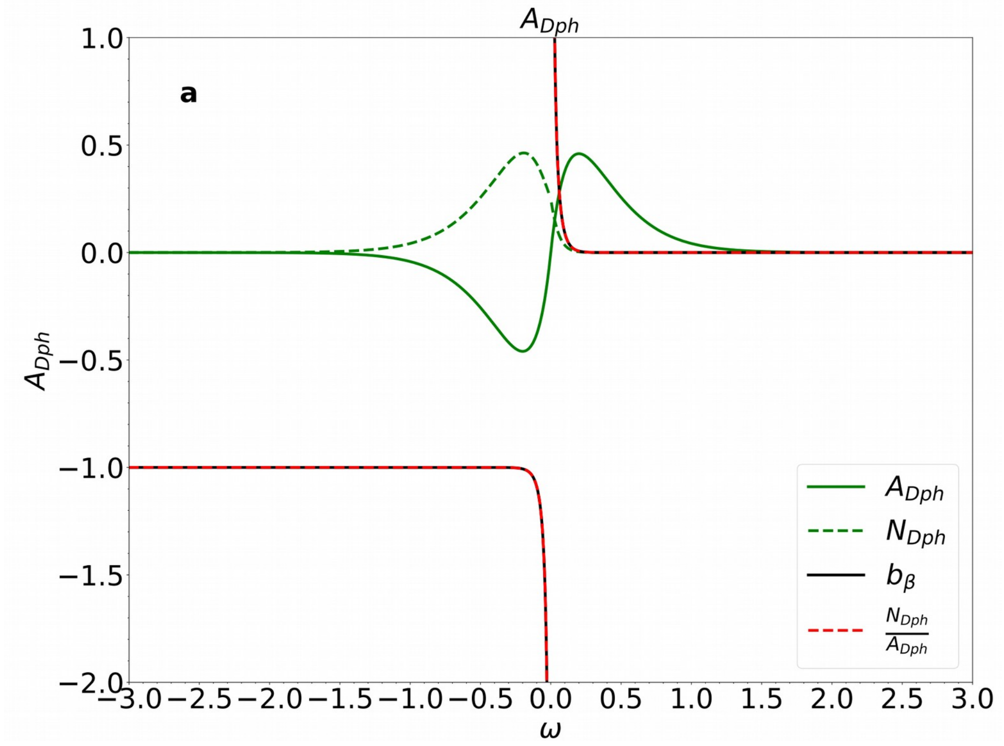
Distribution function and spectral function

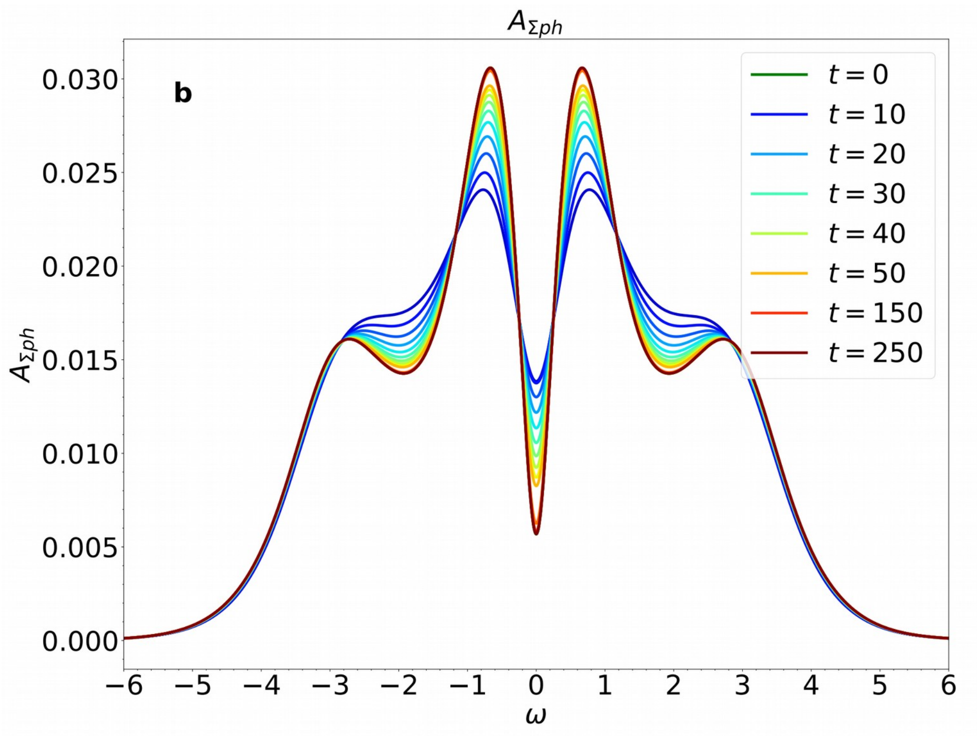
Comparing the results from QBE with the ones from full DMFT



Self-energy of the bosonic bath

The bath is always at equilibrium at temperature T





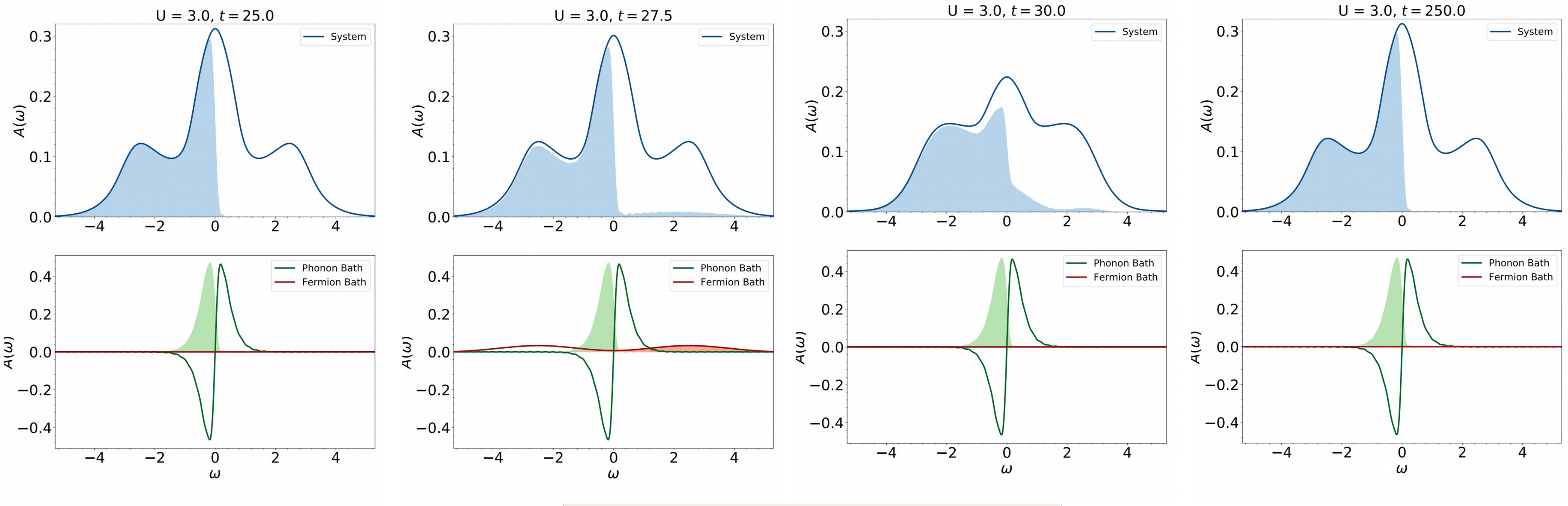
$$\Sigma_{\text{ph}}(t,t') = i g_{\text{ph}}^2 G(t,t') D_{\text{ph}}(t,t')$$

$$A_{\text{D-ph}}(\omega) = \begin{cases} J(\omega), \forall \omega > 0 \\ -J(-\omega) \forall \omega < 0 \end{cases}$$

$$J(\omega) = \omega / (4\omega_{\text{ph}}^2) e^{-\omega/\omega_{ph}}, \forall \omega > 0$$

Fermionic bath at negative temperature

How photo-excitation is simulated



$$\Gamma(t,t') = V(t)G_{\text{bath}}(t,t')V(t')^*$$

$$A_{\text{bath}}(\omega) = A(\omega - 2.5) + A(\omega + 2.5)$$

$$A(\omega) = \frac{1}{\pi} cos^2(\pi\omega/W_{\text{bath}})$$

$$f_{\text{bath}}(\omega) = f_{-\beta}(\omega)$$

Summary

01 Evaluation of the scattering integral in a non-perturbative way

02 Scattering integral calculated from an auxilary non-equilibrium steady-state impurity problem

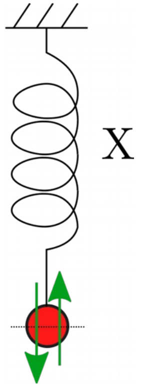
03 This allows the QBE to be combined with non-perturbative methods used in DMFT that allow to reproduce the non-equilibrium electronic structure after photoexcitation

Stochastic semiclassical equations

Impurity problem

On-site electrons interacting with a local vibrational mode

$$H_{\text{imp}} = H_{\text{x}} + H_{\text{cx}} + H_{\text{cc}}$$



$$\text{H}_{\text{x}} = \frac{1}{2}\left(\Omega^2 \hat{X}^2 + \hat{P}^2\right)$$

Free phonons

$$H_{\text{cx}} = \sqrt{2\Omega} g \hat{X} \left(\sum_{\sigma} n_{\sigma} - 1 \right)$$

Electron-phonon interaction

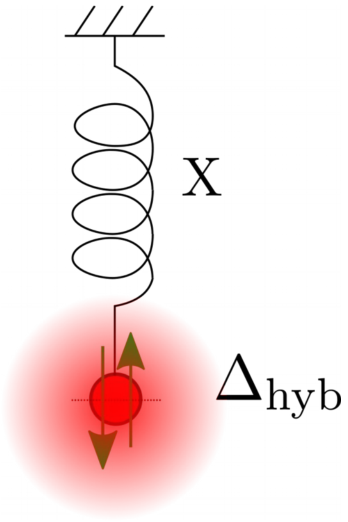
$$H_{\text{cc}} = -\mu \sum_{\sigma} n_{\sigma} + U n_{\uparrow} n_{\downarrow}$$

Purely electronic part of the Hamiltonian

Quantum impurity model

The Keldysh contour

$$H_{QI} = H_{\text{imp}} + H_{\text{res}} + H_{\text{coupl}}$$



$$S[\bar{c}, c, X] = S_x[X] + S_{cx}[\bar{c}, c, X] + S_{cc}[\bar{c}, c]$$

Quantum impurity model

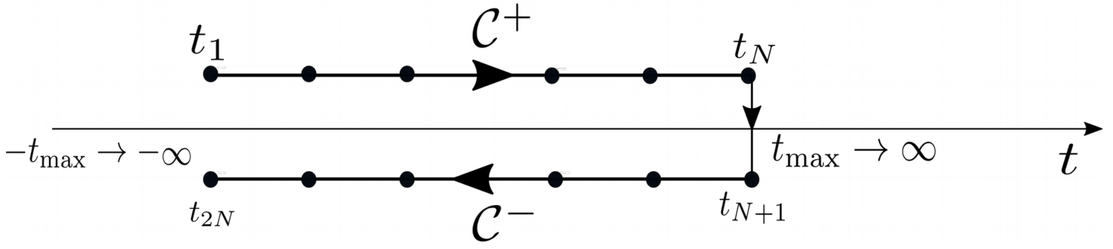
The action terms

$$S_x = \sum_{j=1}^{2N-1} \frac{h_{j+1}}{2}\left[\left(\frac{X_{j+1}-X_j}{h_{j+1}}\right)^2 - \Omega^2 X_j^2\right]$$

$$S_{cx} = -\sqrt{2\Omega}g \sum_{j=1}^{2N-1} h_{j+1}\, O_{j+1} X_j$$

$$S_{cc} = \sum_{j,j'=1}^{2N} \sum_{\sigma} \bar{c}_{j,\sigma} G^{-1}_{jj',\sigma} c_{j',\sigma}$$

$\mathcal{C} = \mathcal{C}^+ \cup \mathcal{C}^-$



S_{cc} is purely electronic and incorporates, e.g., Δ_{hyb} that comes from $H_{\text{res}} + H_{\text{coupl}}$

In continuous time:

$$S_{cc} = \int_{\mathcal{C}} \sum_{\sigma} dt\, dt' \bar{c}_\sigma(t) G_\sigma^{-1}(t,t') c_\sigma(t') \qquad G_\sigma^{-1}(t,t') = (i\partial_t + \mu)\delta_{\mathcal{C}}(t,t') - \Delta_{\text{hyb},\sigma}(t,t')$$

The effective action for the phonons

It is obtained by integrating out the electrons

From the total action one can define the partition function:

$$Z = \int \mathcal{D}[X] \int \mathcal{D}[\bar{c}, c]\, e^{iS[\bar{c},c,X]}$$

The effective action for the phonons is obtained by integrating out the electrons:

$$\mathrm{e}^{iS_{\mathrm{eff}}[X]} = e^{iS_x[X] + i\Gamma[X]}$$

$$\Gamma[X] = -i \log \left\langle e^{iS_{cx}[\bar{c},c,X]} \right\rangle_{cc}$$

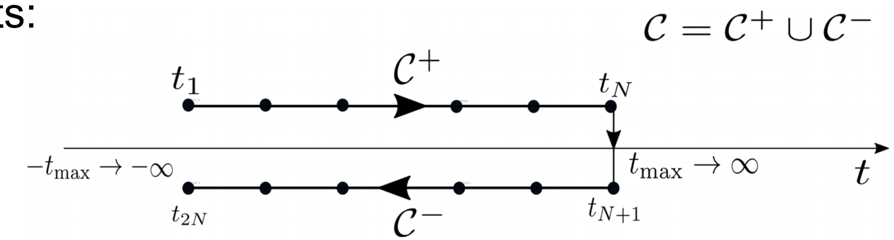
$$\langle \cdots \rangle_{cc} = \frac{1}{Z_{cc}} \int \mathcal{D}[\bar{c}, c] e^{iS_{cc}[\bar{c},c]} \ldots$$

Classical and quantum phonon components

The action terms written in the new basis

X_j is represented in terms of the so-called classical and quantum components:

$$\begin{pmatrix} X_j^{\mathrm{cl}} \\ X_j^{\mathrm{q}} \end{pmatrix} = \frac{1}{2} \begin{pmatrix} X_j^+ + X_j^- \\ X_j^+ - X_j^- \end{pmatrix}$$



In this basis, $S_c, S_{cx}, \Gamma[X]$ become:

$$S_x = -2\delta_t \sum_{j=2}^{N-1} X_j^{\mathrm{q}} \left(\ddot{X}_j^{\mathrm{cl}} + \Omega^2 X_j^{\mathrm{cl}} \right) + b.t.$$

$$S_{cx} = -\delta_t \sqrt{2\Omega} g \sum_{j=2}^{N-1} \left(O_{j+1}^+ - O_{j-1}^- \right) X_j^{\mathrm{cl}} - \delta_t \sqrt{2\Omega} g \sum_{j=2}^{N-1} \left(O_{j+1}^+ + O_{j-1}^- \right) X_j^{\mathrm{q}} + b.t.$$

$$\Gamma[X^{\mathrm{cl}}, X^{\mathrm{q}}] = \sum_{n=0}^{\infty} \frac{1}{n!} \sum_{j_1,\dots,j_n} X_{j_1}^{\mathrm{q}} \cdots X_{j_n}^{\mathrm{q}} \, \tilde{\Pi}_{j_1,\dots,j_n}$$

Where the effective potential Γ is expanded in a Taylor series in the quantum variable

The coefficients in the expansion of Γ

The connected correlation electronic functions

$$\tilde{\Pi}_{j_1,\dots,j_n} = \frac{-i\,\partial^n}{\partial X^{\mathrm{q}}_{j_1}\cdots\partial X^{\mathrm{q}}_{j_n}} \log\left\langle e^{iS_{cx}[\bar{c},c,X]}\right\rangle_{cc}\Big|_{X^{\mathrm{q}}=0}$$

To interpret the coefficients $\tilde{\Pi}$ we define the action:

$$S_{cl} = S_{cc} + S^{cl}_{cx} = S_{cc} - \delta_t\sqrt{2\Omega}g\sum_{j=2}^{N-1}\left(O^{+}_{j+1} - O^{-}_{j-1}\right)X^{\mathrm{cl}}_j + b.t.$$

This describes a purely electronic model where electrons at the impurity are subject to a fluctuating field:

$$\hat{H}_X(t) = \sqrt{2\Omega}gX^{\mathrm{cl}}(t)\,\hat{O}$$

$$\tilde{\Pi}_{j_1,\dots,j_n} \equiv (-i)^{n+1}(2\sqrt{2\Omega}g\delta_t)^n\langle\bar{O}_{j_1}\cdots\bar{O}_{j_n}\rangle^{\mathrm{con}}_{cl}$$

$\langle\cdots\rangle^{\mathrm{con}}_{cl}$ are the connected correlation functions for the electrons in the presence of the fluctuating force $\propto H_X(t)$

$$\tilde{\Pi}_j = -2\sqrt{2\Omega}g\delta_t\langle\bar{O}_j\rangle_{cl} \qquad \bar{O}_j = (O^{+}_{j+1} + O^{-}_{j-1})/2$$

$$\tilde{\Pi}_{j,l} = -\delta_t^2 4\Omega g^2(\chi^K_{cl})_{j,l} \qquad (\chi^K_{cl})_{j,l} = -2i\left(\langle\bar{O}_j\bar{O}_l\rangle_{cl} - \langle\bar{O}_j\rangle_{cl}\langle\bar{O}_l\rangle_{cl}\right)$$

The coefficients in the expansion of Γ

The connected correlation electronic functions

We expand the effective potential Γ in terms of the coefficients $\tilde{\Pi}$:

$$\Gamma = \Gamma^{(1)} + \Gamma^{(2)} + \cdots$$

$$\Gamma^{(1)} = \sum_{j=2}^{N-1} X_j^q \tilde{\Pi}_j = -2\sqrt{2\Omega} g \delta_t \sum_{j=2}^{N-1} \langle \bar{O}_j \rangle_{cl}$$

$$\Gamma^{(2)} = \frac{1}{2} \sum_{j,l=2}^{N-1} X_j^q X_l^q \tilde{\Pi}_{j,l} = -2\Omega g^2 \delta_t^2 \sum_{j,l=2}^{N-1} X_j^q X_l^q (\chi_{cl}^K)_{j,l}$$

Semiclassical approx.: truncate the expansion of Γ at <u>second order</u>

The effective equation for the phonons

Semiclassical approximation: expansion at second order in X^q

$$S_x + \Gamma^{(1)} = -2\delta_t \sum_{j=2}^{N-1} X_j^{\mathrm{q}} \left(\ddot{X}_j^{\mathrm{cl}} + \Omega^2 X_j^{\mathrm{cl}} + \sqrt{2\Omega} g \langle \bar{O}_j \rangle_{cl} \right)$$

$$e^{i\Gamma^{(2)}} = e^{-\frac{1}{2}\delta_t^2 4\Omega \sum_{j,l} X_j^{\mathrm{q}} \; (ig^2(\chi_{cl}^K)_{j,l}) \; X_l^{\mathrm{q}}} = \frac{1}{Z_\xi} \int \mathcal{D}[\xi] e^{-\frac{1}{2}\sum_j \xi_j A_{j,l} \xi_l + i2\delta_t \sqrt{\Omega} \sum_j \xi_j X_j^{\mathrm{q}}}$$

$$(A^{-1})_{j,l} = ig^2 (\chi_{cl}^K)_{j,l}$$

$$e^{i(S_x + \Gamma^{(1)} + \Gamma^{(2)})} = e^{-i2\delta_t X_j^{\mathrm{q}} F_j}$$

$$F_j = \ddot{X}_j^{\mathrm{cl}} + \Omega^2 X_j^{\mathrm{cl}} + \sqrt{2\Omega} g \langle \bar{O}_j \rangle_{cl} - \sqrt{\Omega} \xi_j$$

ξ can be viewed as a stochastic force whose statistics is determined by the matrix A, which itself depends on the trajectory X_j^{cl}

White noise limit $\tau_e \ll \frac{1}{\Omega}$

On the timescale of the phonons the noise from the electrons is delta-correlated

By choosing a Δt such that $\tau_e \ll \Delta t \ll 1/\Omega$:

$$\langle \xi(t)\xi(t')\rangle \to 0 \quad \text{for} \quad |t-t'| \gg \tau_e$$

In the limit $\tau_e \to 0$ the equations of motions become:

$$\mathrm{X}^{\mathrm{cl}}_{j+1} = X^{\mathrm{cl}}_j + \Delta t V_j$$

$$V_j = V_{j-1} + \Delta t F_{j-1}$$

$$F_j = -\Omega^2 X^{\mathrm{cl}}_j - g\sqrt{2\Omega}\langle \bar{O}_j\rangle_{cl} - \Omega\Gamma_j V_j + \sqrt{\Omega}\xi_j$$

$$\Gamma_j = -2g^2\partial_\omega \Im\chi^R_{cl}(t_j,\omega)|_{\omega=0}$$

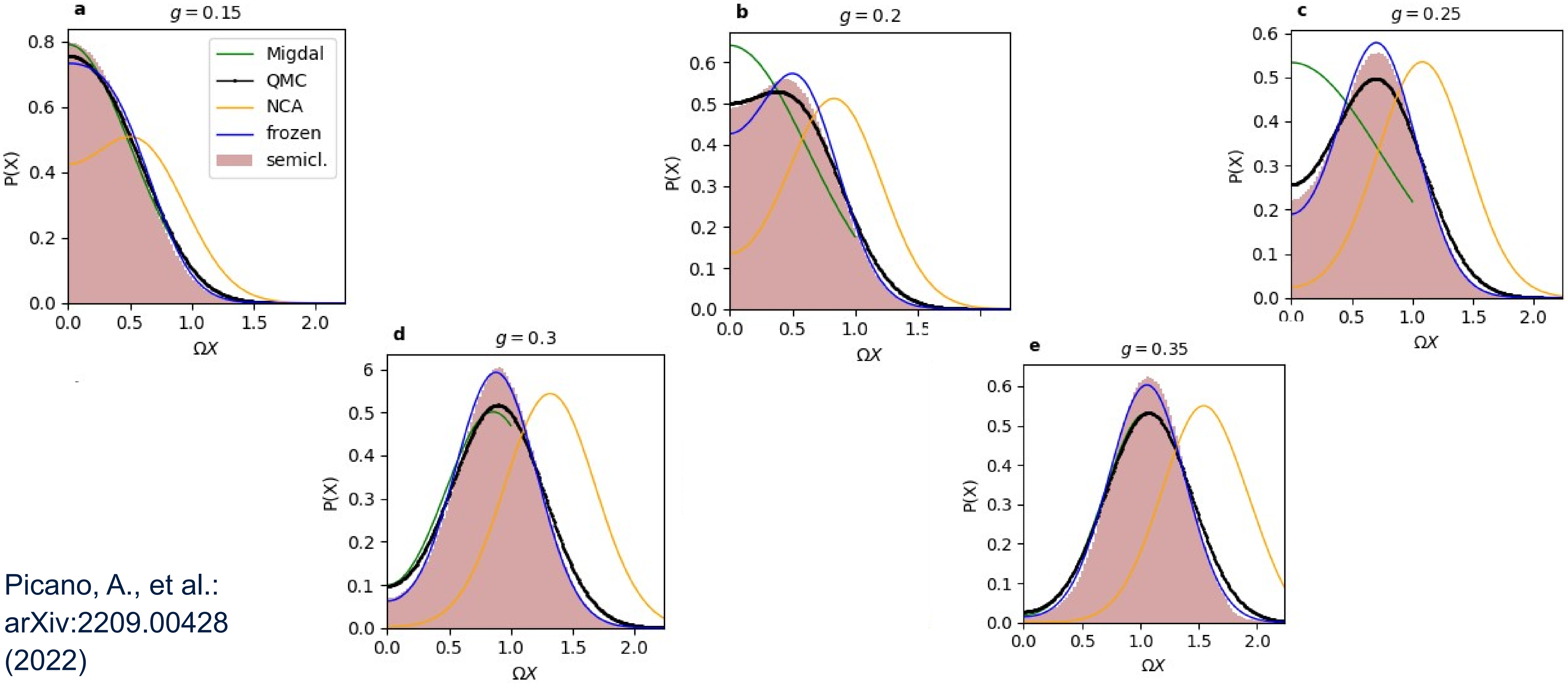
$$\langle \xi_j\rangle = 0,\ \langle \xi_j\xi_{j'}\rangle = K_j\delta_{j,j'}\Delta t^{-1} \qquad K_j = -g^2\Im\chi^K_{cl}(t_j,\omega)|_{\omega=0}$$

Fluctuation - dissipation theorem:

$$\chi^K_{cl}(\omega) = 2i\coth\left(\frac{\omega\beta}{2}\right)\Im\chi^R_{cl}(\omega) \quad \longleftrightarrow \quad K = 2\Gamma T$$

Comparison between semiclassical theory and QMC

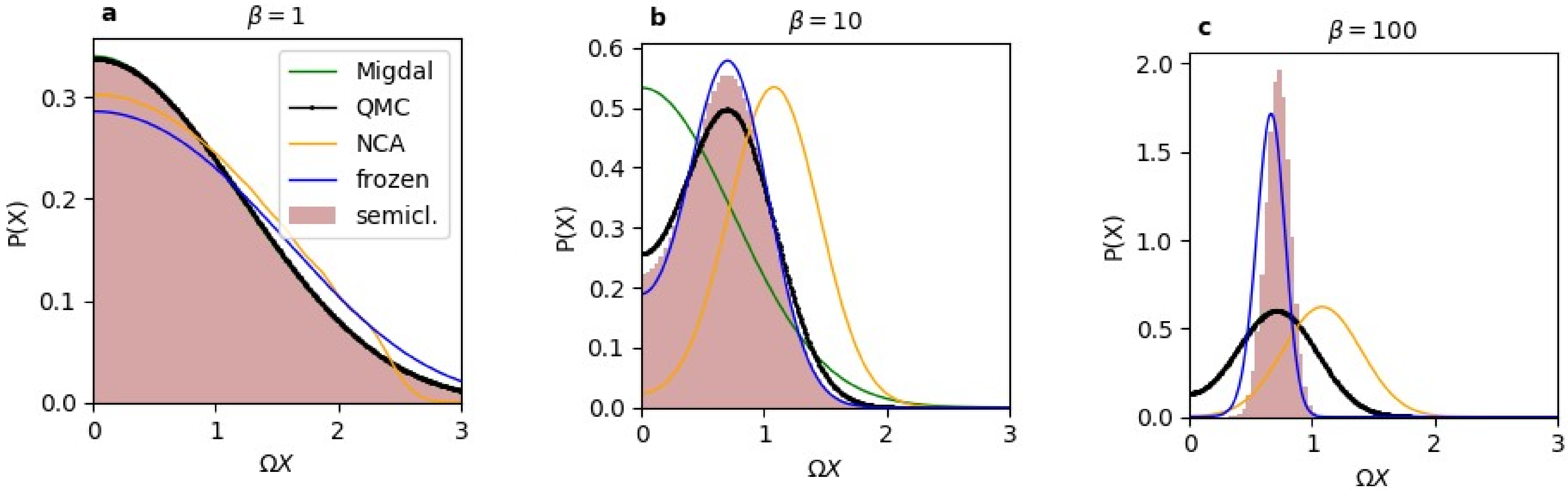
At high values of *g*, the semiclassical solution is nore localized with respect to QMC



Picano, A., et al.:
arXiv:2209.00428
(2022)

Comparison between semiclassical theory and QMC

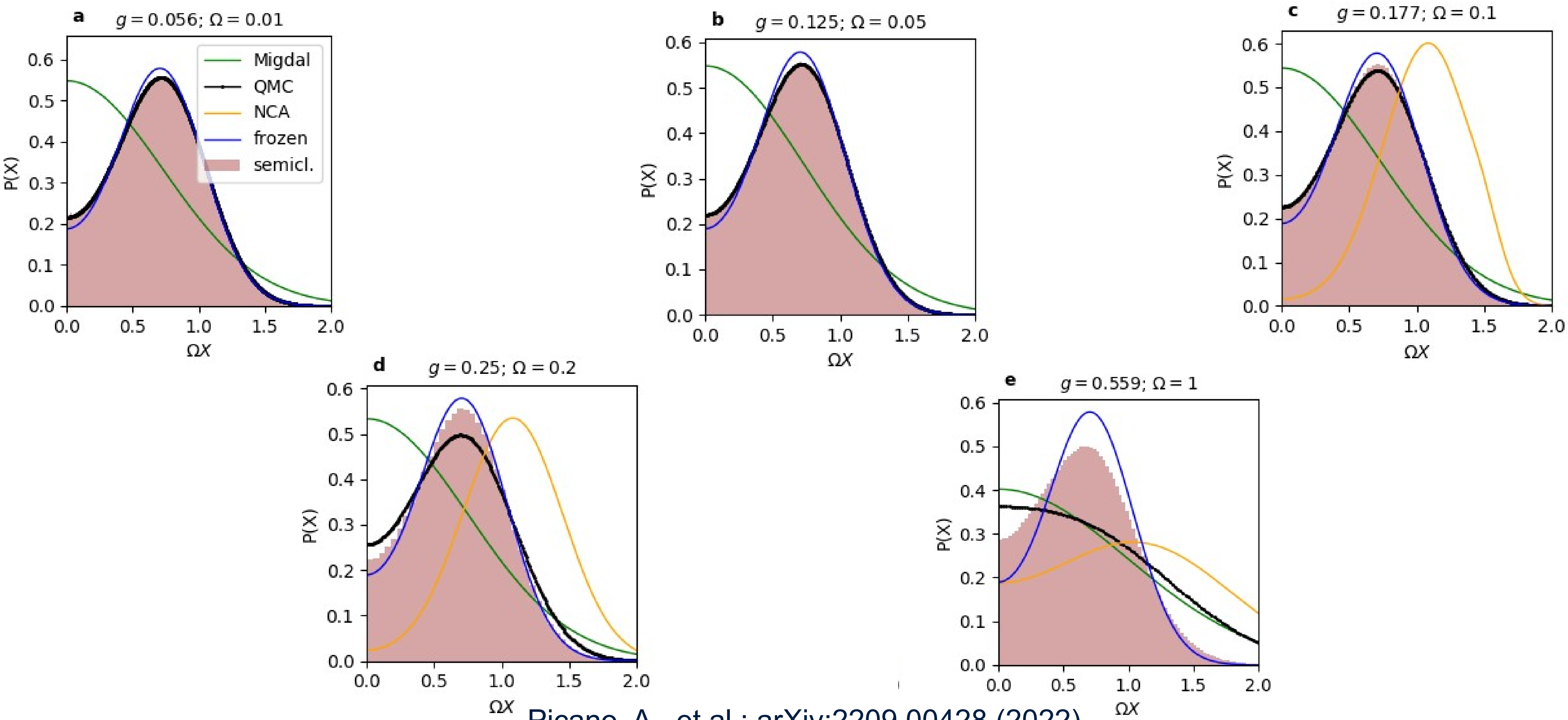
As the temperature decreases, the semiclassicla results deviate from QMC



Picano, A., et al.: arXiv:2209.00428 (2022)

Comparison between semiclassical theory and QMC

As the phonon frequency increases, the white noise approximation is less justified

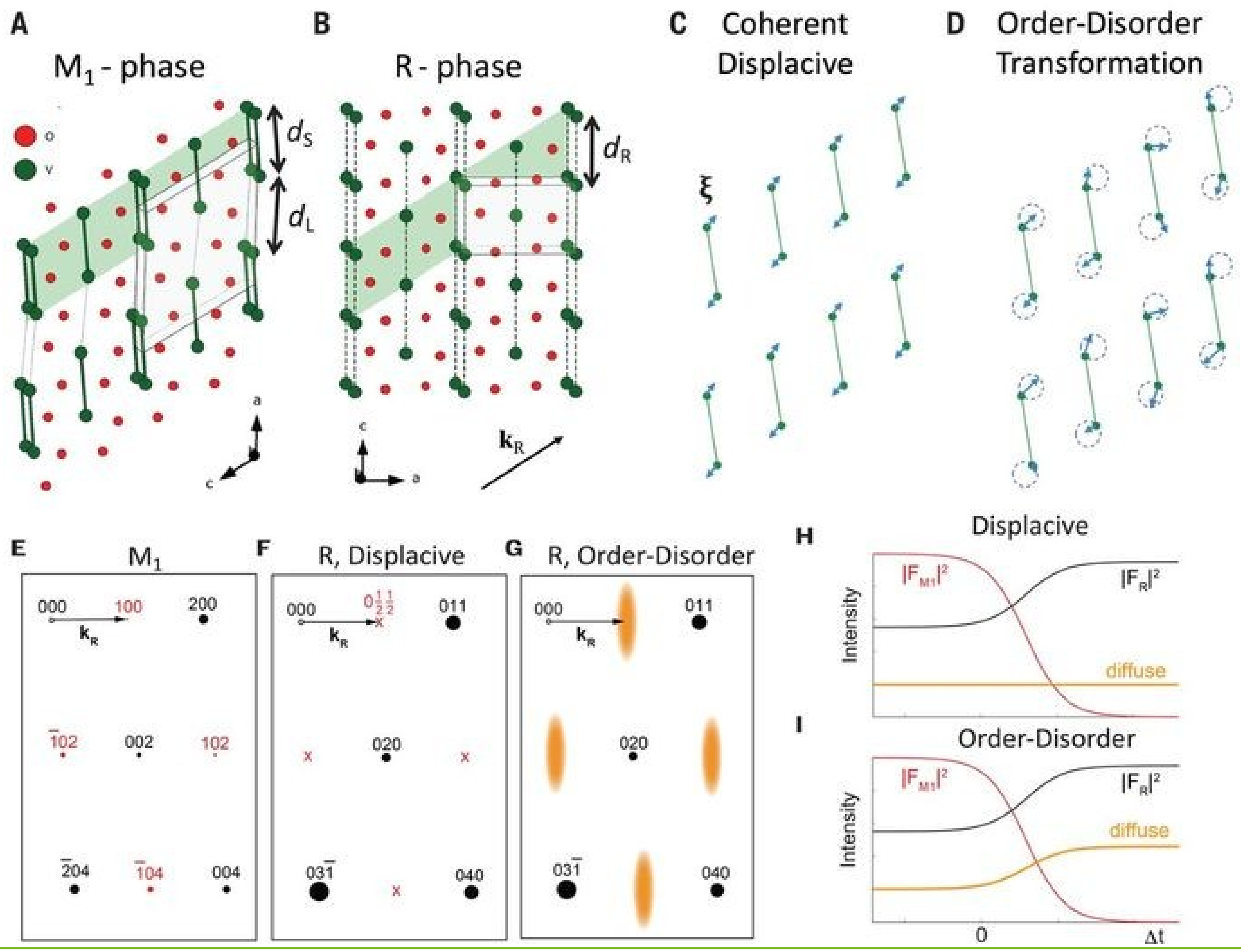


Picano, A., et al.: arXiv:2209.00428 (2022)

Inhomogeneous disordering

The case for VO_2

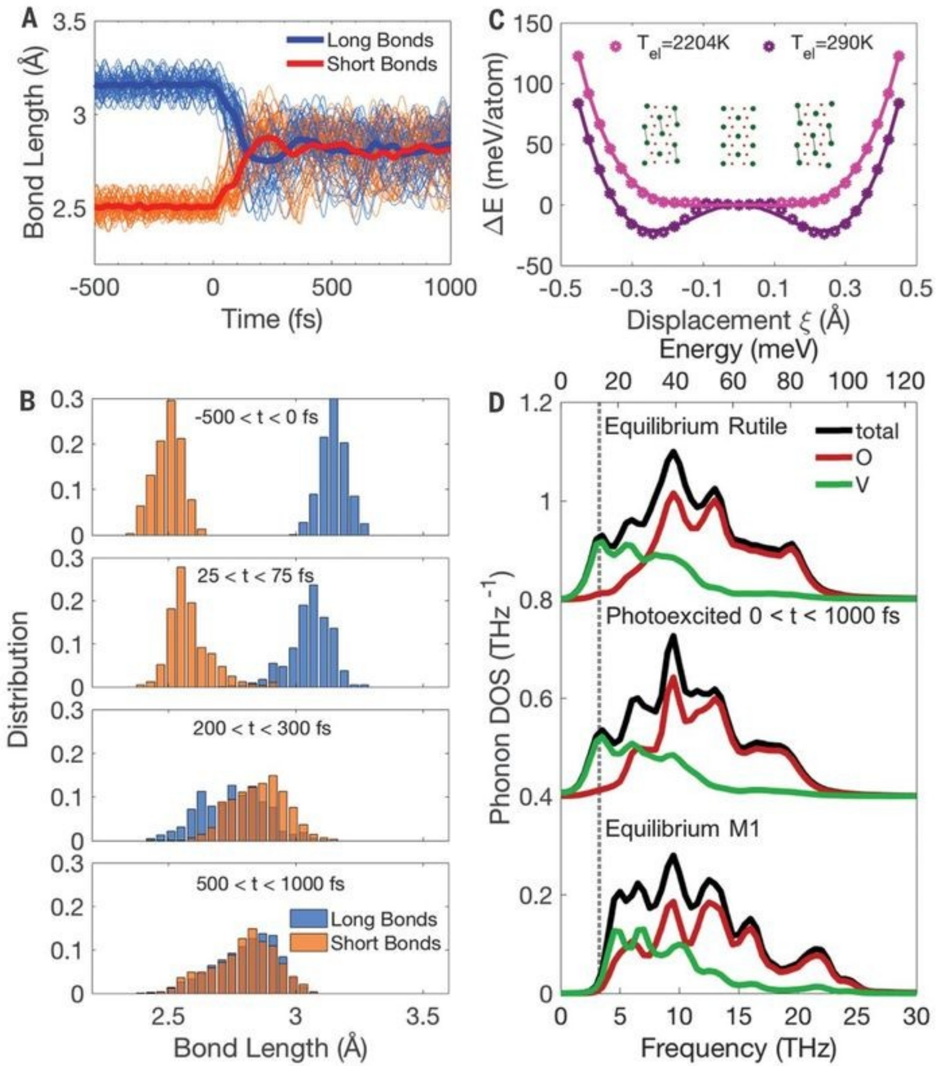
Displacive dynamics or order-disorder transition?



Wall, S. et al.: Science 362, 572 (2018)

The case for VO_2

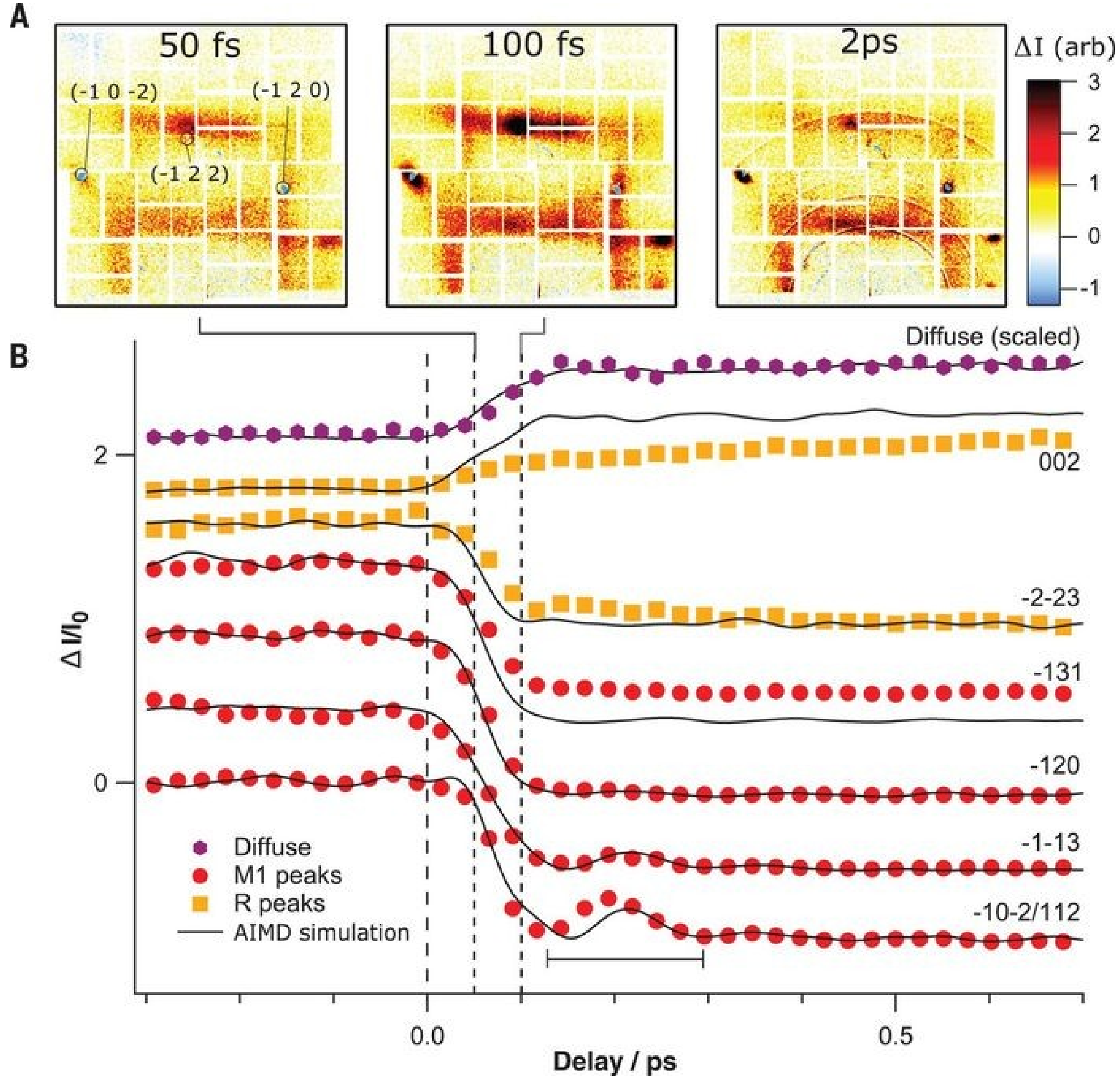
Displacive dynamics or order-disorder transition?



Wall, S. et al.: Science 362, 572 (2018)

The case for VO_2

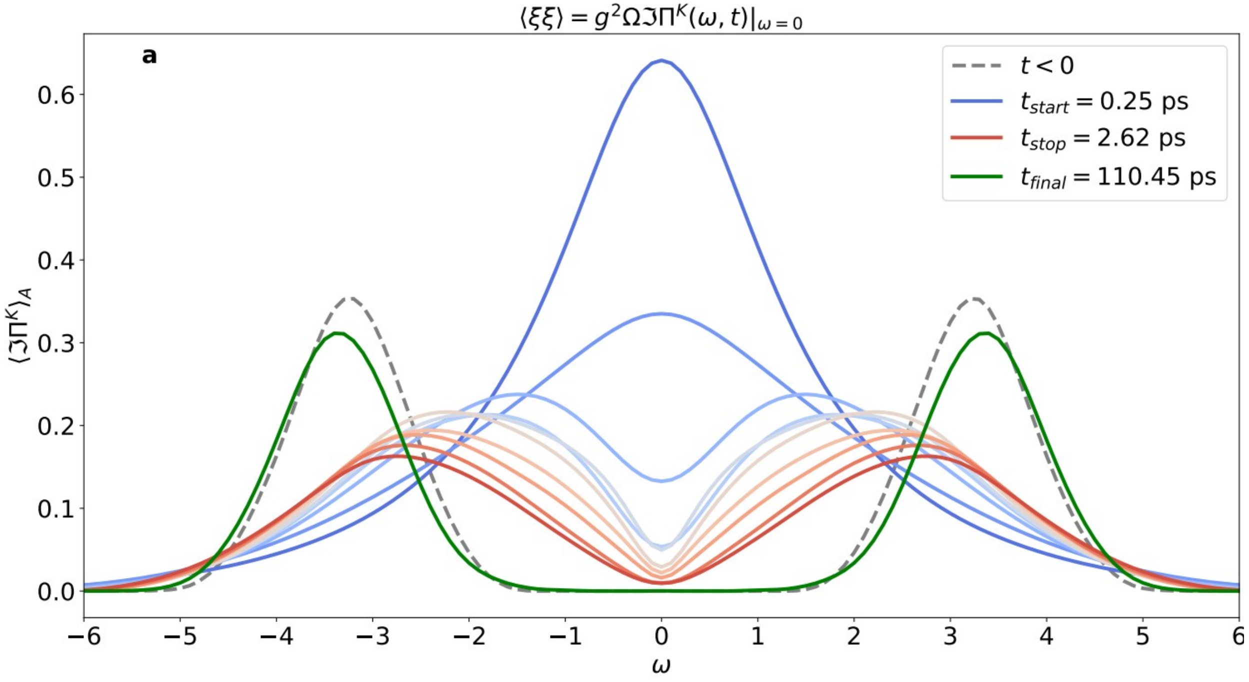
Displacive dynamics or order-disorder transition?

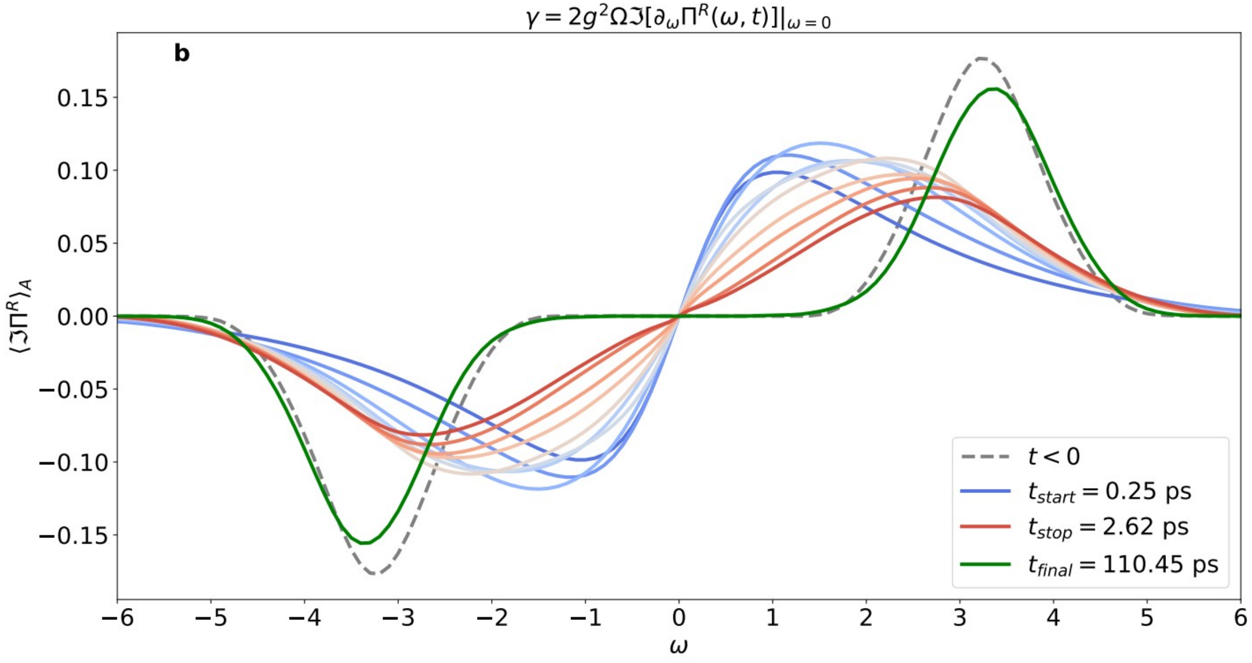


Wall, S. et al.: Science 362, 572 (2018)

Average connected electronic density correlation function

The variance of the Gaussian noise on the phonons and the damping

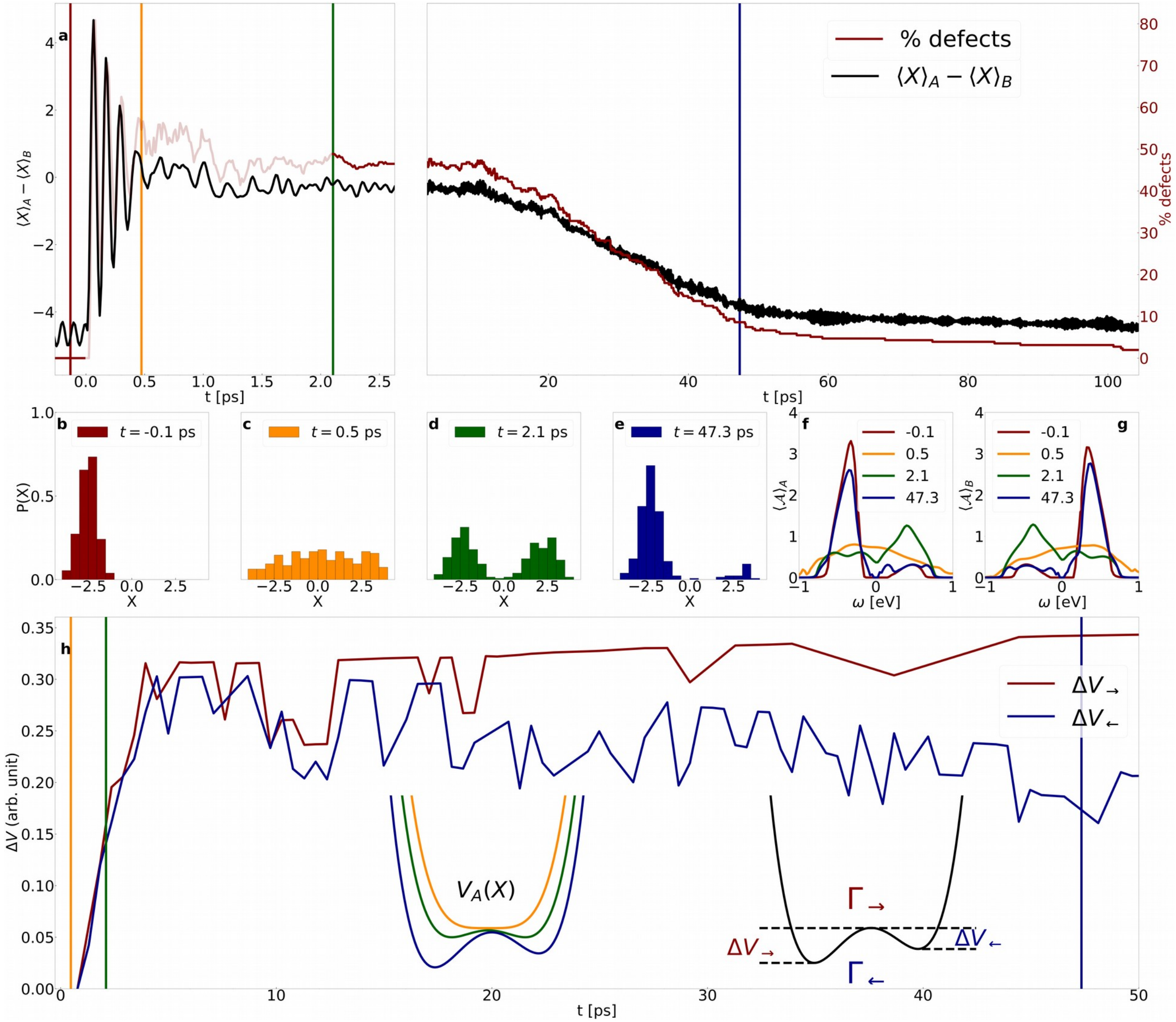




Picano, A. et al. : arXiv: 2112.15323 (2021)

The non-equilibrium dynamics

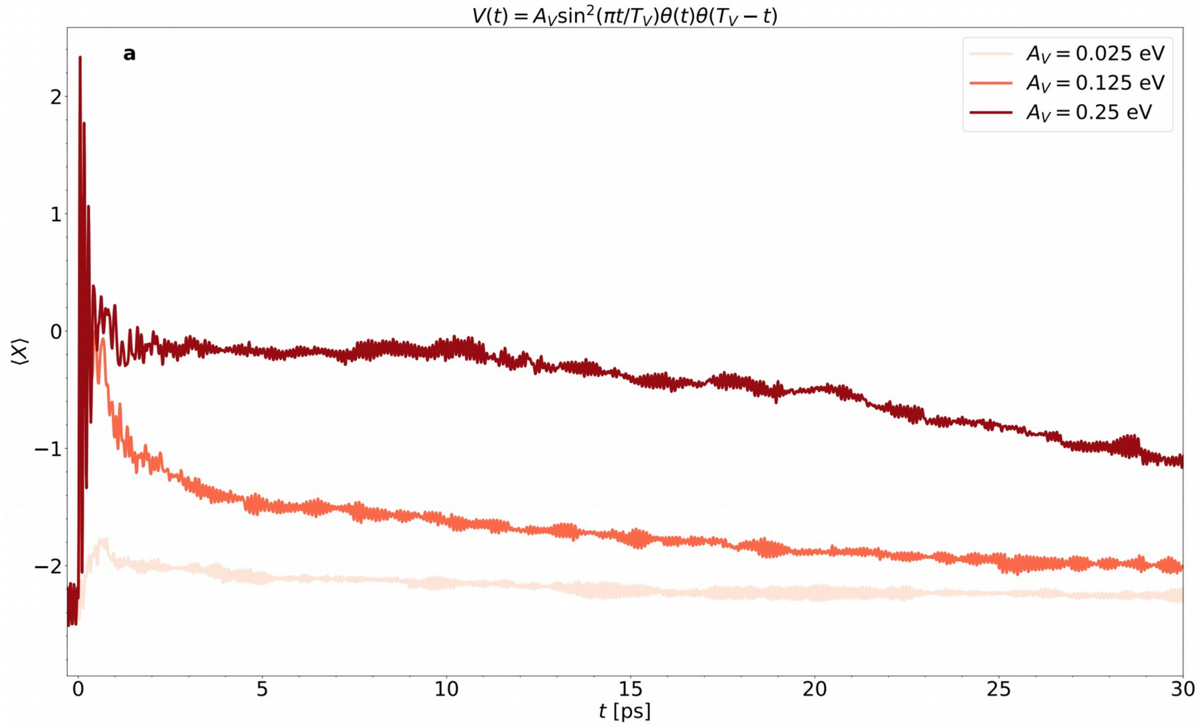
The potential energy barriers and the bimodal distribution

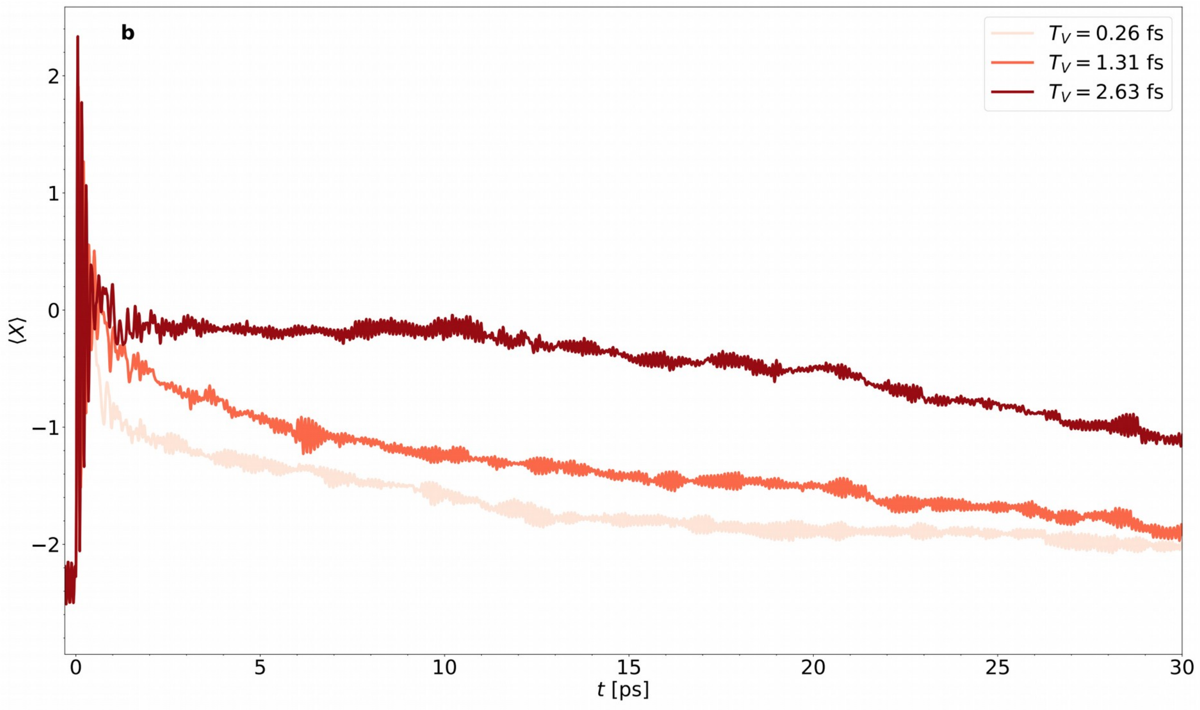


Picano, A. et al. : arXiv: 2112.15323 (2021)

Time evolution of the staggered order parameter

Different amplitudes of the driving field A_V and different durations T_V of the coupling with the fermionic bath

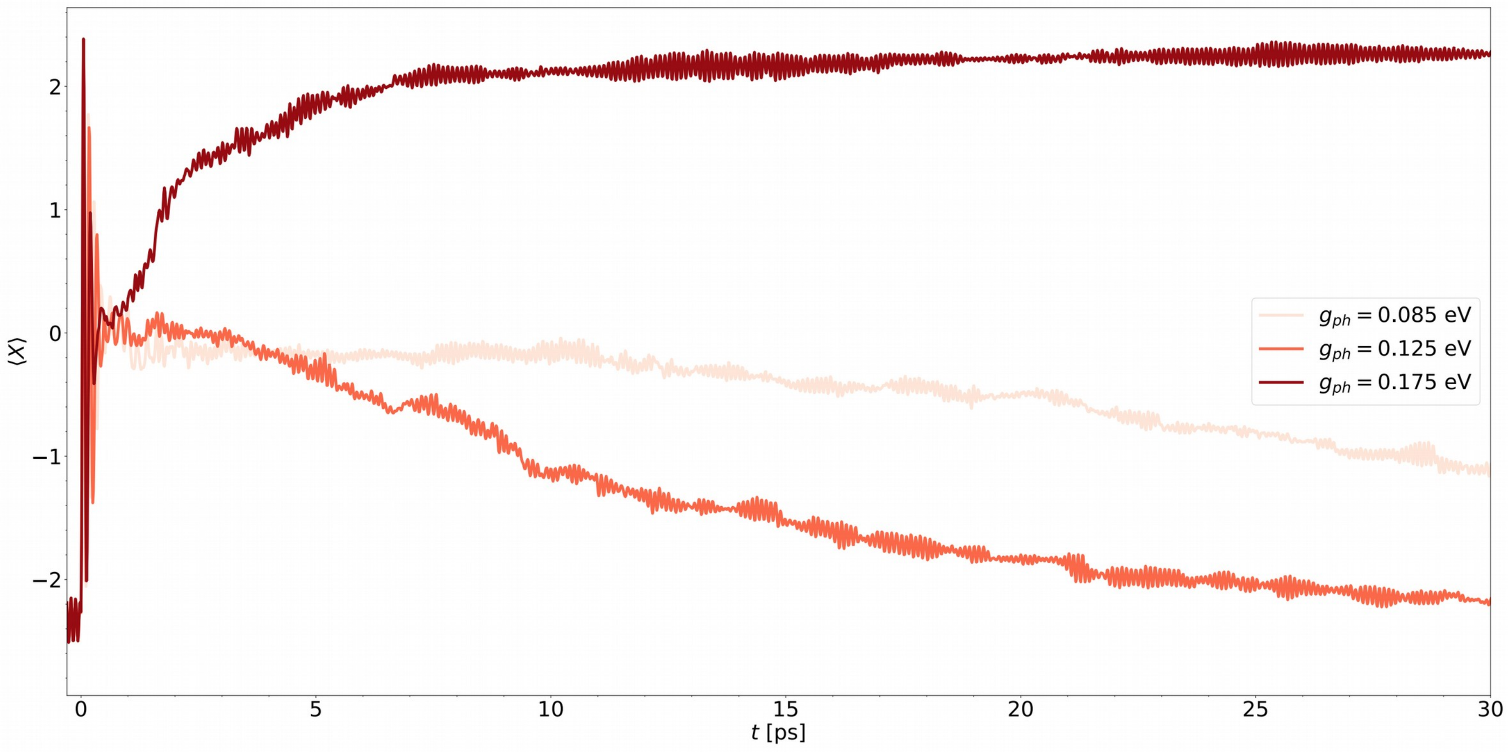




$$\Gamma(t,t') = V(t) G_{\text{bath}}(t,t') V(t')^*$$

Time evolution of the staggered order parameter

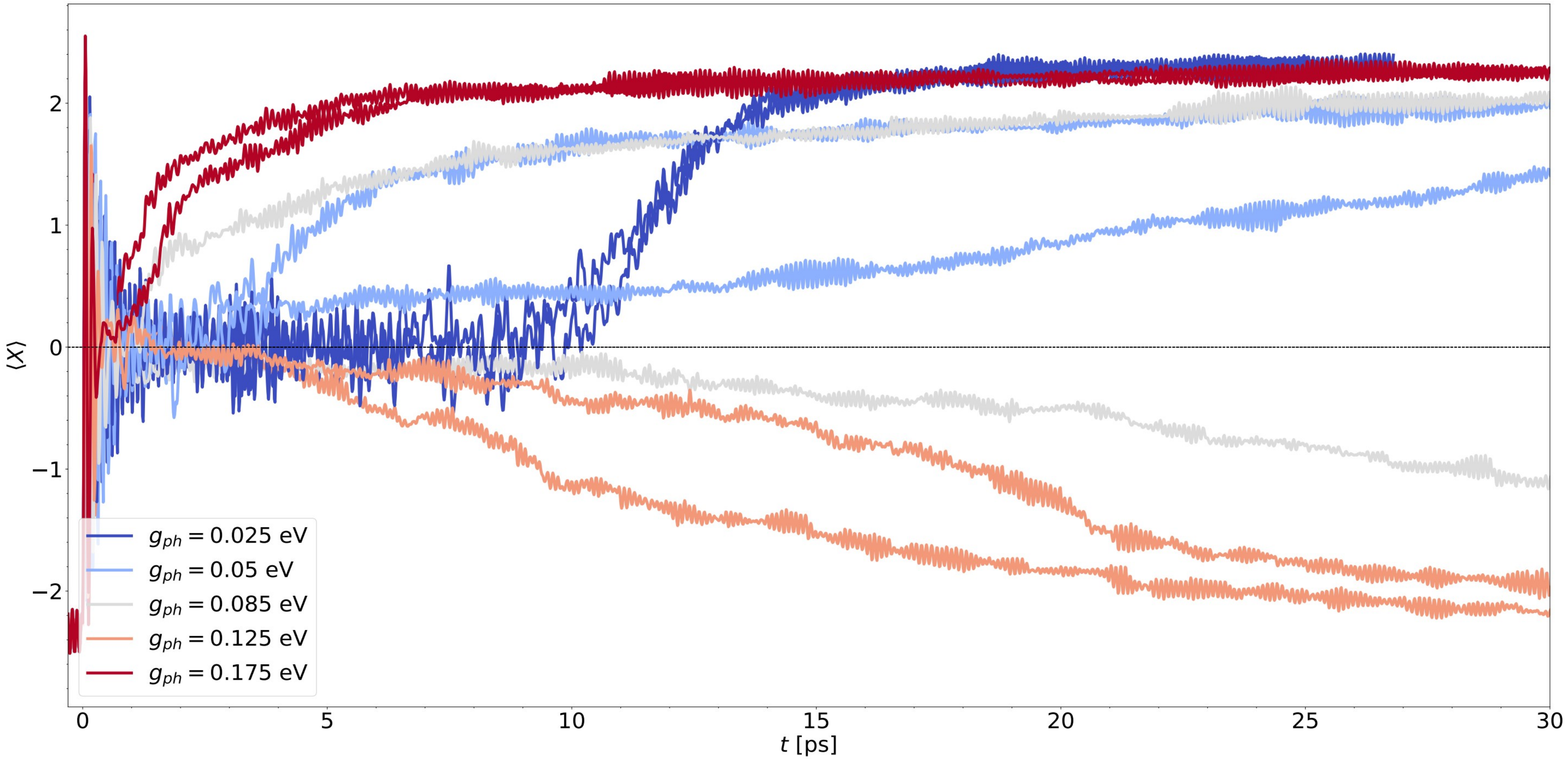
Different values of the coupling g_{ph} to the phononic bath



$$\Sigma_{\mathrm{ph}}(t,t') = i g_{\mathrm{ph}}^2 G(t,t') D_{\mathrm{ph}}(t,t')$$

Time evolution of the staggered order parameter

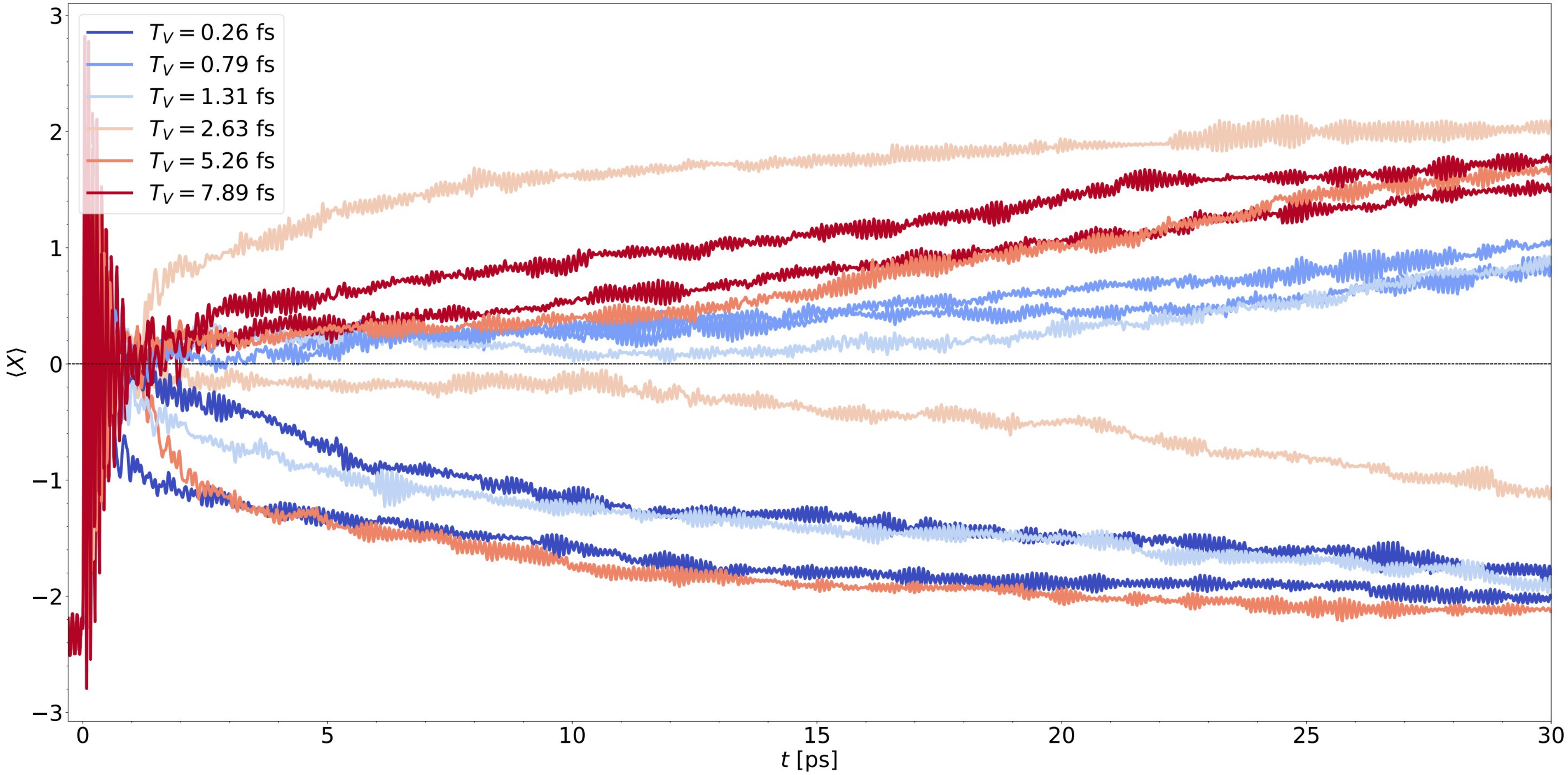
Different values of the coupling g_{ph} to the phononic bath



$$\Sigma_{\rm ph}(t,t') = i g_{\rm ph}^2 G(t,t') D_{\rm ph}(t,t')$$

Time evolution of the staggered order parameter

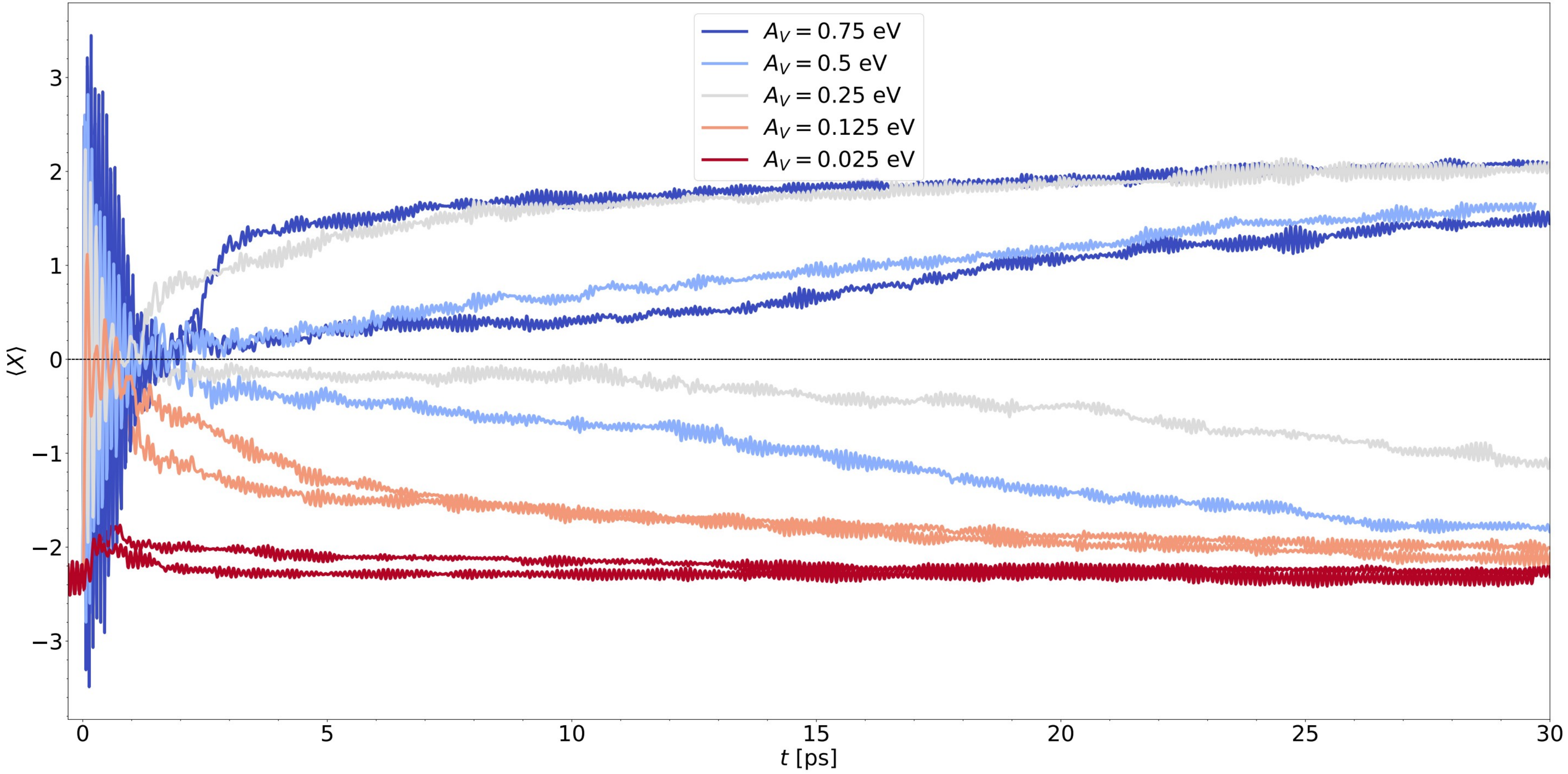
Different durations T_V of the coupling with the fermionic bath



$$\Gamma(t,t') = V(t)G_{\text{bath}}(t,t')V(t')^*$$

Time evolution of the staggered order parameter

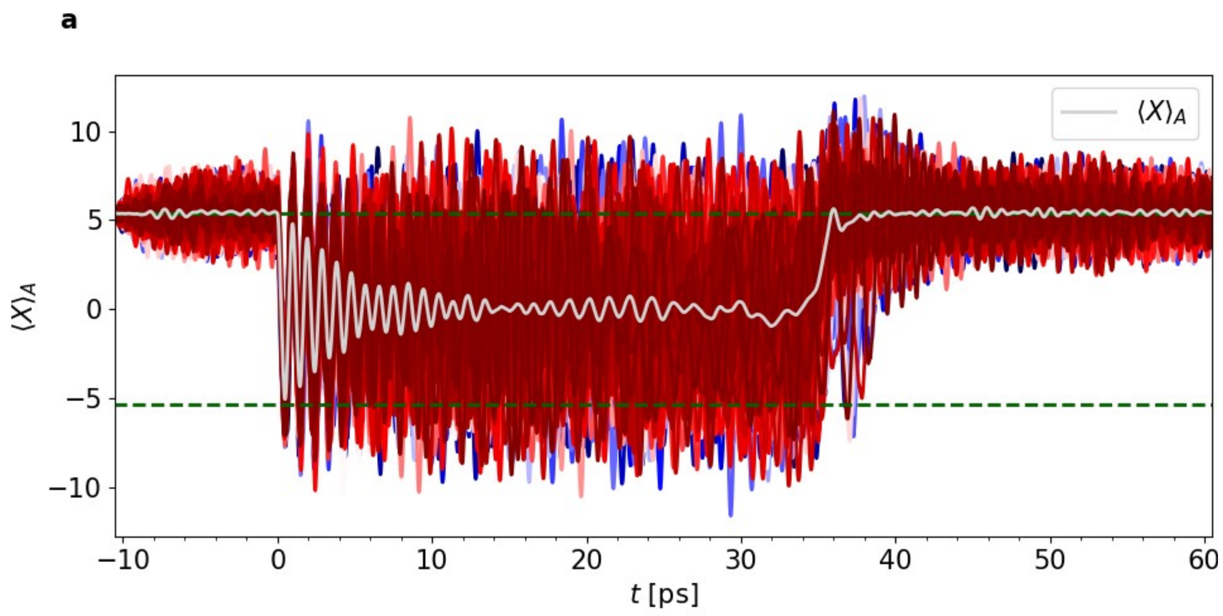
Different amplitudes of the driving field A_V

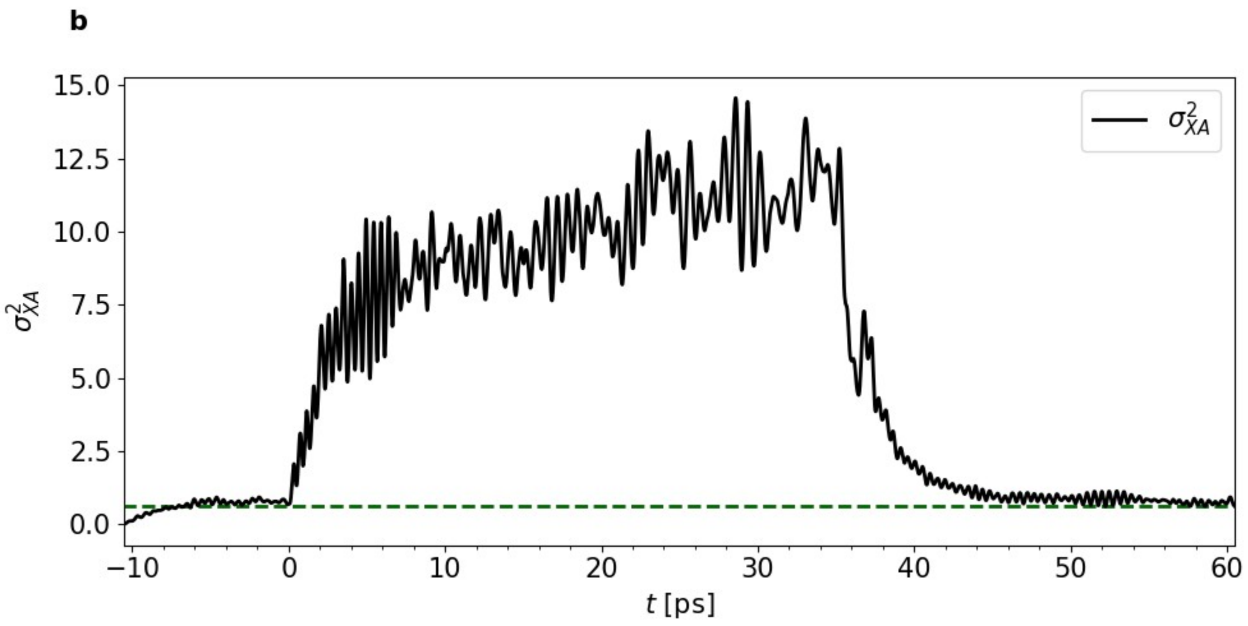


$$\Gamma(t,t') = V(t)G_{\mathrm{bath}}(t,t')V(t')^*$$

Nonequilibrium dynamics after photodoping

Coherent phonon oscillations at short times

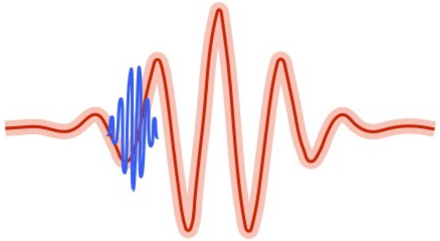


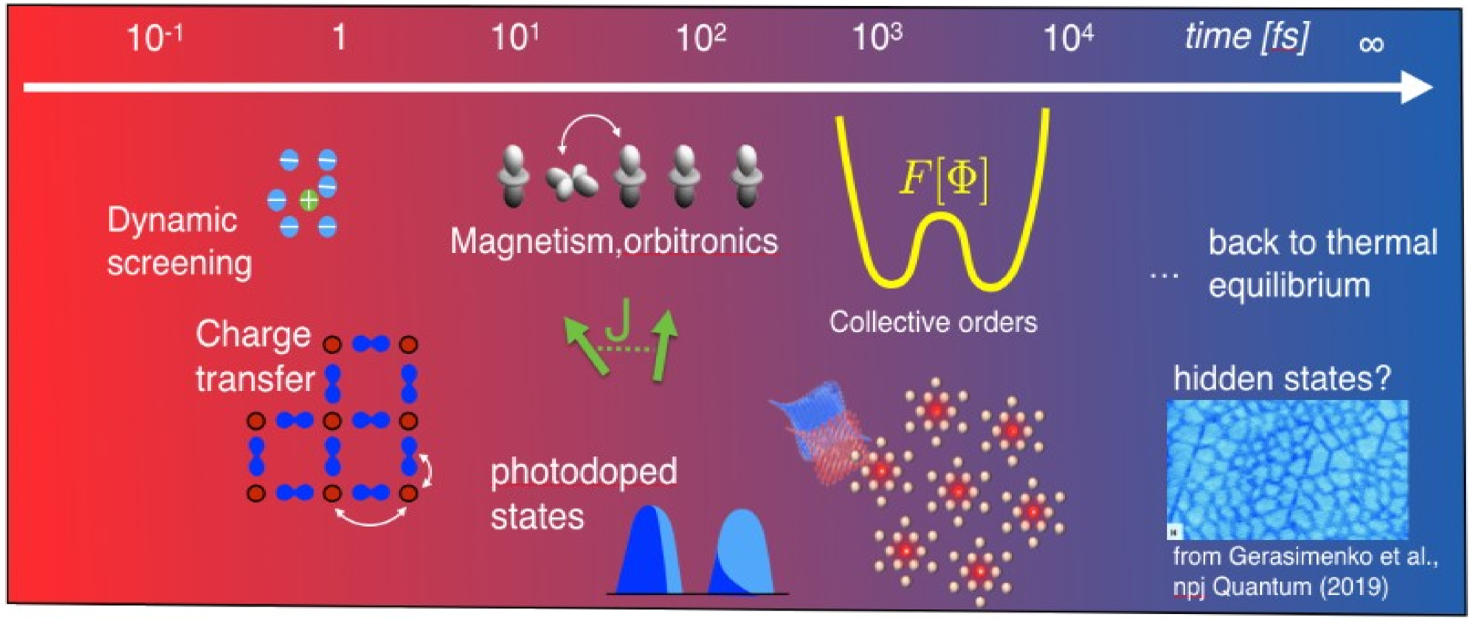


Additional slides

Non-equilibrium phenomena in condensed matter

The wide range of relevant timescales in the relaxation dynamics

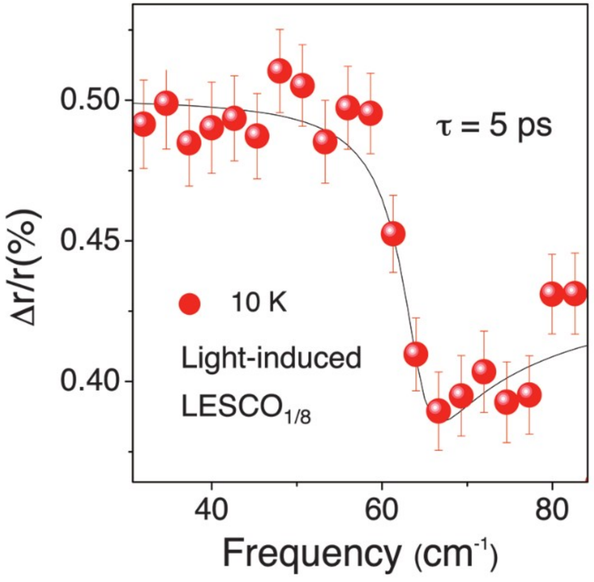




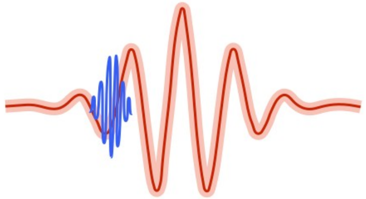
The interplay between the electrons and the other degrees of freedom in solids leads to the most fascinating out-of-equilibrium phenomena in condensed matter

Non-equilibrium phenomena in condensed matter

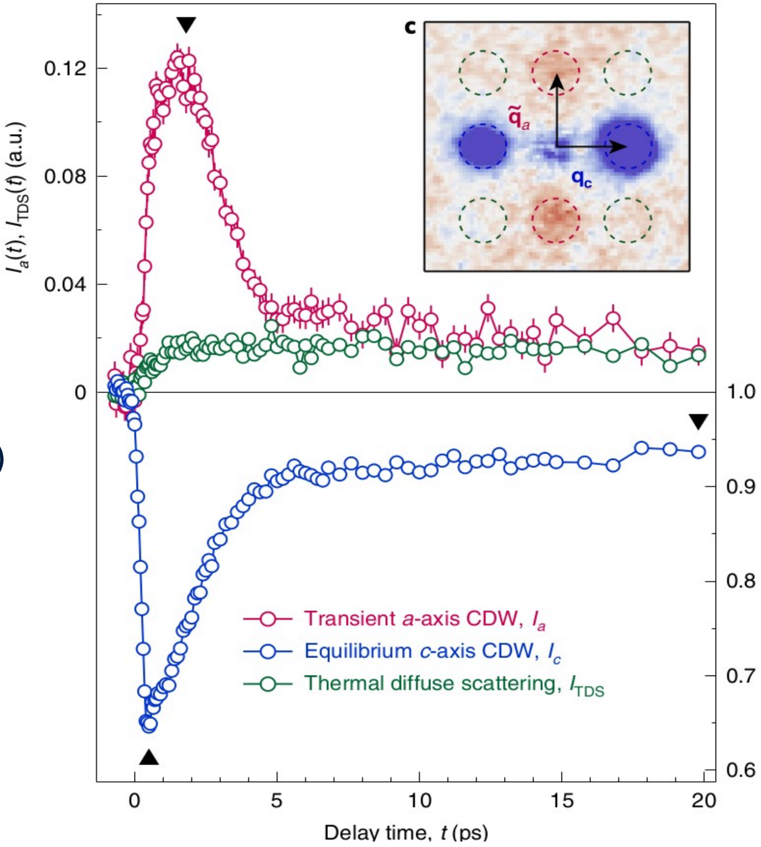
Pump-probe experiments

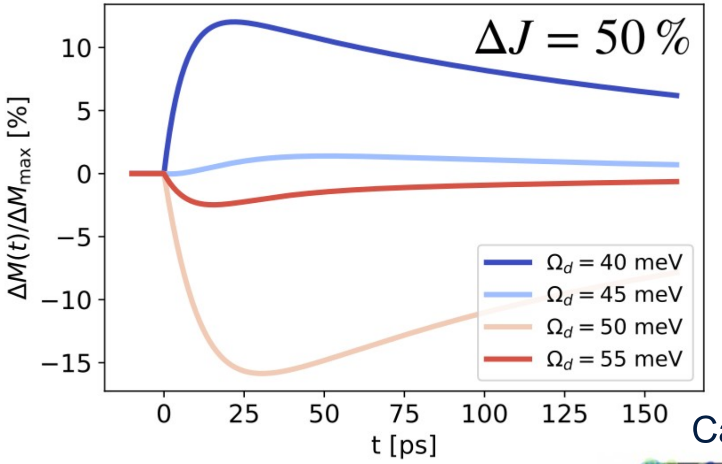


Fausti, D. et al.,
Science 331, 189-191 (2011)



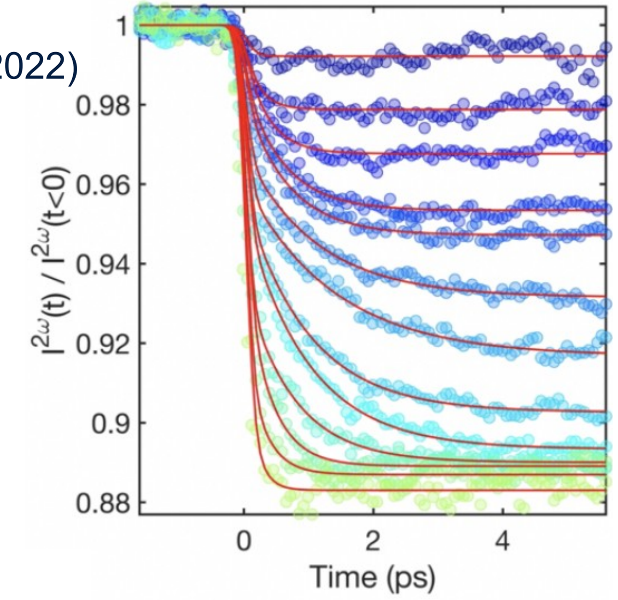
Kogar, A. et al.,
Nat. Phys. 16, 159-163 (2019)





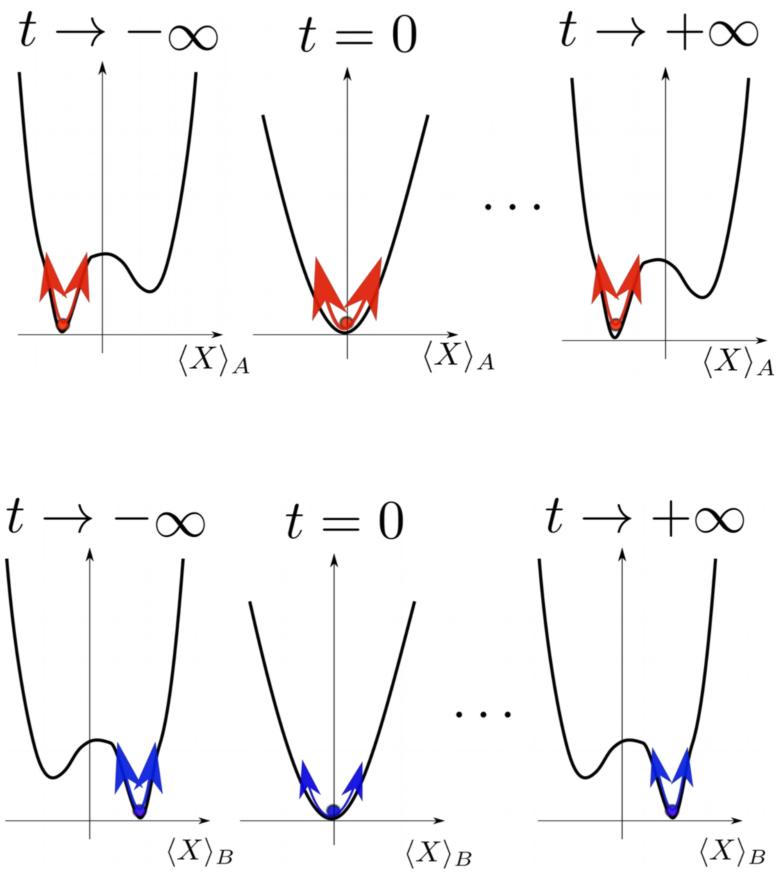
Curtis, J. et al,
arXiv:2209.10567v1 (2022)

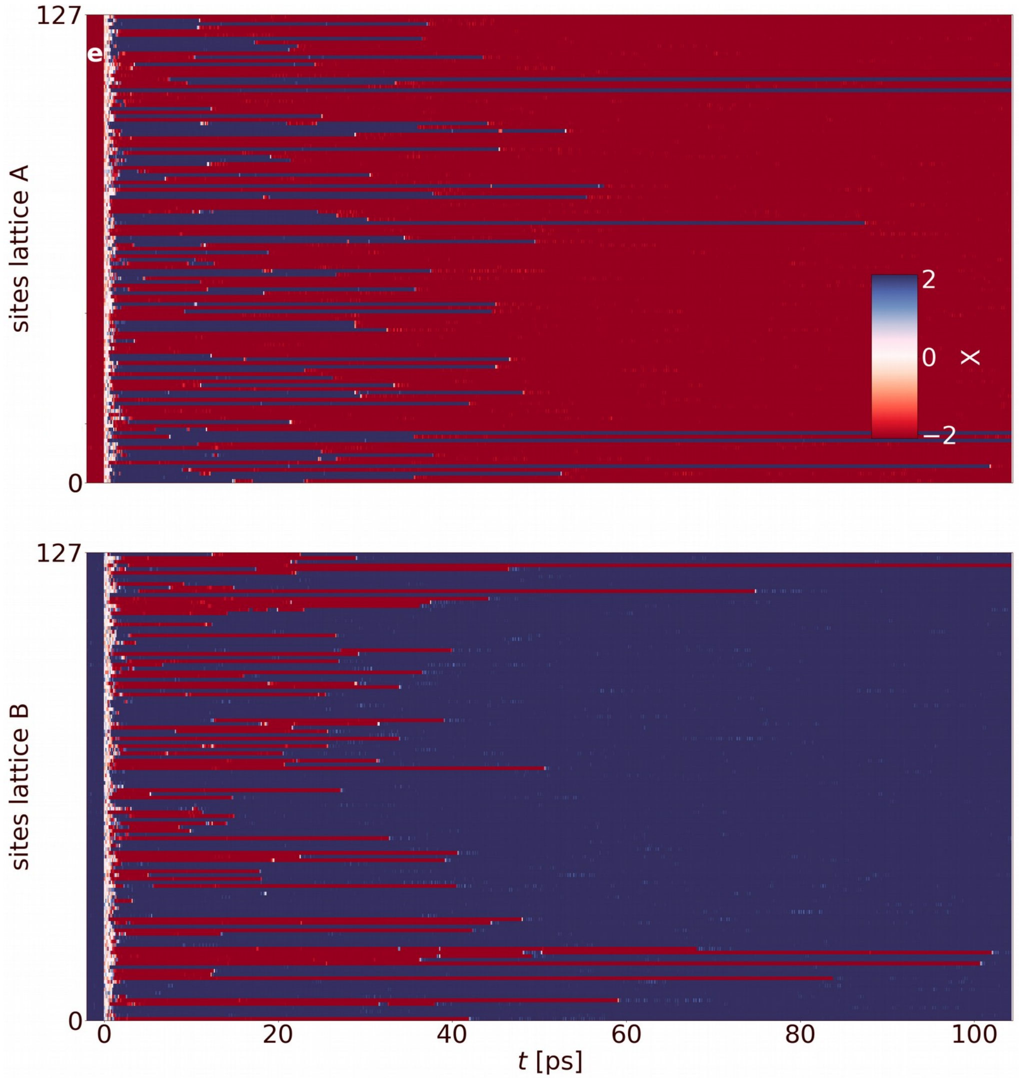
Carbin, T. et al., (2022)



Semiclassical solution: dynamics

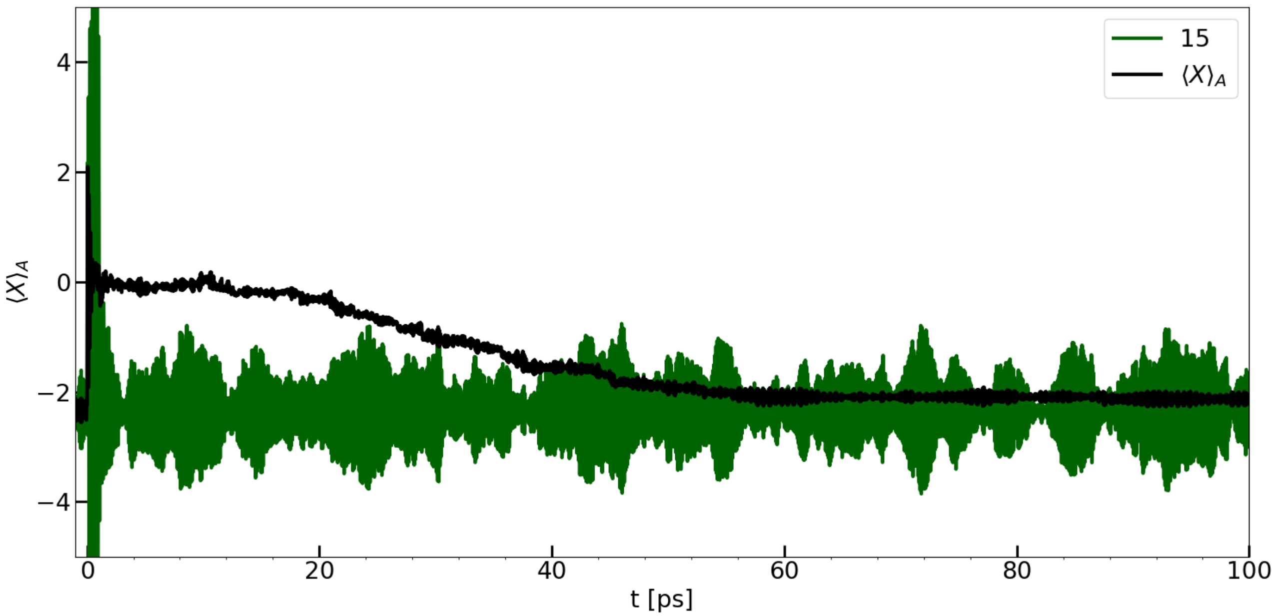
Long-time evolution: the average displacement and the single trajectories

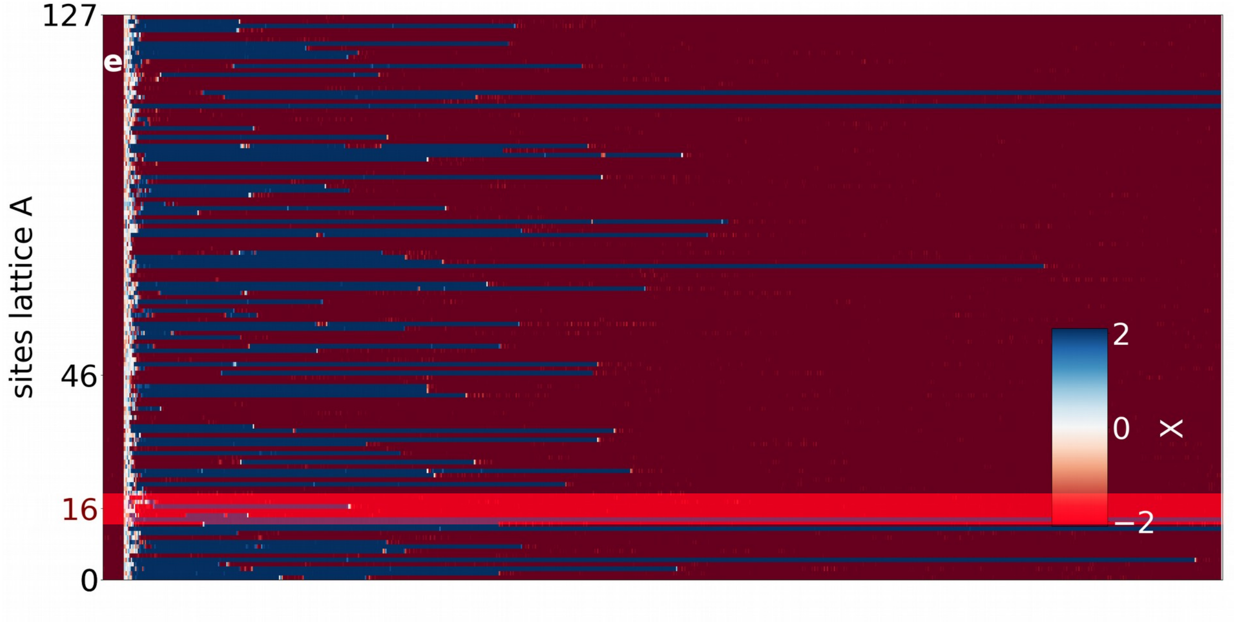




Semiclassical solution: dynamics

Long-time evolution: single trajectories not always representative of the average displacement





Trajectory 15 jumps to the global minimum well before the average does

Evaluation of memory integrals

Truncated Kadanoff-Baym equations

If the self-energy decays to zero, for large enough time one can impose:

$$\begin{aligned} &\Sigma^R(t,t') = \Sigma^<(t,t') = 0 && \text{for} && t - t' > t_c, \\ &\Sigma^\rceil(t,\tau) = 0 && && t > t_c \end{aligned}$$

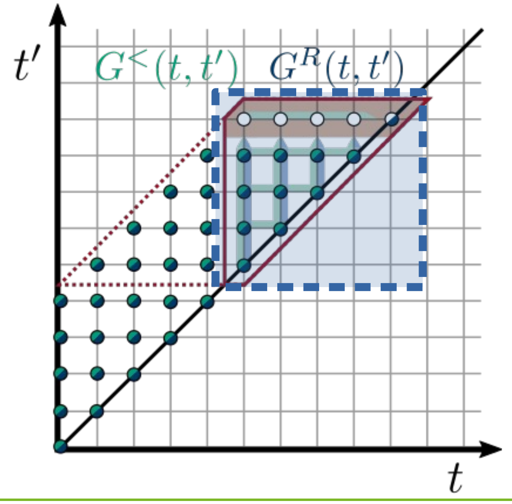
The self-consistency scheme therefore reduces to:

$$i\partial_t G^R(t,t') - \int_{t-t_c}^{t} d\bar{t}\, [\Sigma^R + \Delta^R](t,\bar{t}) G^R(\bar{t},t') = 0$$

$$i\partial_t G^<(t,t') - \int_{t-t_c}^{t} d\bar{t}\, [\Sigma^R + \Delta^R](t,\bar{t}) G^<(\bar{t},t') = \int_{t'-t_c}^{t'} d\bar{t}\, [\Sigma^< + \Delta^<](t,\bar{t}) G^A(\bar{t},t')$$

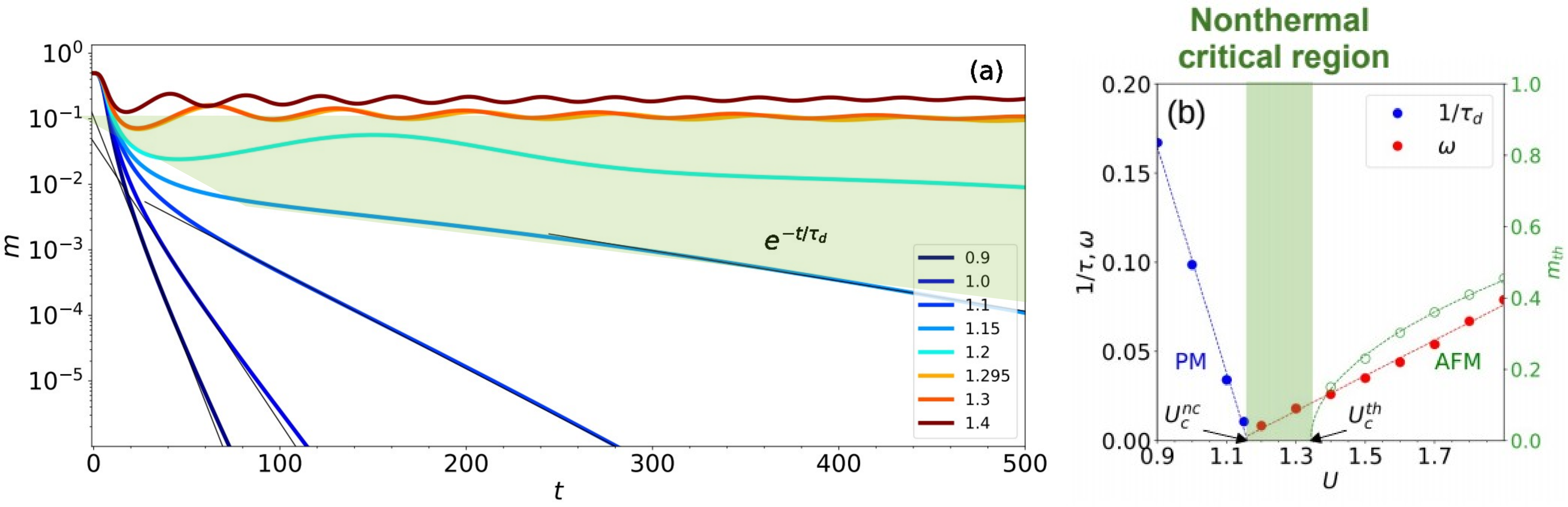
Computational effort $\sim \mathcal{O}(n n_c^2)$

Memory occupation $\sim \mathcal{O}(n_c^2)$



Short-time dynamics

Nonthermal critical region



Staggered magnetization $$m = \frac{1}{2}(n_{A,\uparrow} - n_{B,\uparrow}) = \frac{1}{2}(n_{B,\downarrow} - n_{A,\downarrow})$$

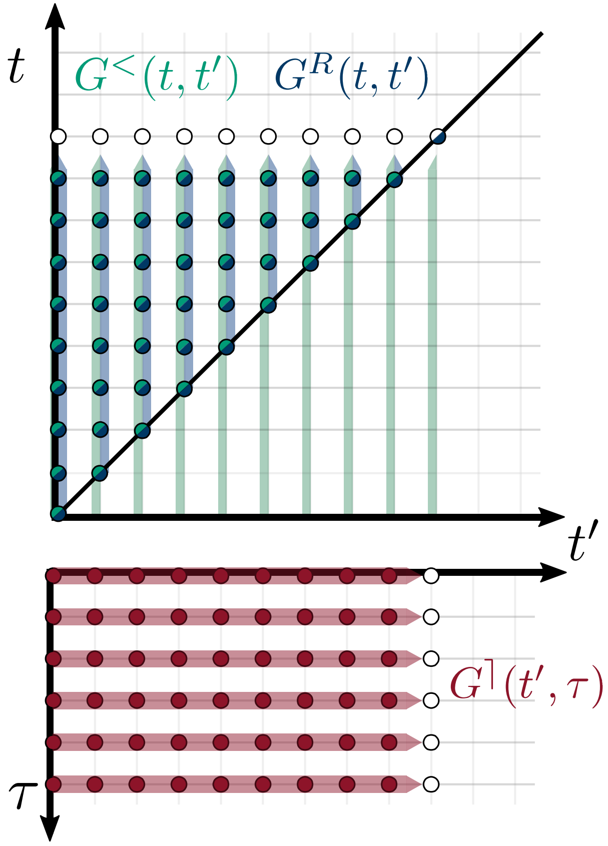
Picano A. et al. : Phys. Rev. B 103, 165118 (2021)

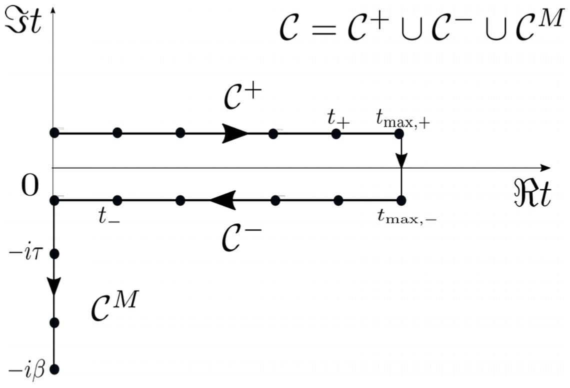
Evaluation of memory integrals on Keldysh contour

Kadanoff-Baym equations

Time evolution of the impurity local Green's function G:

$$i\partial_t G(t,t') - \int_{\mathcal{C}} d\bar{t}\ \ [\Sigma + \Delta](t,\bar{t})G(\bar{t},t') = \delta_{\mathcal{C}}(t,t')$$



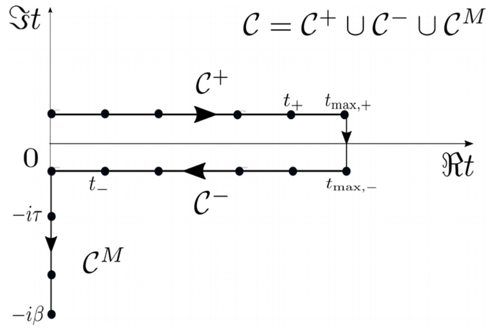


Computational effort $\sim \mathcal{O}(n^3)$

Memory occupation $\sim \mathcal{O}(n^2)$

Evaluation of memory integrals on Keldysh contour

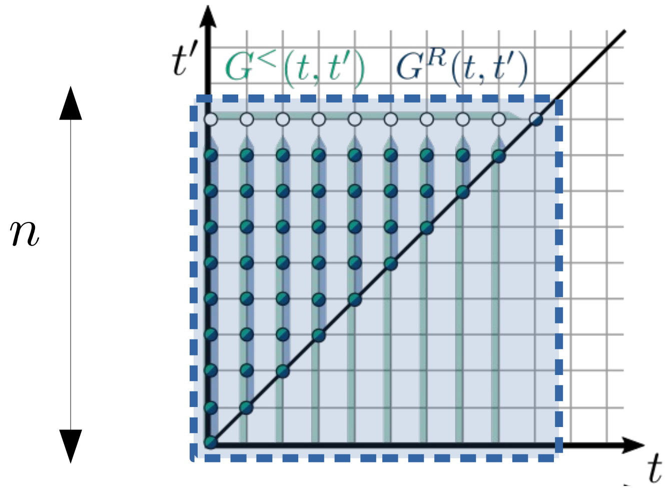
Kadanoff-Baym equations



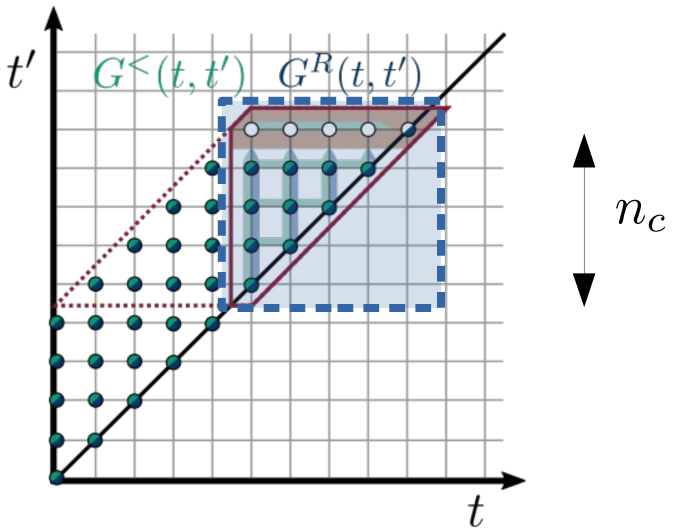
Time evolution of the impurity local Green's function G:

$$i\partial_t G(t,t') - \int_{\mathcal{C}} d\bar{t} \;\; [\Sigma + \Delta](t,\bar{t})G(\bar{t},t') = \delta_{\mathcal{C}}(t,t')$$

Memory untruncated scheme



Memory truncated scheme



Computational effort $\sim \mathcal{O}(n^3)$ $\qquad \sim \mathcal{O}(n n_c^2)$

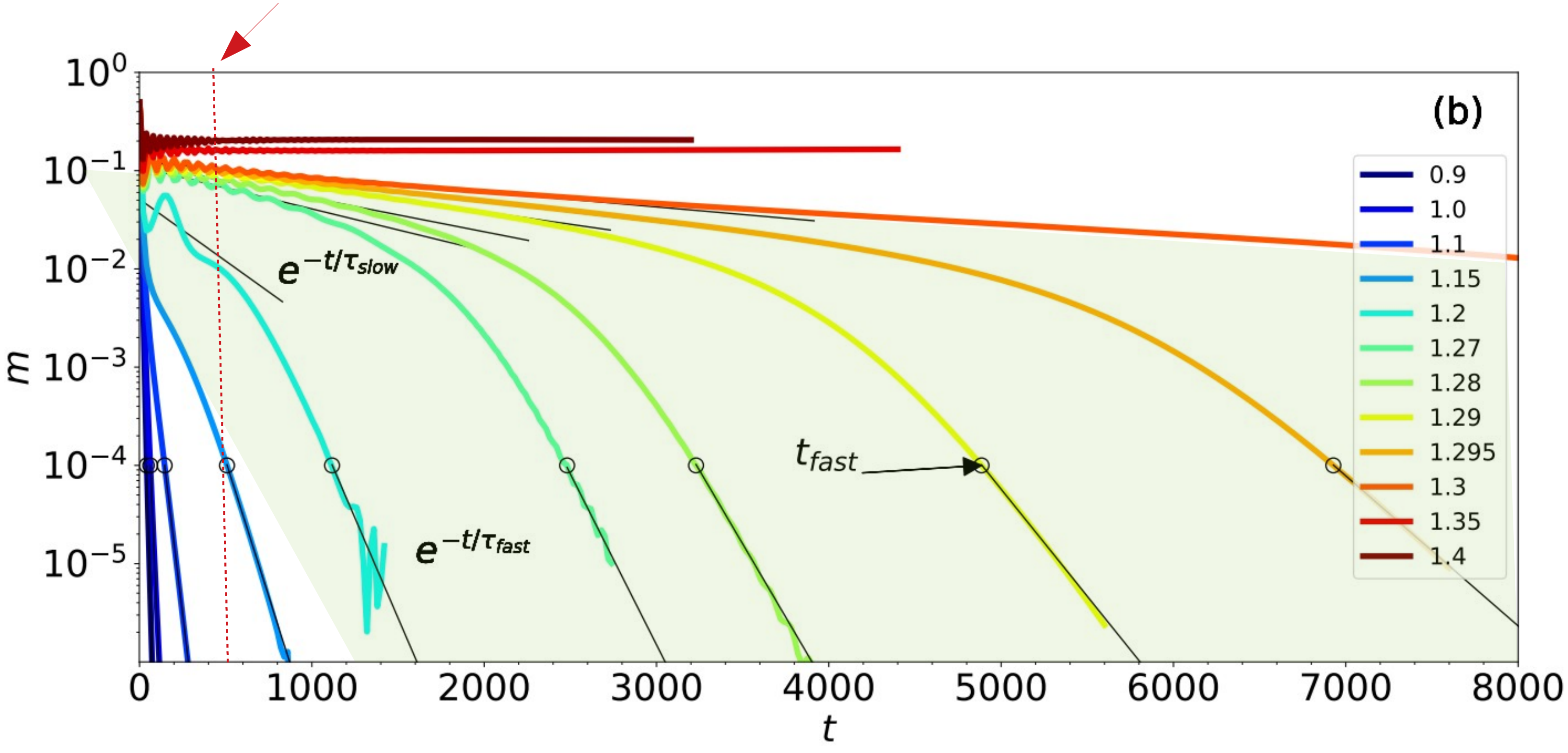
Memory occupation $\sim \mathcal{O}(n^2)$ $\qquad \sim \mathcal{O}(n_c^2)$

Stahl, C.; Dasari, N.; Li, J.; Picano, A. et al.: Phys. Rev. B 105, 115146 (2022)

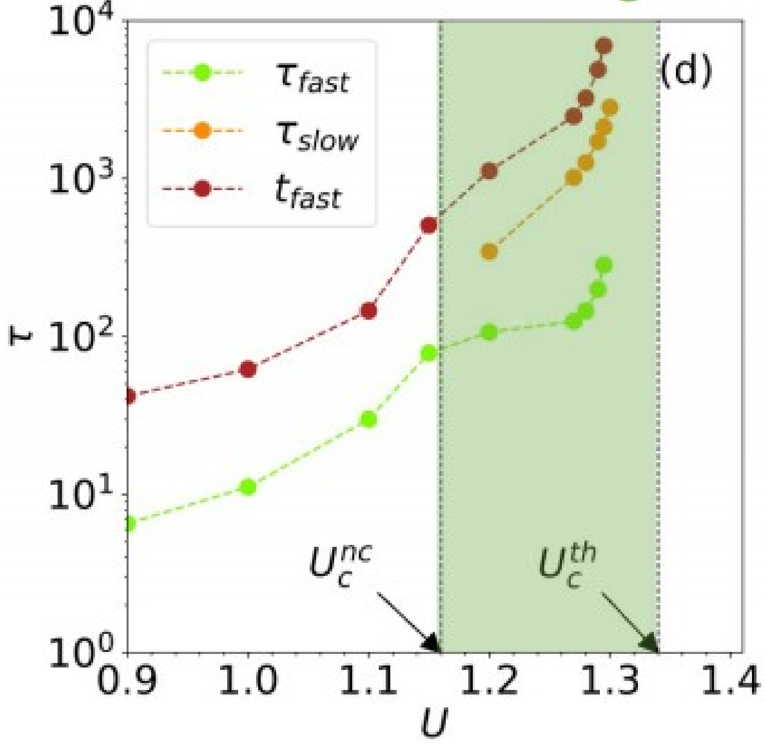
Long-time dynamics

Accelerated gap collapse

Two order of magnitudes longer in time!



Nonthermal critical region

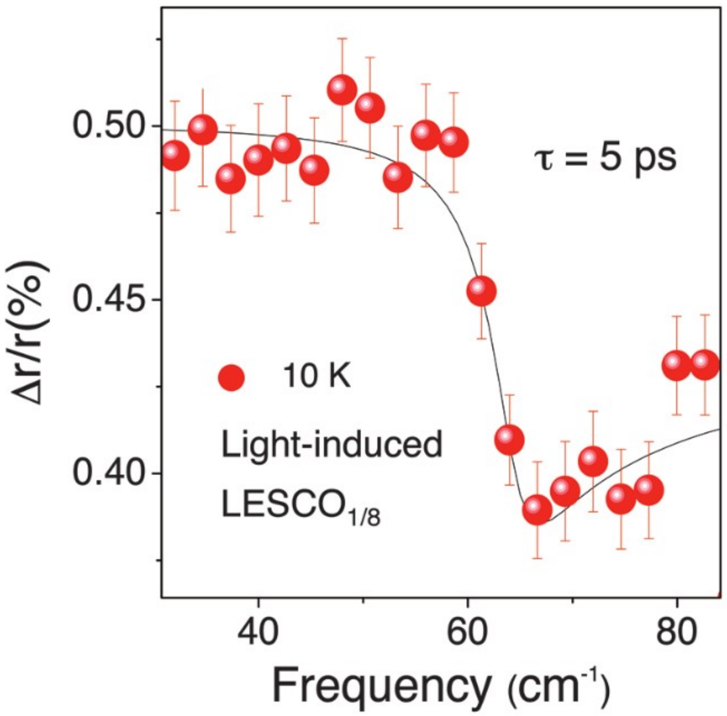


Picano, A. et al.: Phys. Rev. B 103, 165118 (2021)

Non-equilibrium phenomena in condensed matter

So-called light-induced superconductivity above the equilibrium T_C

Transient *c*-axis reflectance of $LESCO_{1/8}$



Agenda

Subheadline möglich. Gegebenenfalls löschen.