# Photoinduced pairing in Mott insulators

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## Abstract

**Utilizing time-evolution techniques in (infinite) matrix-product-state representation, we study the nonequilibrium dynamics of driven Mott insulators and demonstrate photoinduced $\eta$ pairing directly in the thermodynamic limit. Analyzing the time evolution of the corresponding pairing correlations, we determine the optimal laser pump parameters for which long-range $\eta$-pairing becomes dominant after pulse irradiation. The time-dependent photoemission spectra for this optimal pump parameter set show clear signatures of the photoinduced insulator-to-metal phase transition related to the formation of $\eta$ pairs.**

## Contents



## 1 Introduction

$\eta$ pairing, proposed first by C. N. Yang in 1989 [1], gives rise to a paring-density-wave-like off-diagonal long-range order in the Hubbard model. While it can be used to construct exact eigenstates of this model, $\eta$ pairing is absent in the Hubbard model's ground state, and

therefore has been attracted only specific attention, mostly from mathematical point of view. Recently, however, it was pointed out that the $\eta$-pairing state will be enforced by pulse irradiation [2]. The respective enhancement of pairing correlations emerged in time-dependent exact diagonalisations: Calculating all eigenstates as well as pairing correlations for a small cluster and taking the selection rule of $\eta$ pairs into account, Kaneko et al. showed that this photoinduced state is related to the $\eta$-pairing state [2].

Meanwhile, as a result of on-going developments in (time-depenent) density-matrix renormalisation group [(t-)DMRG] technique [3,4], optically driven systems in (quasi-)one-dimension can be simulated directly in the thermodynamic limit. In doing so, static correlation functions such as $\eta$-pair correlations can be computed by means of the infinite time-evolving block decimation (iTEBD) technique [5], taking advantage of translational invariance in the infinite matrix-product-state (iMPS) representation. Building window sites with so-called infinite boundary conditions (IBC) in the uniform update scheme [6] enables us to simulate nonequilibrium dynamics of excited (quasi-)one-dimensional (1D) systems by a laser electric field [7].

On this basis, in this study, we reexamine the time-evolution of photoinduced $\eta$-pairing, mainly to confirm or put in question previous small cluster results. Thereby we emphasize the importance of using optimal pump pulse parameters. Furthermore, we reconsider the relation between the $\eta$-pairing correlations and the optical spectrum in the small-amplitude regime after pulse irradiation. Finally we prove the photoinduced insulator-to-metal phase transition by simulating time-dependent photoemission spectra of driven Mott insulators.

## 2 Model

Let us consider the 1D half-filled Hubbard model,

$$\hat{H} = -t_{\mathrm{h}} \sum_{j,\sigma} \left( \hat{c}^{\dagger}_{j,\sigma} \hat{c}_{j+1,\sigma} + \mathrm{H.c.} \right) + U \sum_{j} \left( \hat{n}_{j,\uparrow} - 1/2 \right) \left( \hat{n}_{j,\downarrow} - 1/2 \right) , \tag{1}$$

where $t_{\mathrm{h}}$ is the nearest-neighbor transfer amplitude and $U$ gives the on-site part of the Coulomb interaction. In Eq. (1), $\hat{c}^{\dagger}_{j,\sigma}$ ($\hat{c}_{j,\sigma}$) creates (annihilates) a spin-$\sigma$ (=$\uparrow,\downarrow$) electron at Wannier lattice site $j$, and $\hat{n}_{j,\sigma} = \hat{c}^{\dagger}_{j,\sigma} \hat{c}_{j,\sigma}$. In the repulsive case ($U > 0$) the model realizes a Mott insulating ground state with a finite charge gap $\Delta$.

Exact eigenstates of the Hubbard model can be constructed by means of the operators $\hat{\eta}^{+} = \sum_j (-1)^j \hat{\Delta}^{\dagger}_j$, $\hat{\eta}^{-} = (\hat{\eta}^{+})^{\dagger}$, and $\hat{\eta}^{z} = \frac{1}{2} \sum_j (\hat{n}_{j,\uparrow} + \hat{n}_{j,\downarrow} - 1)$, where $\hat{\Delta}^{\dagger}_j = \hat{c}^{\dagger}_{j,\downarrow} \hat{c}^{\dagger}_{j,\uparrow}$ denotes the singlet pair-creation operator [1]. These so-called $\eta$ operators fulfill SU(2) commutation relations $[\hat{\eta}^{+}, \hat{\eta}^{-}] = 2\hat{\eta}^{z}$ and $[\hat{\eta}^{z}, \hat{\eta}^{\pm}] = \pm \hat{\eta}^{\pm}$. Apparently, the Hubbard Hamiltonian (1) commutes with $\hat{\eta}^2 = \frac{1}{2}(\hat{\eta}^{+}\hat{\eta}^{-} + \hat{\eta}^{-}\hat{\eta}^{+}) + (\hat{\eta}^{z})^2$, i.e., $\langle \eta^2 \rangle$ is a conserved quantity. Long-ranged pairing correlations $\langle \hat{\eta}^{+}_j \hat{\eta}^{-}_{\ell} \rangle$ develop when the expectation value $\langle \hat{\eta}^2 \rangle$ becomes finite, but such $\eta$-pairing states cannot be the ground state of the Hubbard model [1]. Pulse irradiation can establish $\eta$-paired states in Mott insulators however [2].

To address this issue, we apply a pump pulse with amplitude $A_0$, frequency $\omega_{\mathrm{p}}$ and width $\sigma_{\mathrm{p}}$, centered at time $t_0 (> 0)$:

$$A(t) = A_0 e^{-(t-t_0)^2/(2\sigma_{\mathrm{p}}^2)} \cos\left[ \omega_{\mathrm{p}} (t - t_0) \right] . \tag{2}$$

The external time-dependent electric field $A(t)$ changes the hopping amplitude by a Peierls phase [8]: $t_{\mathrm{h}} \hat{c}^{\dagger}_{j,\sigma} \hat{c}_{j+1,\sigma} \rightarrow t_{\mathrm{h}} e^{\mathrm{i}A(t)} \hat{c}^{\dagger}_{j,\sigma} \hat{c}_{j+1,\sigma}$, i.e., $\hat{H} \rightarrow \hat{H}(t)$. As a result, the system being initially in the ground state, is driven out of equilibrium, $|\psi(0)\rangle \rightarrow |\psi(t)\rangle$.

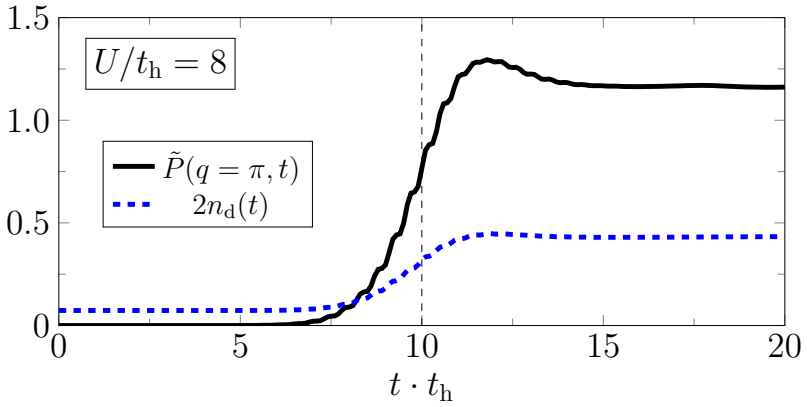


Figure 1: Typical time-evolution process of $\tilde{P}(q=\pi,t)$ and $2n_\mathrm{d}(t)$ for the photoinduced $\eta$-pairing states in the strong-coupling regime of the driven Hubbard model with $U/t_\mathrm{h} = 8$ and pump parameters $A_0$=0.4, $\omega_\mathrm{p}/t_\mathrm{h} = 7.0$, $\sigma_\mathrm{p} = 2t_\mathrm{h}^{-1}$ and $t_0 = 10t_\mathrm{h}^{-1}$. The iTEBD data are obtained for bond dimension $\chi = 1200$, ensuring a truncation error smaller than $10^{-5}$. For the iTEBD calculations, we employ a second-order Suzuki-Trotter decomposition with time step $0.1t_\mathrm{h}^{-1}$ ($0.01t_\mathrm{h}^{-1}$).

## 3 Pairing correlations

The $\eta$-pairing state can be detected evaluating the time evolution of the pair-correlation function

$$P(r,t) = \frac{1}{L}\sum_j \langle\psi(t)|\hat{\Delta}^\dagger_{j+r}\hat{\Delta}_j + \text{H.c.})|\psi(t)\rangle \tag{3}$$

and its Fourier transform $\tilde{P}(q,t) = \sum_r e^{iqr}P(r,t)$. As found in Refs. [2, 9] for small clusters, $\tilde{P}(\pi,t)$ is enhanced after pulse irradiation, indicating the formation of an $\eta$-pairing state. By means of iTEBD this is confirmed directly in the thermodynamic limit which is demonstrated in Fig. 1 for a pump with $A_0 = 0.4$, $\omega_\mathrm{p}/t_\mathrm{h} = 7.0$ and $\sigma_\mathrm{p} = 2t_\mathrm{h}^{-1}$ centered at $t_0 = 10t_\mathrm{h}^{-1}$. $\tilde{P}(\pi,t)$ shows a clear response to pulse irradiation and is strengthened as the system progresses in time until saturation is reached. Obviously, the nonlocal contributions have a stronger impact on $\tilde{P}(\pi,t)$ than the double occupancy $n_\mathrm{d}(t)$ [note that $P(r=0,t) = 2n_\mathrm{d}(t)$].

The enhancement process of $\eta$-pairing can be described as follows [2]: The initial state before pulse irradiation is the ground state of the Hubbard chain with $|\eta = 0, \eta^z = 0\rangle$, which is consistent with the numerical finding: $\tilde{P}(0, t=0) \simeq 0$ (see Fig. 1). Turning on the pump pulse, the Hamiltonian does not commute with the $\eta$-operators anymore,

$$[\hat{H}(t), \eta^+] = [\hat{H}, \eta^+]\cos[A(t)] + \sum_k F(k,t)\hat{c}^\dagger_{\pi-k,\downarrow}\hat{c}^\dagger_{k,\uparrow}, \tag{4}$$

where $F(k,t) = 4t_\mathrm{h}\sin[A(t)]\sin k$. This alters the initial state to a state with a finite expectation value $\langle\hat{\eta}^2\rangle$. Even though the commutation relation is recovered for $t \gg t_0$, i.e., $[\hat{H}(t), \hat{\eta}^+] \to [\hat{H}, \hat{\eta}^+]$ [since $A(t) \to 0$], $|\psi(t)\rangle$ now includes components of $|\eta > 0, \eta^z = 0\rangle$ leading to the enhancement of $\tilde{P}(\pi,t)$, see Fig. 1 for $t > t_0$.

Note that the enhancement of $\eta$ pairing after pulse irradiation depends, however, strongly on the pump pulse parameters. The optimal parameter set for inducing $\eta$-pairing states can be determined examining the $A_0$ and $\omega_\mathrm{p}$ dependences of $\tilde{P}(\pi,t)$ by iTEBD. Figure 2(a) shows the contour plot of $\tilde{P}(\pi,t)$ after pulse irradiation ($t = 15t_\mathrm{h}^{-1}$). We find a single maximum around $A_0 \approx 0.4$ and $\omega_\mathrm{p}/t_\mathrm{h} \approx 7.0$ (marked by the "×" symbol), instead of the stripe structure observed in the finite-system ($L = 14$) exact diagonalisation (ED) simulations [2].

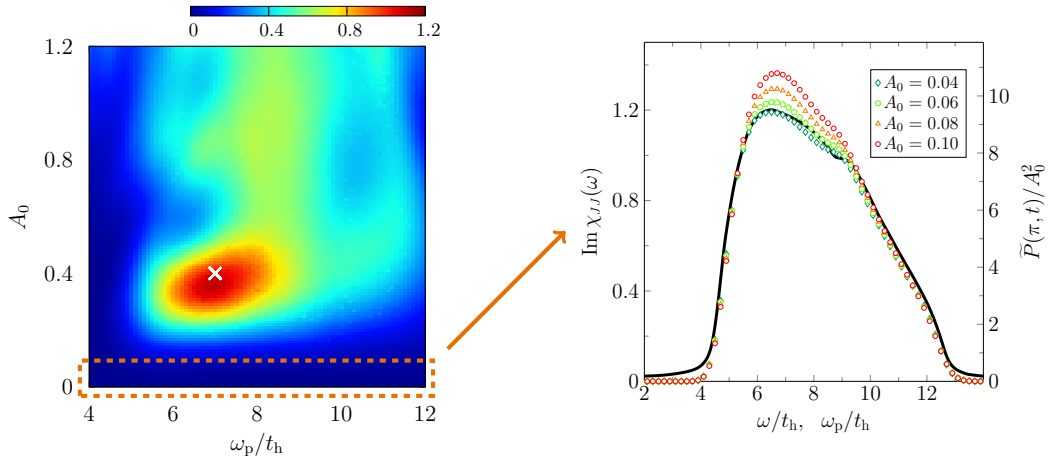


Figure 2: (a): Contour plots of $\tilde{P}(q=\pi,t)$ in the $\omega_p$-$A_0$ plane at $t=15t_h^{-1}$. Again $U/t_h=8$, and the pump is parametrized by $\sigma_p=2t_h^{-1}$ at $t_0=10t_h^{-1}$. (b): $\tilde{P}(\pi,t)$ at $t=15t_h^{-1}$ in the small-$A_0$ area enclosed by the dashed square in panel (a). Dividing by $A_0^2$, data can be rescaled to $\mathrm{Im}\chi(\omega)$ (black line), where $\mathrm{Im}\chi(\omega)$ is the imaginary part of the optical spectrum $\chi_{JJ}(\omega)$.

Another notable results of previous ED calculations [2] was that the peak structure of $\tilde{P}(\pi,t)$ as a function of $\omega_p$ for small $A_0$ is essentially the same as those of the ground-state optical spectrum,

$$\chi_{JJ}(\omega>0)=-\frac{1}{L}\langle\psi_0|\hat{J}\frac{1}{E_0-\hat{H}+\hbar\omega+i\eta_L}\hat{J}|\psi\rangle\,, \tag{5}$$

where $|\psi_0\rangle$ is the ground state having energy $E_0$ and Lorentzian width $\eta_L$. In (5), the Hubbard-model charge-current operator is $\hat{J}=it_h\sum_{j,\sigma}(\hat{c}^\dagger_{j,\sigma}\hat{c}_{j+1,\sigma}-\hat{c}^\dagger_{j+1,\sigma}\hat{c}_{j,\sigma})$.

Figure 2(b) compares the iTEBD data, obtained for $\tilde{P}(\pi,t)$ at various small $A_0$ and $t=15t_h^{-1}$, with the t-DMRG results for $\chi_{JJ}(\omega)$ (using $\eta_L/t_h=0.2$), in dependence on $\omega_p$ respectively $\omega$. Most notably, $\tilde{P}(\pi,t)$ divided by $A_0^2$ scales to the imaginary part of the optical spectrum $\mathrm{Im}\chi(\omega)$. This can be understood as follows: The hopping term including the Peierls phase can be divided into kinetic and current operators as

$$-t_h\sum_{j,\sigma}\left(e^{iA(t)}\hat{c}^\dagger_{j,\sigma}\hat{c}_{j+1,\sigma}+\mathrm{H.c.}\right)=\hat{K}\cos[A(t)]+\hat{J}\sin[A(t)]\,, \tag{6}$$

where $\hat{K}=-t_h\sum_{j,\sigma}(\hat{c}^\dagger_{j,\sigma}\hat{c}_{j+1,\sigma}+\mathrm{H.c.})$. For small $A_0$ and large $t$, the second term in Eq. (6) can be approximated by $\hat{J}A_0$, yielding a significant contribution of $A_0^2$ to the pair correlations. Needless to say that the finite-size effects are eliminated by simulating the pair correlations directly in thermodynamic limit by iTEBD, leading to the single-peak structure in Fig. 2(b), in strong contrast to the multiple-peak structure observed in the ED calculations [2].

## 4 Nonequilibrium dynamics

We now analyze the nonequilibrium photoemission spectra $A(k,\omega;t)=\sum_{\sigma=\uparrow,\downarrow}A_\sigma(k,\omega;t)$ for the optimal pump parameter set marked by the "×"-symbol in Fig. 2(a). To explore the system dynamics in a nonequilibrium situation, time-dependent spectral functions of the form [10]

$$A_\sigma(k,\omega;t)=\sum_r e^{-ikr}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\tau_1 d\tau_2\, f(\tau_1,\tau_2;\omega)\cdot C_\sigma(r,\tau_1,\tau_2;t) \tag{7}$$

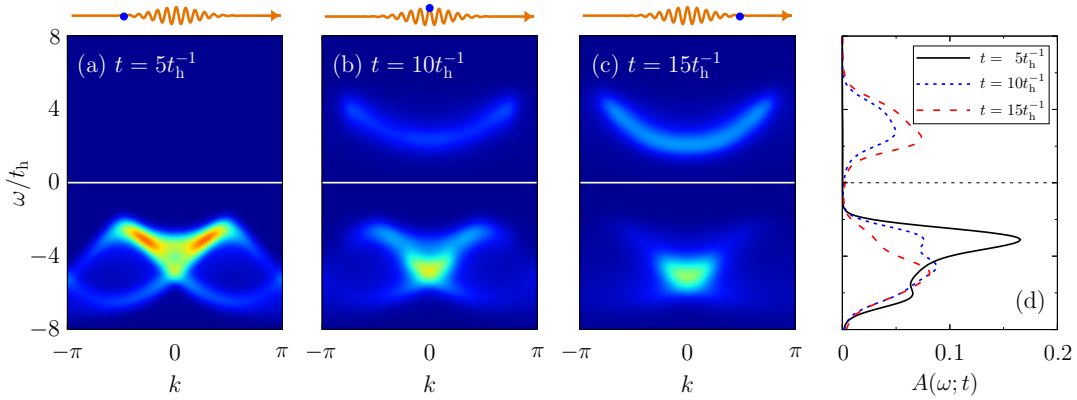


Figure 3: Snapshots of the photoemission spectra $A(k,\omega;t)$ indicating photoinduced $\eta$-pairing during the pump at times $t = 5t_h^{-1}$ (a), $10t_h^{-1}$ (b) and $15t_h^{-1}$ (c). The pump is parametrized by $A_0 = 0.4$, $\omega_p/t_h = 7.0$ [see '×'-symbol in Fig. 2(a)], and $\sigma_p = 2t_h^{-1}$ at $t_0 = 10t_h^{-1}$. The transient integrated density of states $A(\omega;t)$ obtained from the data of panels (a)-(c) is depicted in panel (d). All data are obtained by the (i)TEBD technique with IBC for the 1D half-filled Hubbard model with $U/t_h = 8$. Note that the time cutoff in the simulation of time-dependent correlation functions is $T = 5t_h^{-1}$, i.e., the integration in Eq. 7 extends only over the interval $-T \le \tau_1, \tau_2 \le T$. As a compromise between time and frequency resolutions we have chosen a probe pulse width $\sigma_{pr} = 2t_h^{-1}$.

are of interest. Here, the nonequilibrium two-point correlator

$$C_\sigma(r,\tau_1,\tau_2;t) = \langle\phi(t)|\hat{c}^\dagger_{j+r,\sigma}(\tau_1;t)\hat{c}_{j,\sigma}(\tau_2;t)|\phi(t)\rangle \tag{8}$$

is defined relative to $t$, and

$$f(\tau_1,\tau_2;\omega) = e^{i\omega(\tau_1-\tau_2)}g(\tau_1)g(\tau_2),\ \ g(\tau) = \exp[-\tau^2/2\sigma^2_{pr}]/\sqrt{2\pi}\sigma_{pr} \tag{9}$$

specify the shape of the probe pulse, e.g., in a time-dependent photoemission spectroscopy experiment. How numerically simulate two-time-dependent quantities such as $C_\sigma(r,\tau_1,\tau_2;t)$ has been explained in detail in Ref. [7] [see paragraphs below Eq. (1)].

Figure 3 displays our (i)TEBD results for the 1D half-filled Hubbard model in the strong-coupling regime ($U/t_h = 8$). Before pump irradiation the state is a Mott insulator with a noticable single-particle gap, see Fig. 3(a) for $t = 5t_h^{-1}$. In the midst of the pump ($t = 10t_h^{-1}$), an extra dispersion above Fermi energy ($\omega > E_F$) appears and persists afterwards [Fig. 3(c)].

Evaluating the integrated density of states

$$A(\omega;t) = \frac{1}{L}\sum_k A(k,\omega;t), \tag{10}$$

we see more clearly how the spectral weight is shifted from $\omega < E_F$ to $\omega > E_F$ due to the photoinduced $\eta$-pairing. Figure 3(d) gives $A(\omega;t)$ for the photoinduced $\eta$-pairing state. Obviously, the spectral weight for $\omega > E_F$ increases distinctly over time, indicating a photoinduced phase transition from a Mott insulator to a metallic $\eta$-pairing state. This photoinduced insulator-to-metal transition should be observed in time- and angle-resolved photoemission spectroscopy, when the pure Hubbard model is realized experimentally, e.g., in optical lattices. We note that the photoinduced phase transition cannot be observed by simulating the time-dependent photoemission spectra with not-optimized pump-pulse parameters, see Ref. [7].

## 5 Conclusions

To summarize, combining tensor-network algorithms with infinite time-evolving block decimation techniques, we revisited the problem of photoinducing $\eta$-pairing states in the one-dimensional Hubbard model at half band filling. This allowed us to prove the enhancement of the pairing correlations directly in the thermodynamic limit. We also determined the optimal pump-pulse parameter set that maximizes the $\eta$-pairing tendency. An $\eta$-pairing related Mott insulator to metal transition could be extracted from the time-dependent photoemission spectrum.

We wish to stress that the numerical approach presented here can be applied to simulate the nonequilibrium dynamics of any (quasi-)one-dimensional translational-invariant system in entire ranges of interacting and driving parameters. For example, the photoinduced metallization of excitonic insulators was demonstrated quite recently in accordance with time- and angle-resolved photoemission spectroscopy experiments on $Ta_2NiSe_5$ [14, 15].

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