# Quasi-steady state descriptions for photo-doped Mott insulators

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YM, S. Takayoshi, T. Kaneko, Z. Sun, D. Golež, A. J. Millis and P. Werner, Comm. Phys. 5, 23 (2022).
 YM, S. Takayoshi, T. Kaneko, A. Läuchli and P. Werner, Phys. Rev. Lett. 130, 106501 (2023).
 Review: YM, D Golež, M Eckstein, P Werner, arXiv:2310.05201.



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#### Background: Physics out of strong light-matter coupling

#### Weak light excitation (Linear regime)



Same frequencies of input and output

Matter stays in equilibrium

Strong light excitation (Nonlinear regime)



 Change in properties of output light ex) High-harmonic generation

 Change in properties of matter ex) Insulator-metal transition, light-induced superconductor



Control of properties of light and matter

## Strong light-matter coupling and emergent phenomena

Rapid development on strong laser techniques in THz and mid-infrared regime



Potential impact on next generation photo-electronics technology & new spectroscopy techniques

ex) Fast memory, Spintronics, 6G telecommunication, Attosecond spectroscopy, etc..

#### Appeal of strongly correlated systems

Strongly correlated systems: Crucial role of interactions between electrons

➡ Various emergent collective phenomena in and out of equilibrium



#### Examples of Photo-induced phase transitions



General question: Origin of nonequilibrium phases?

## Doping charge carriers into Mott insulators: Equilibrium



A. Kordyuk, Low. Temp. Phys. 41, 319 (2015)

Doping activates correlations between spin, orbital and charge



Emergence of rich phases

### Doping charge carriers into Mott insulators: Equilibrium



A. Kordyuk, Low. Temp. Phys. 41, 319 (2015)

Doping activates correlations between spin, orbital and charge

> Emergence of rich phases

#### Doping charge carriers into Mott insulators



#### Various types of charge carriers are activated at the same time

cf. Equilibrium doping  $\rightarrow$  holon **or** doublon

#### Long-life time of photo-carriers and metastable states

#### Life-time of doublon • holon

Just after excitation

$$U \gg v$$
  $\Box > \tau_{
m rec} \gg 1/v$  (Exponential with  $U/v$  )

N. Strohmaiser, et. al., PRL 104, 080401 (2010).
R. Sensarma, et. al., PRB 82, 224302 (2010).
A. Rosch, et. al., PRL 101, 265301 (2008).

#### Metastable steady state



- (Approximate) conservation of doublons and holons
- Intraband relaxation + Cooling via environment



## General question & three complementary approaches

What kinds of metastable states emerge in photo-doped Mott insulators?

1) Direct time-evolution

Methods: Exact Diagonalization, Tensor network, Dynamical mean-field theory, etc...

2) Quasi-NESS approach

Approximate quasi-steady state with a true steady state supported by external bath

J. Li, et. al., PRB **102**, 165136 (2020). J. Li and M. Eckstein, PRB **103** 045133 (2021).





Review: YM, D Golež, M Eckstein, P Werner, arXiv:2310.05201

3) Quasi-equilibrium approach

Analogous to photo-doped semiconductor
 Mainly used in this talk

A. Rosch, et. al., PRL **101**, 265301 (2008). Y. Kanamori, et al., PRL 107, 167403 (2011). YM, et. al., Comm. Phys. 5, 23 (2022).

## Quasi-equilibrium approach for photo-doped semiconductors <sup>12</sup>

K. Asano, Bussei Kenkyu (2013). L. V. Keldysh, *Contemporary Phys.* **27**, 395 (1986).



Strongly correlated systems?

## Quasi-equilibrium description for strongly correlated systems <sup>13</sup>



Introducing chemical potential for local multiplets and effective temperature

$$\hat{K}_{\text{eff}} = \hat{H}_{\text{eff}} - \sum_{g \in \text{ps}} \mu_g \hat{n}_g \qquad \hat{\rho}_{\text{eff}} = \exp(-\beta_{\text{eff}} \hat{K}_{\text{eff}}) \qquad \text{GGE type description}$$

Solve the effective problem with existing **equilibrium** methods

Step3

#### Example : Extended Hubbard model

$$\begin{split} \hat{H} &= -v \sum_{\langle i,j \rangle,\sigma} \hat{c}_{i}^{\dagger} \hat{c}_{j} + \hat{H}_{U} + \hat{H}_{V} \quad \text{with} \\ \begin{aligned} \hat{H}_{U} &= U \sum_{i} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} \\ \hat{H}_{V} &= V \sum_{\langle ij \rangle} \hat{n}_{i} \hat{n}_{j} \end{aligned} \qquad U \gg v, V \\ \hat{H}_{V} &= V \sum_{\langle ij \rangle} \hat{n}_{i} \hat{n}_{j} \end{aligned} \qquad U \gg v, V \end{split}$$

$$\begin{split} \text{Effective model with conserved local multiplets and effects of virtual fluctuation} \\ \hat{H}_{\text{eff}} &= \hat{H}_{U} \qquad \leftarrow \mathcal{O}(U) \qquad 4 \text{ types of pseudo-particles} \\ &+ \hat{H}_{\text{kin,LHB}} + \hat{H}_{\text{kin,UHB}} \qquad \leftarrow \mathcal{O}(v) \qquad \mathcal{O}(J_{\text{ex}}) \ J_{\text{ex}} = \frac{4v^{2}}{U} \qquad \textbf{d} \qquad \textcircled{e} \qquad \textcircled{$$

#### Previous analysis : metastable η pairing phase

Cold atom with extreme doping

A. Rosch, et. al., PRL 101, 265301 (2008).

Metastable state with doublon or holon



#### Photo-doped metastable states in 1D?





#### Main points

- $\triangleright$  Exact from of wave function of photo-doped states:  $|\Psi\rangle = |\Psi_{\rm SF}^{\rm GS}\rangle |\Psi_{\rm spin}^{\rm GS}\rangle |\Psi_{\eta-{\rm spin}}^{\rm GS}\rangle$
- $\triangleright$  Spin, charge and  $\eta$ -spin separation
- Intuitive insight into physics of metastable states

Emergent degrees of freedoms by photo-doping lead to intriguing nonequilibrium phases!

#### Exact wave function of photo-doped metastable states <sup>17</sup>

YM, et al., PRL. 130, 106501 (2023).

Wave function @  $U \rightarrow \infty$  ,  $V/J_{ex} = \text{const}$ ,  $T_{eff} = 0$ 

$$\begin{split} |\Psi\rangle = |\Psi_{SF}^{GS}\rangle |\Psi_{spin}^{GS}\rangle |\Psi_{\eta-spin}^{GS}\rangle \\ \downarrow_{\gamma-spin}^{\text{Spinless fermion}} \\ \downarrow_{\gamma-spin}^{\text{Squeezed}} \\ H_{SF,free} \\ H_{spin}^{(SQ)} \\ H_{\eta-spin}^{(SQ)} \\ \downarrow_{\gamma-spin}^{(SQ)} \\ \downarrow_{\gamma-spin}^{$$

ho Extension of Ogata-Shiba state in equilibrium  $|\Psi
angle=|\Psi^{
m GS}_{
m SF}
angle|\Psi^{
m GS}_{
m spin}
angle$ 

M. Ogata & H. Shiba, PRB 41 2326 (1990).

 $\triangleright$  Spin, charge and  $\eta$ -spin separation

Useful insight into physics

## Explanation of $|\Psi\rangle = |\Psi_{\rm SF}^{\rm GS}\rangle |\Psi_{\sigma}^{\rm GS}\rangle |\Psi_{\eta}^{\rm GS}\rangle$

YM, et al., PRL. 130, 106501 (2023). *L*: System size New expression of states:  $\hat{U}$  $N_{\rm s}$ : Number of singly occupied sites  $N_{\eta}$ : Number of doublons and holons Spinless fermion Squeezed Squeezed (Position of Singlons) spin space  $\eta$  -spin space L sites L sites  $N_{\rm s}$  sites  $N_n$  sites Hamiltonian for  $J_{ex} = 0$  in the new expression 0 th order wave function  $|\Psi_{\rm SF}^{\rm GS}\rangle|\Psi_{\sigma,\eta}\rangle$  $\hat{U}\hat{H}_{\rm kin}\hat{U}^{\dagger} = -t_{\rm hop}\sum_{i}(\hat{c}_{i}^{\dagger}\hat{c}_{j} + {\rm h.c.}) (\equiv \hat{H}_{\rm SF, free}) \quad \Box \searrow$  $\langle i,j \rangle$ i.e. Degeneracy of  $2^{N_s} \cdot 2^{N_\eta}$ 

 $|\Psi_{\sigma,\eta}
angle$  is determined by degenerate perturbation theory

## Explanation of $|\Psi\rangle = |\Psi_{\rm SF}^{\rm GS}\rangle|\Psi_{\sigma}^{\rm GS}\rangle|\Psi_{\eta}^{\rm GS}\rangle$

with

YM, et al., PRL. 130, 106501 (2023).

#### $\mathcal{O}(J_{ m ex})~~{ m terms}~{ m projected}~{ m to}~|\Psi_{ m SF}^{ m GS} angle|m{\sigma} angle$

$$\hat{H}_{\rm spin}^{\rm (SQ)} = J_{\rm ex}^{s} \sum_{i} \hat{\mathbf{s}}_{i+1} \cdot \hat{\mathbf{s}}_{i},$$
$$\hat{H}_{\eta-\rm spin}^{\rm (SQ)} = -J_{X}^{\eta} \sum_{j} (\hat{\eta}_{j+1}^{x} \hat{\eta}_{j}^{x} + \hat{\eta}_{j+1}^{y} \hat{\eta}_{j}^{y}) + J_{Z}^{\eta} \sum_{j} \hat{\eta}_{j+1}^{z} \hat{\eta}_{j}^{z},$$

 $\tilde{x} = n_s - \frac{\sin^2(\pi n_s)}{\pi^2 n_s}, \qquad \qquad \tilde{x}' = \frac{\sin(2\pi n_s)}{2\pi} - \frac{\sin^2(\pi n_s)}{\pi^2 n_s},$ 

 $\tilde{y} = n_{\eta} - \frac{\sin^2(\pi n_{\eta})}{\pi^2 n_n}, \qquad \qquad \tilde{y}' = \frac{\sin(2\pi n_{\eta})}{2\pi} - \frac{\sin^2(\pi n_{\eta})}{\pi^2 n_n}.$ 

3-site terms

$$J_{\text{ex}}^{\text{s}} = (\tilde{x} - \tilde{x}')J_{\text{ex}}$$
$$J_X^{\eta} = (\tilde{y} - \tilde{y}')J_{\text{ex}}$$
$$J_Z^{\eta} = -(\tilde{y} - \tilde{y}')J_{\text{ex}} + 4\tilde{y}V$$

 $n_s$ : Density of singly occupied sites  $n_n$ : Density of doublons and holons

 $\triangleright$  spin and  $\eta$ -spin are separated

Exchange couplings are renormalized

Summary

2-site terms

$$|\Psi\rangle = |\Psi_{\mathrm{SF}}^{\mathrm{GS}}\rangle |\Psi_{\mathrm{spin}}^{\mathrm{GS}}\rangle |\Psi_{\eta-\mathrm{spin}}^{\mathrm{GS}}\rangle$$
  
 $H_{\mathrm{SF,free}} = H_{\mathrm{spin}}^{\mathrm{(SQ)}} = H_{\eta-\mathrm{spin}}^{\mathrm{(SQ)}}$ 

## Indication to nonequilibrium phases

YM, et al., PRL. **130**, 106501 (2023).

η-spin sectors

Described by the XXZ model

 $\Rightarrow$ 

Two types of phases

 $J_Z < J_X$  : **Gapless** phase of the XXZ model

η-pairing state with slowly decaying

 $\chi_{\text{pair}}(r) \equiv \langle \hat{\eta}^x(r) \hat{\eta}^x(0) \rangle$ 

 $\dot{X}$  Alternating sign in definition of  $\hat{\eta}_i^+ = (-)^i \hat{c}_{i\downarrow}^{\dagger} \hat{c}_{i\uparrow}^{\dagger}$ 



 $J_Z > J_X$ : **Gapful** phase of the XXZ model

CDW state with slowly decaying

 $\chi_{\rm charge}(r) \equiv \langle \hat{\eta}^z(r) \hat{\eta}^z(0) \rangle$ 

% Long range order in the squeezed  $\eta$  spin space

String type order cf. Haldane phase

#### 





## Phase diagram of the photo-doped states at $T_{eff} = 0$



- $\triangleright$  3 site terms favor  $\eta$  pairing phase
- $\triangleright$  Analytic argument well explains numerically obtained phase diagram for H<sub>eff2</sub> (no 3 site terms)
- ▷ Picture at U $\rightarrow$ ∞ works well even for finite U

#### Insight into total central charge

Total central charge ( $\mathbf{c}$ ) ~ Number of massless modes

$$|\Psi
angle = |\Psi_{\mathrm{SF}}^{\mathrm{GS}}
angle |\Psi_{\mathrm{spin}}^{\mathrm{GS}}
angle |\Psi_{\eta-\mathrm{spin}}^{\mathrm{GS}}
angle$$
  
 $H_{\mathrm{SF,free}} = H_{\mathrm{spin}}^{\mathrm{(SQ)}} = H_{\eta-\mathrm{spin}}^{\mathrm{(SQ)}}$ 

Charge (SF) sector: gapless

## Total central charge: iTEBD analysis for H<sub>eff2</sub>



c=3 in single-band Hubbard model is not expected in equilibrium

Emergence of extra degrees of freedom by photo-doping!

## Naïve expectation of single-particle spectrum

$$A_k(\omega) = -\frac{1}{\pi} \text{Im} G_k^R(\omega) \text{ with } G_k(t,t') = -i \langle \mathcal{T} c_k(t) c_k^{\dagger}(t') \rangle$$

Equilibrium doped system

Electron = charge (SF) degree + spin degree

gapless gapless

Gapless around Fermi level

Photo-doped system

Electron = charge (SF) degree + spin degree + η spin degreegaplessgaplessgaplessgaplessCDW: gapful

η pairing phase : Gapless around Fermi level ?

CDW phase : Gapful around Fermi level ?

## Single partici

## ring state and CDW state <sup>25</sup>



#### Summary



Review on nonequilibrium Mott insulators: YM, D Golež, M Eckstein, P Werner, arXiv:2310.05201

## Supplement

#### Previous analysis : metastable η pairing phase



#### Indication to spin properties

$$\begin{split} \Psi \rangle &= |\Psi_{\rm SF}^{\rm GS}\rangle |\Psi_{\rm spin}^{\rm GS}\rangle |\Psi_{\eta-{\rm spin}}^{\rm GS}\rangle \\ & \overline{H_{\rm SF,free}} \ \overline{H_{\rm spin}^{\rm (SQ)}} \ \overline{H_{\eta-{\rm spin}}^{\rm (SQ)}} \end{split}$$

SF and spin part is independent of the ratio between  $N_h$  and  $N_d$ .

➡ Chemical doping and photo-doping have the same effect on spin correlations

cf. DMFT results



J. Mentink & M. Eckstein PRL 113 057301 (2014).

#### Nonequilibrium phase diagram @ U = 10, $J_{ex} = 0.4$



Quasi-long range order

 $\chi(r) \propto \cos(qr)/r^a$  with  $q = \pi (\eta$ -SC)  $q = 2n_d \pi (\text{CDW})$   $q = (1 - 2n_d)\pi (\text{SDW})$ 

▷ Boundary of  $\eta$ SC and CDW = V=J<sub>ex</sub>/2

Special kinematics of doublons and holons in one dimensional system

#### Long-life time of photo-carriers and their relaxation



#### Cooling of carriers in Photo-doped Mott

#### **Cluster DMFT study**



M. Eckstein & P. Werner Sci. Rep. 6 21235 (2015)



## 光誘起されたMott絶縁体の理論研究

NESS @ coupling with heat and particle bath

J. Li, et. al., PRB **102**, 165136 (2020).
J. Li and M. Eckstein, PRB **103** 045133 (2021).



- ▷ Transient state ≔ NESS
- ▷ NESS ← Effective temp + doping level description looks good

#### Analysis with infinite boundary condition



FIG. 5. Calculated single-particle excitation spectra of the 1DEHM at (a), (e), (i)  $\Delta t_{\rm pr} = -\infty$  (GS); (b), (f), (j)  $\Delta t_{\rm pr} = 0$ ; and (c), (g), (k)  $\Delta t_{\rm pr} = 8$ . (d), (h), (l) TDOSs at  $\Delta t_{\rm pr} = -\infty$  (black solid line) and  $\Delta t_{\rm pr} = 8$  (red dashed line). The on-site interaction is set to U = 10, and the intersite interaction, the pump-light frequency, and its intensity are set to (a)-(d) V = 0,  $\omega_0 = 8.0$ , and  $A_0 = 0.6$ ; (e)-(h) V = 3,  $\omega_0 = 6.04$ , and  $A_0 = 0.3$ ; and (i)-(l) V = 6,  $\omega_0 = 6.34$ , and  $A_0 = 0.3$ .

