

LINEAR OPTICAL SPECTRUM IN STRONGLY CORRELATED SYSTEMS ANALYZED BY NEW APPROACH TO EXTRACT IMPORTANT DEGREES OF FREEDOM USING SINGULAR VALUE DECOMPOSITION

A. Takahashi

Nagoya Institute of Technology

coworkers

J. Tokimoto

S. Ohmura

K. Iwano

H. Okamoto

K. Nakanishi

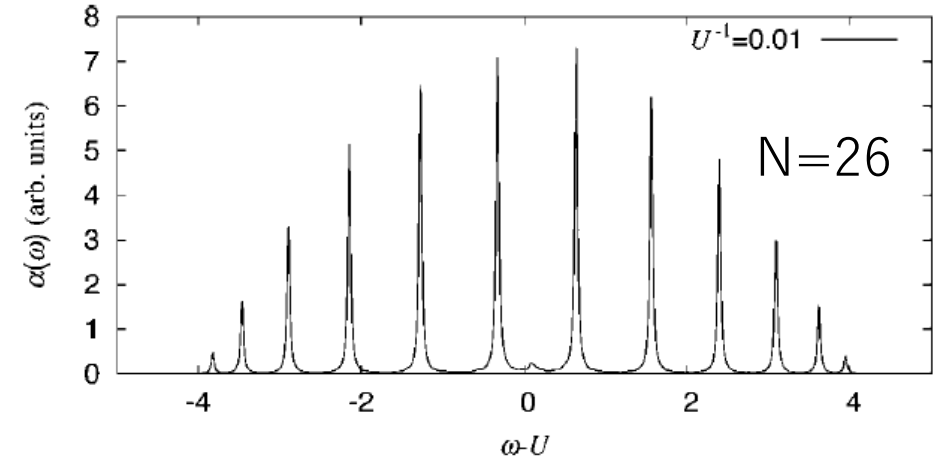
supported by JST CREST in Japan (Grant No. JPMJCR1661)

outline

- Motivation
- singular value decomposition (SVD) of coefficient matrix
extraction of the important degrees of freedom in quantum dynamics
- Light absorption spectrum and photoexcited states in 2D Mott insulators
- Light absorption spectrum and photoexcited states in charge-ordered insulators

In strongly correlated electron systems, it is not easy to understand their physical properties from the light absorption spectrum.

Ex. The light absorption spectrum for the 2D Mott insulators is essentially different from that of the 1D ones



[Phys. Rev. B **73**, 075110 \(2006\).](#)

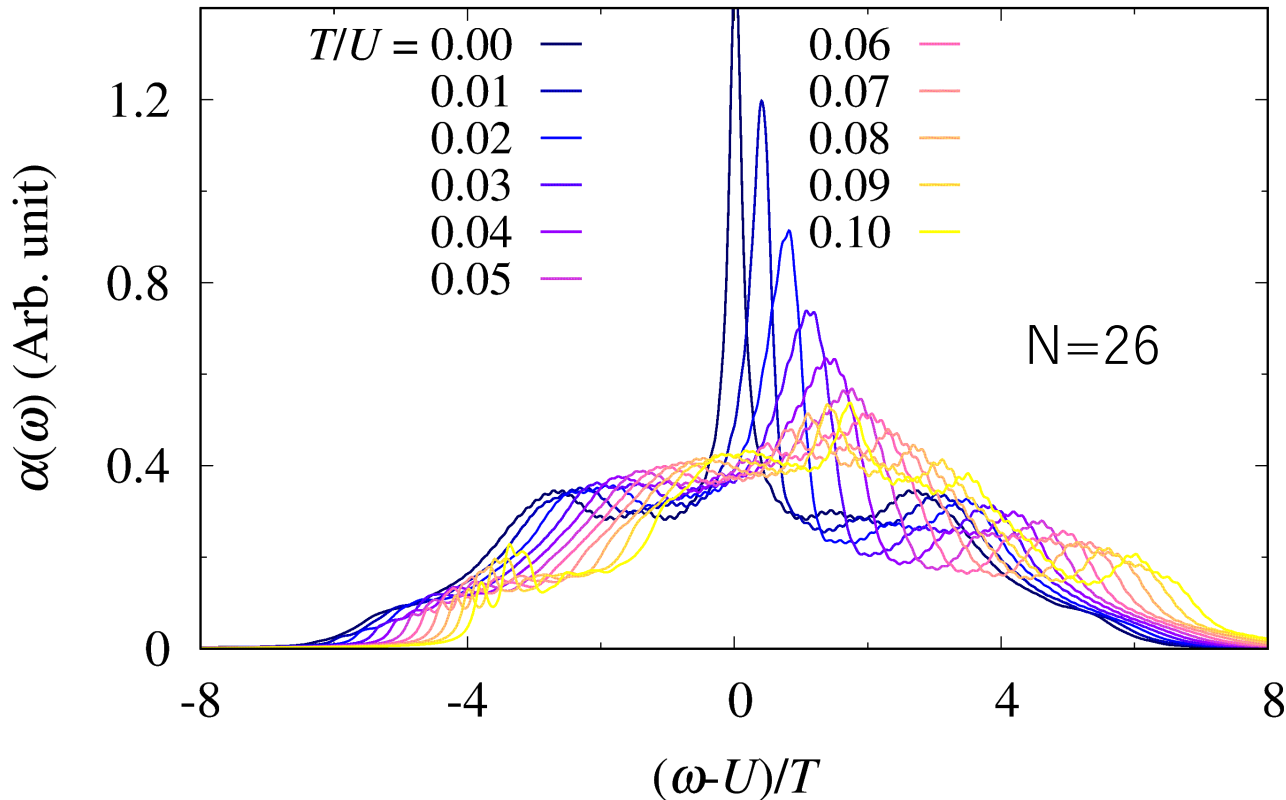
1D Mott insulators

- Light absorption spectrum consists a few isolated peaks in the small size cluster.
- The spectrum shape is almost unchanged when U/T is changed in the strong correlation region $U/T \geq 10$.

separation between spin and charge degrees of freedom

[Phys. Rev. B **100**, 235134 \(2019\).](#)

2D Mott insulators



[Phys. Rev. B **109**, 195150 \(2024\).](#)

We propose a new approach to extract the important degrees of freedom in the solution of the time-dependent Schrödinger equation using singular value decomposition (SVD) of the coefficient matrix.

- a band is formed
- The spectrum shape is strongly dependent on U/T .

The strong dimensionality dependence is closely related to the difference in the spin-charge coupling between 1D and 2D Mott insulators.

Light absorption spectrum is an important stage to investigate the spin-charge coupling.

Method of calculations

- Coefficient matrix

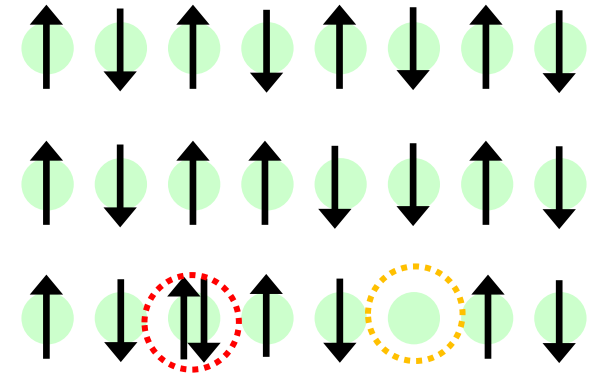
Numerical solution of time-dependent Schrödinger equation

$$|\psi(t)\rangle = \sum_{l=1}^{N_b} c_l(t) |\varphi_l\rangle$$

coefficient matrix $N_t \times N_b$

$$C = \begin{bmatrix} c_1(t_1) & c_2(t_1) & \cdots & c_{N_b}(t_1) \\ c_1(t_2) & c_2(t_2) & \cdots & c_{N_b}(t_2) \\ \vdots & \vdots & \ddots & \vdots \\ c_1(t_{N_t}) & c_2(t_{N_t}) & \cdots & c_{N_b}(t_{N_t}) \end{bmatrix}$$

$|\varphi_l\rangle$: basis states



N_b : dimension of the Hilbert space

N_t : number of time steps

$$t_j = t_0 + \Delta t j$$

- singular value decomposition (SVD) of coefficient matrix

$$C = U\Sigma V^\dagger$$

$U : N_t \times R$ unitary matrix

$\Sigma : R \times R$ diagonal matrix

$V : N_b \times R$ unitary matrix

R : rank of C

$$\Sigma = \begin{bmatrix} \sigma_1 & 0 & \cdots & 0 \\ 0 & \sigma_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \sigma_R \end{bmatrix}$$

singular values $\sigma_1 \geq \sigma_2 \geq \cdots \geq \sigma_R \geq 0$

The solution is given by the linear combination of modes $|\Phi_k\rangle$.

$$|\psi(t_j)\rangle = \sum_{k=1}^R U_{j,k} \overset{\text{weight}}{\sigma_k} |\Phi_k\rangle$$



$K \ll R \leq N_b$ holds in most cases.

$$|\Phi_k\rangle = \sum_{l=1}^{N_b} V_{l,k}^* |\varphi_l\rangle$$

$$\sigma_1 \gg \sigma_{K+1}$$

$$|\psi(t_j)\rangle \approx \sum_{k=1}^K U_{j,k} \sigma_k |\Phi_k\rangle$$

linear combination of
far fewer important modes
than N_b

Important degrees of freedom for the dynamics are extracted by SVD.

effective Hamiltonian of 2D Hubbard model with 26 system size

$$N_b = 1.7 \times 10^7$$

$$N_t = 6000$$

C is a giant matrix.

SVD of C cannot be done by conventional algorithms



Randomized SVD (RSVD) *N. Halko, P.-G. Martinsson, and J. A. Tropp, SIAM Review 53, 217 (2011).*

RSVD provides **practically rigorous** SVD for the **important modes** where the singular values are not negligible.

Randomized SVD enables SVD of giant matrices

N. Halko, P.-G. Martinsson, and J. A. Tropp, SIAM Review 53, 217 (2011).

flow of RSVD

If we have Q that satisfies

$$A \approx QQ^T A$$

$m \times n$ $m \times k$ k : target rank
($k \ll m$)

SVD of $k \times n$ matrix

$$Q^T A = \tilde{U} \Sigma V^T$$



$$A \approx Q \tilde{U} \Sigma V^T$$



$$U = Q \tilde{U}$$

$$A \approx U \Sigma V^T \text{ SVD is completed}$$

How to find the matrix Q

w_r random vectors of m -dimension
 $1 \leq r \leq k$

$$y_r = A w_r$$

orthogonalization
 q_r

$$Q = (q_1, q_2, \dots, q_k)$$

Vector y_r is linearly independent with high probability and well reflects the structure of matrix A

(Johnson-Lindenstrauss theorem)

Model

2D Hubbard Hamiltonian

$$H = \hat{K} + \hat{V}$$

$$\hat{K} = -T \sum_{\langle n,m \rangle} \sum_{\sigma} (c_{n,\sigma}^{\dagger} c_{m,\sigma} + \text{H.c.})$$

transfer term
- T is the transfer integral
between these nearest-neighbor sites

$$\hat{V} = U \sum_n c_{n,\uparrow}^{\dagger} c_{n,\uparrow} c_{n,\downarrow}^{\dagger} c_{n,\downarrow}$$

Coulomb interaction term
 U is the on-site Coulomb repulsion energy

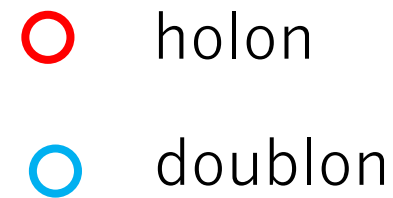
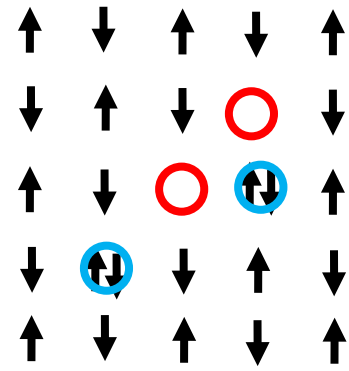
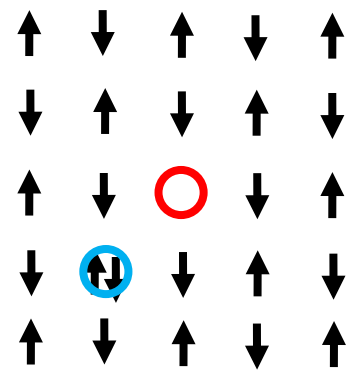
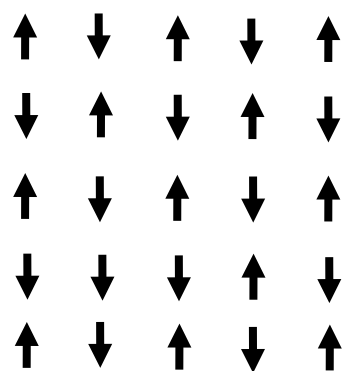
Finite-size effect \rightarrow we need to use the system size $N \geq 26$ to consider $\alpha(\omega)$.
SVD of the coefficient matrices is practically impossible to perform with $N \geq 26$
even when using the RSVD method.



effective Hamiltonian of the Hubbard model in the strong interaction case

Basis states in the half-filled case

$$T/U=0$$



1 H-D pair

2 H-D pairs

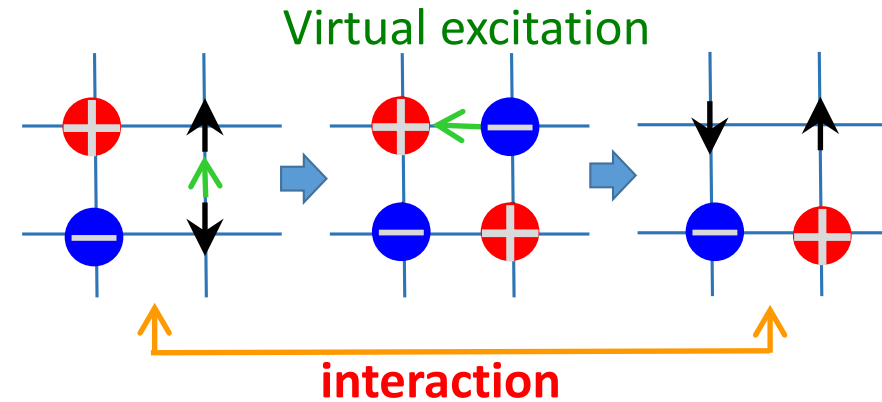
$$E_n=0$$

$$E_n=U$$

$$E_n=2U$$

effective model of the 2D Hubbard model in the strong interaction case $U \gg T$

Incorporate interactions through virtual electronic excitations that changes the number of H-D pairs perturbatively (up to the second order of T/U)



*A. Takahashi, S. Yoshikawa, and M. Aihara,
Phys. Rev. B **65**, 085103 (2002)*

effective Hamiltonian for one-photon excited states (one H-D pair)

$$H_{\text{eff}} = P_1 \hat{K} P_1 - U^{-1} P_1 \hat{K} P_2 \hat{K} P_1 + U^{-1} P_1 \hat{K} P_0 \hat{K} P_1$$

P_m : projection onto the space with m H-D pairs

effective Hamiltonian for the ground state (zero H-D pair)

$$H_{\text{eff}} = -U^{-1} P_0 \hat{K} P_1 \hat{K} P_0$$

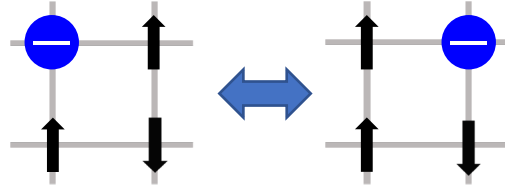
Heisenberg spin Hamiltonian

effective Hamiltonian for one-photon excited states (one H-D pair)

$$H_{\text{eff}} = P_1 \hat{K} P_1 - U^{-1} P_1 \hat{K} P_2 \hat{K} P_1 + U^{-1} P_1 \hat{K} P_0 \hat{K} P_1$$

P_m : projection onto the space with m H-D pairs

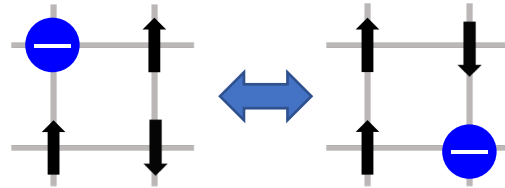
- $P_1 \hat{K} P_1 \propto T$
the transfer of H and D to nearest neighbor spin sites



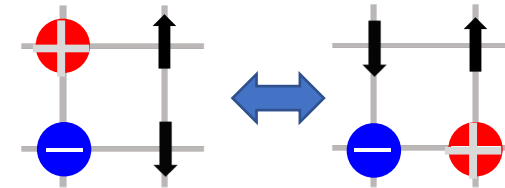
- terms $\propto J = 4T^2/U$

AF Heisenberg interaction $J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$

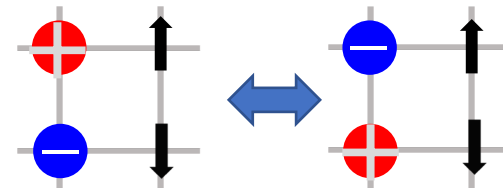
a virtual three-site transfer



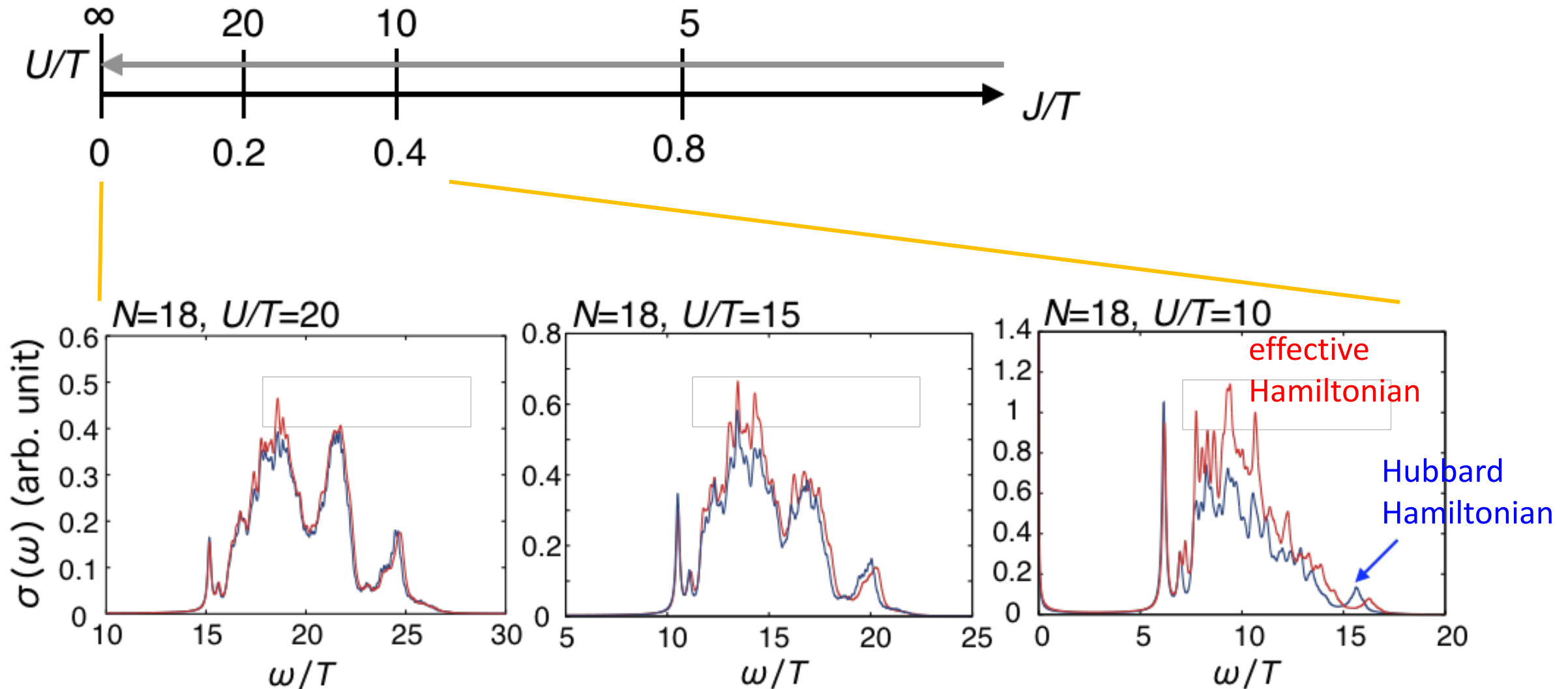
the transfer of an H-D pair on nearest-neighbor site



the exchange of an H and a D on nearest-neighbor sites



The optical conductivity calculated by using the original Hubbard model can be reproduced by using the effective model for $U/T \gtrsim 10$.



We numerically solve time-dependent Schrödinger equation

$$i \frac{\partial}{\partial t} |\psi(t)\rangle = H_{\text{eff}} |\psi(t)\rangle$$

system size $N=26$

initial condition

$$|\psi(0)\rangle = \hat{J} |\phi_0\rangle, \quad |\phi_0\rangle \quad : \text{ground state, } \hat{J} = \hat{\mathbf{J}} \cdot \mathbf{e}_x$$

light field is polarized
in the x -direction.

$$\hat{\mathbf{J}} = -iT \sum_{\langle n,m \rangle, \sigma} (\mathbf{r}_n - \mathbf{r}_m) (c_{n,\sigma}^\dagger c_{m,\sigma} - c_{m,\sigma}^\dagger c_{n,\sigma}) \quad : \text{current operator}$$



the dynamics when the ground state $|\phi_0\rangle$ is excited
by an ultrashort weak light pulse that is polarized along the x -direction.

We consider the coefficient matrix C in the time range when light is off.

We adopt large enough time range $T_I = N_t \Delta t$. $N_t = 6000, \Delta t T = 0.1$

uniqueness of the SVD



Each of the SVD modes is identical to an energy eigenstate. $|\phi_k\rangle = |\Phi_k\rangle$

$$C = U \Sigma V^\dagger$$

$V \rightarrow$ SVD modes = all the important energy eigenstates

$U \rightarrow$ energy eigenvalues

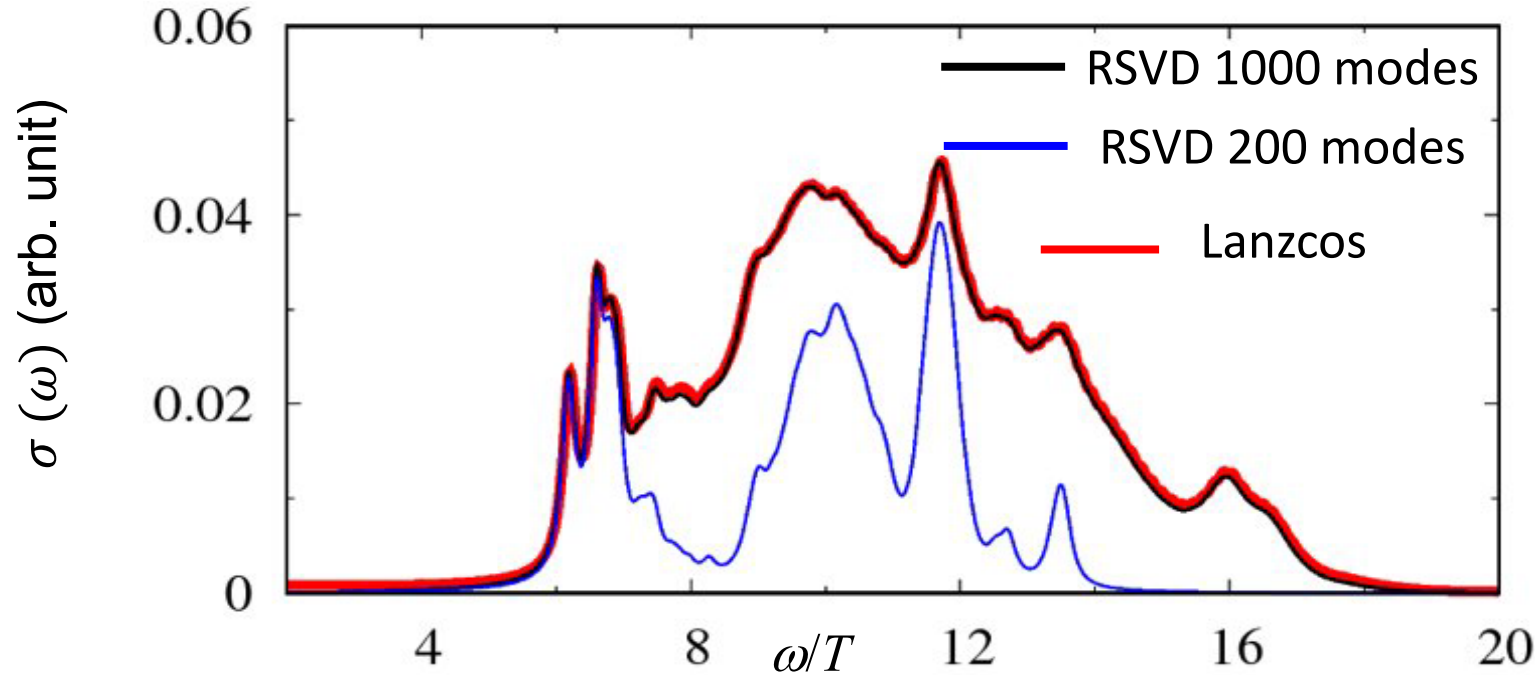
Fourier transformation

$\Sigma \rightarrow$ transition dipole moments : $J_{k,0} = \frac{1}{\sqrt{N_t}} \sigma_k$



light absorption
spectrum
optical conductivity

extraction of important degrees of freedom for the dynamics by SVD



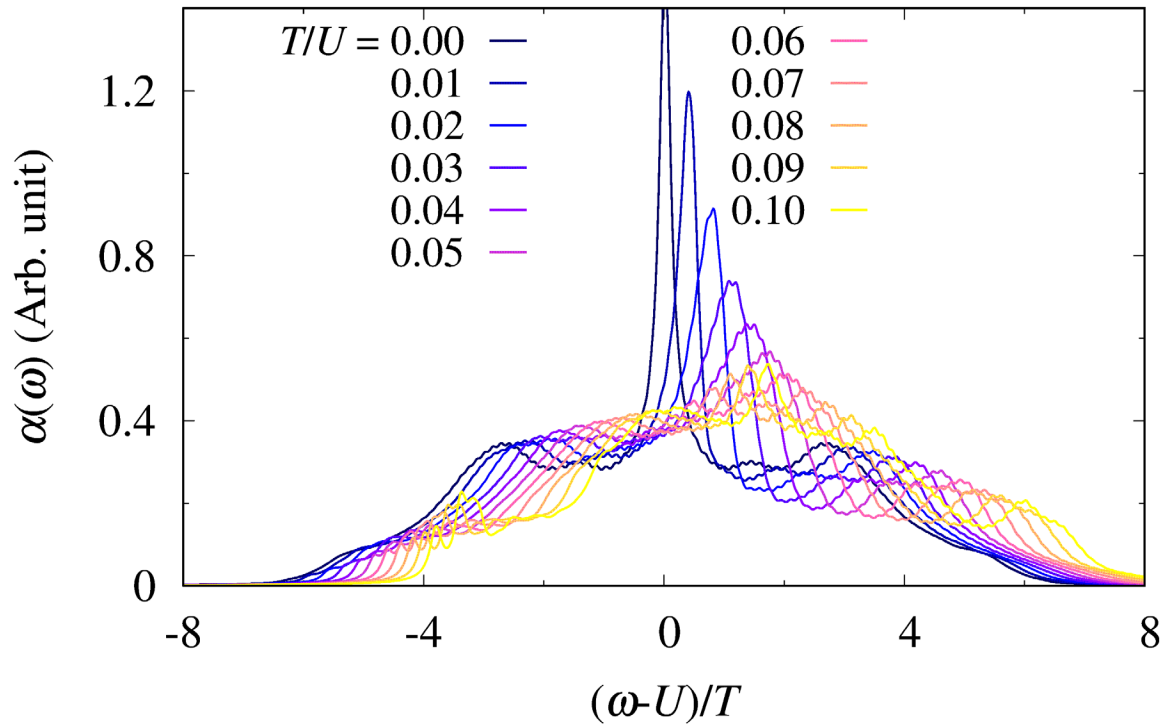
system size $N = 26$

$U/T = 10$

(typical magnitude of Coulomb interaction in Mott insulators)

- Optical conductivity can be reproduced almost exactly by considering **1000** important energy eigenstates ($K=1000$) in 1.7×10^7 -dimension Hilbert space.
 - All the energy eigenstates that contribute to $\sigma(\omega)$ are obtained by SVD.
- ↓
- Important degrees of freedom for linear optical spectrum are extracted by SVD.
 - The number (K) of important modes is far fewer than Nb

2D Mott insulators



- a band is formed
- The spectrum shape is strongly dependent on U/T .

The strong dimensionality dependence is closely related to the difference in the spin-charge coupling between 1D and 2D Mott insulators.

important stages to investigate the coupling

New approach

We calculate all the energy eigenstates that have non-negligible contribution to $\alpha(\omega)$, and investigate their physical properties.

spin correlation for $|\phi_k\rangle$

$$\eta(\mathbf{r}_n - \mathbf{r}_m) = \langle \phi_k | \mathbf{S}_n \cdot \mathbf{S}_m | \phi_k \rangle$$

spin structure factor

$$S(\mathbf{q}) = \sum_m \eta(\mathbf{r}_m) \exp[i\mathbf{q} \cdot \mathbf{r}_m]$$

$$I_{\text{AF}} = N^{-1} \{ S(\pi, \pi) - \eta(0) \}$$

the quantity that represents the magnitude of the AF spin order

entanglement entropy

$$S = -\text{Tr}_A [\rho_A \ln(\rho_A)]$$

$$\rho_A = \text{Tr}_B [\rho], \quad \rho = |\phi_k\rangle\langle\phi_k|$$

charge correlation for $|\phi_k\rangle$

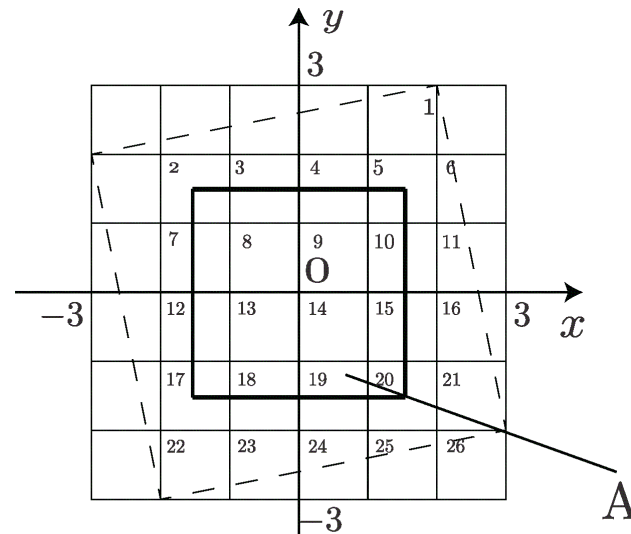
$$\xi(\mathbf{r}_n - \mathbf{r}_m) = \begin{cases} \langle \phi_k | (1 - n_n)(1 - n_m) | \phi_k \rangle & \text{for } n \neq m \\ 0 & \text{for } n = m \end{cases}$$

$$n_m = \sum_{\sigma} c_{m,\sigma}^{\dagger} c_{m,\sigma}$$

$-\xi(\mathbf{r})$: D density at the site \mathbf{r} when H exists at the site (0,0)

σ_c : standard deviation of D density

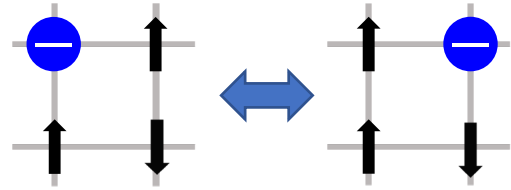
$D(E)$: density of states



Results in the strong correlation limit $U \rightarrow \infty$ ($T/U=0$)

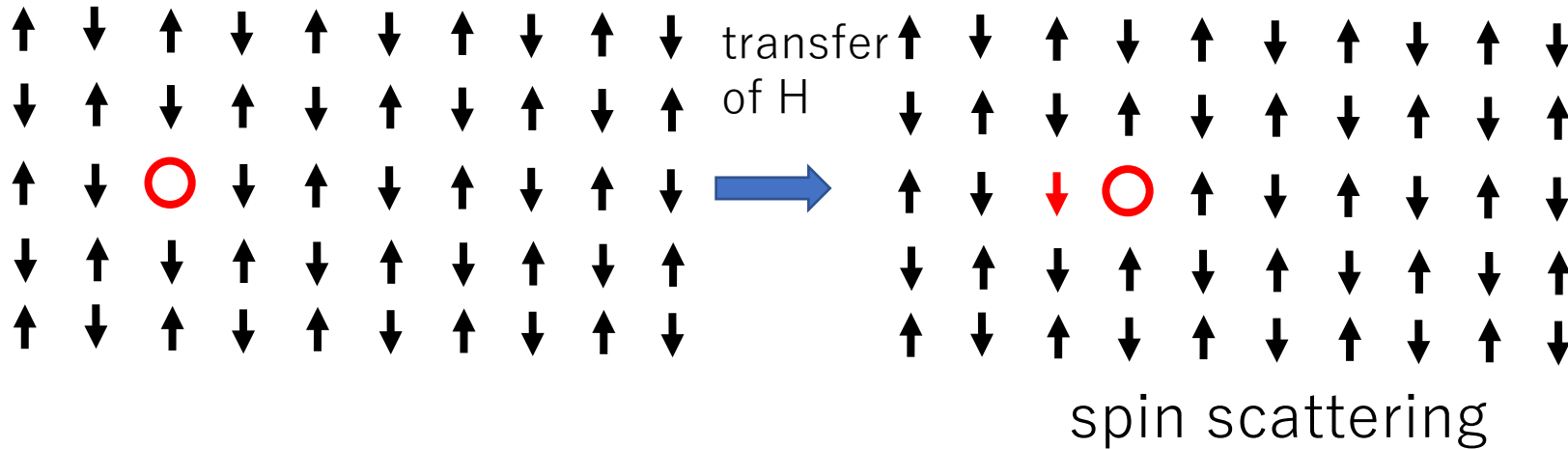
$$H_{\text{eff}} = P_1 \hat{K} P_1$$

- $P_1 \hat{K} P_1 \propto T$
the transfer of H and D to
nearest neighbor spin sites



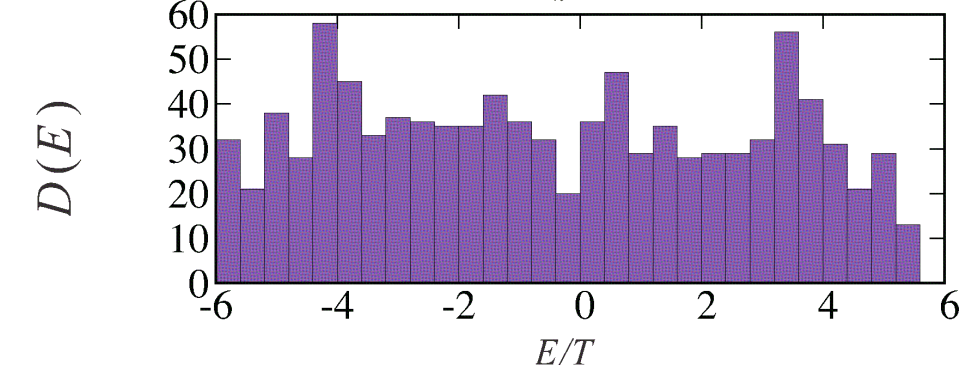
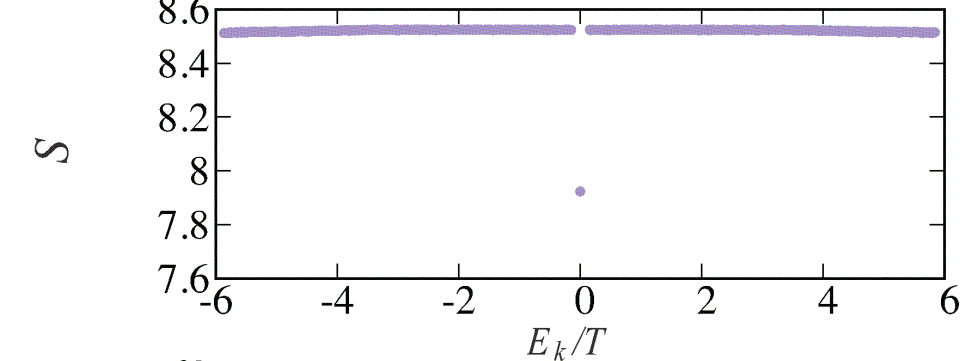
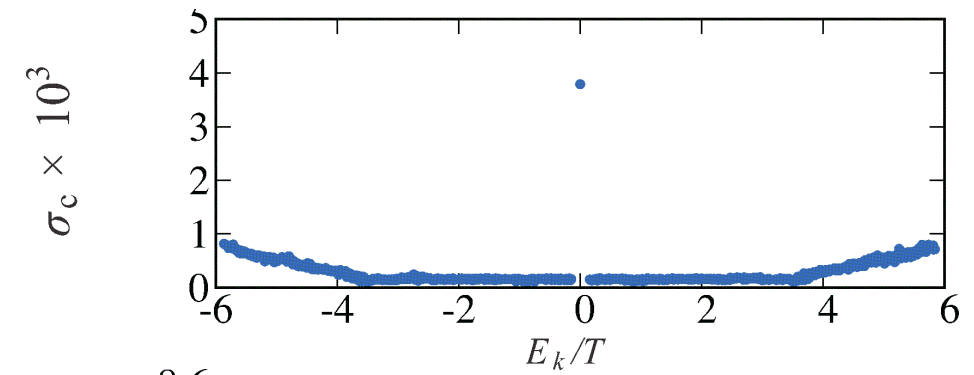
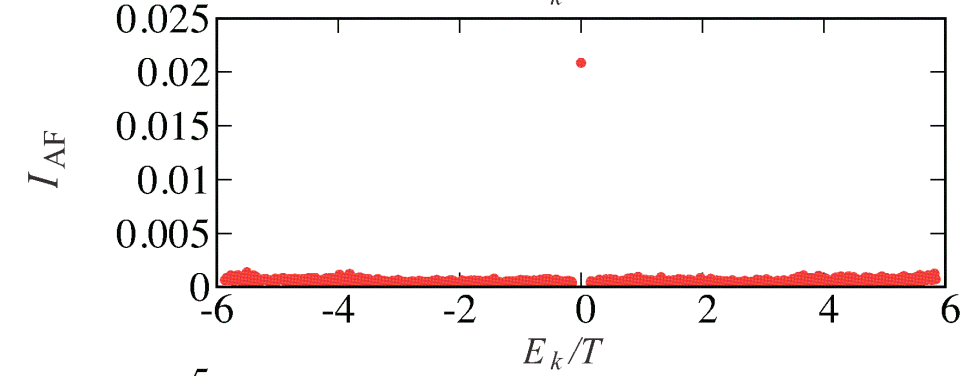
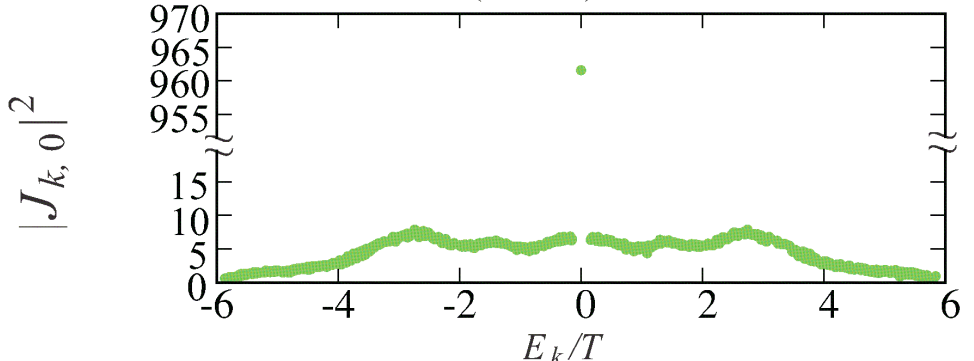
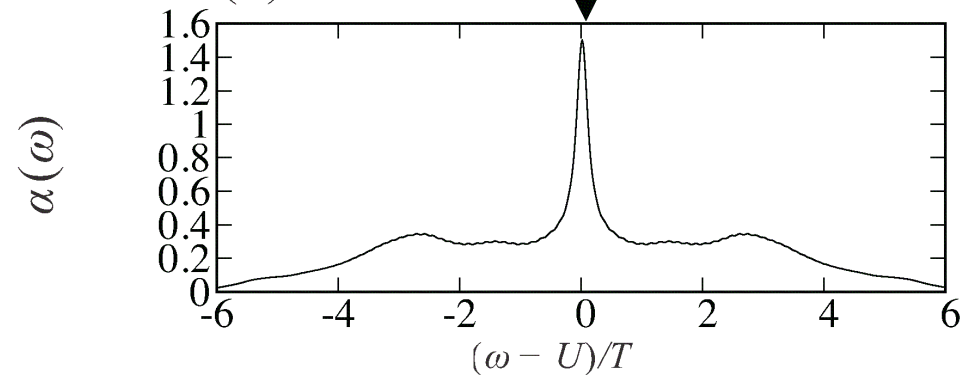
Spin-spin interaction is NOT included.

H_{eff} is not a simple two-body Hamiltonian of H and D.

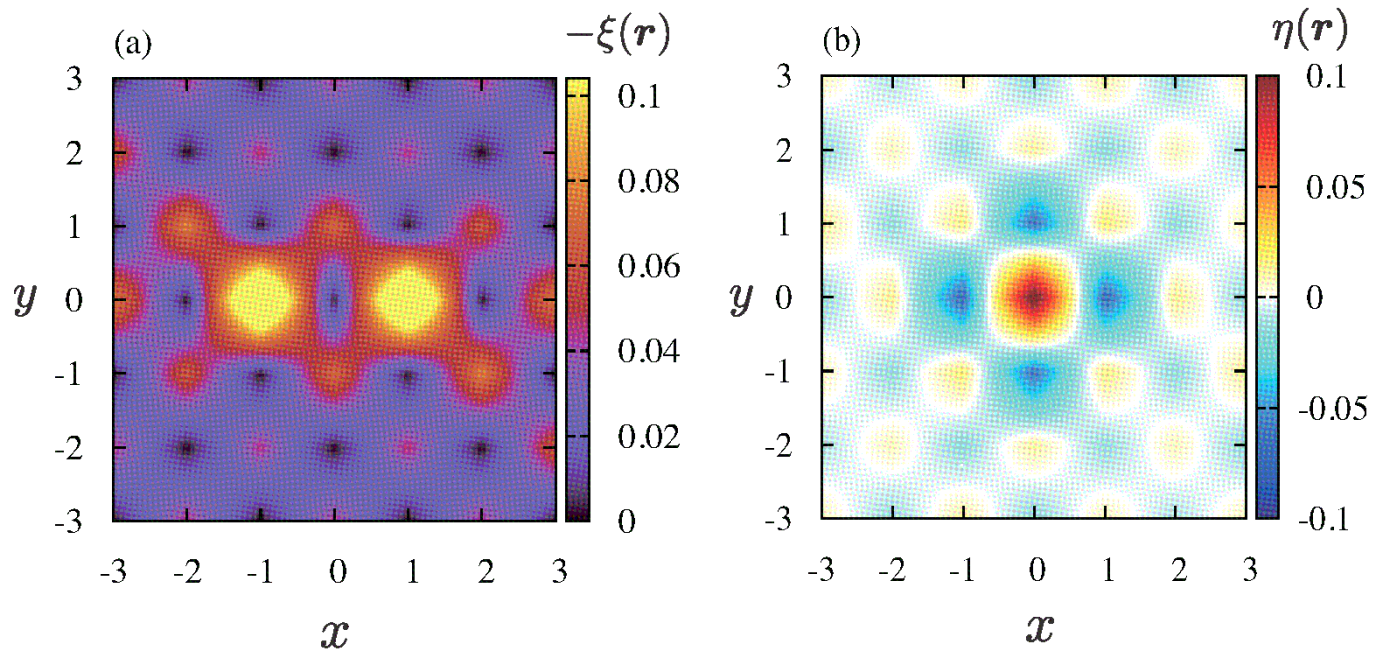


spin-charge coupling

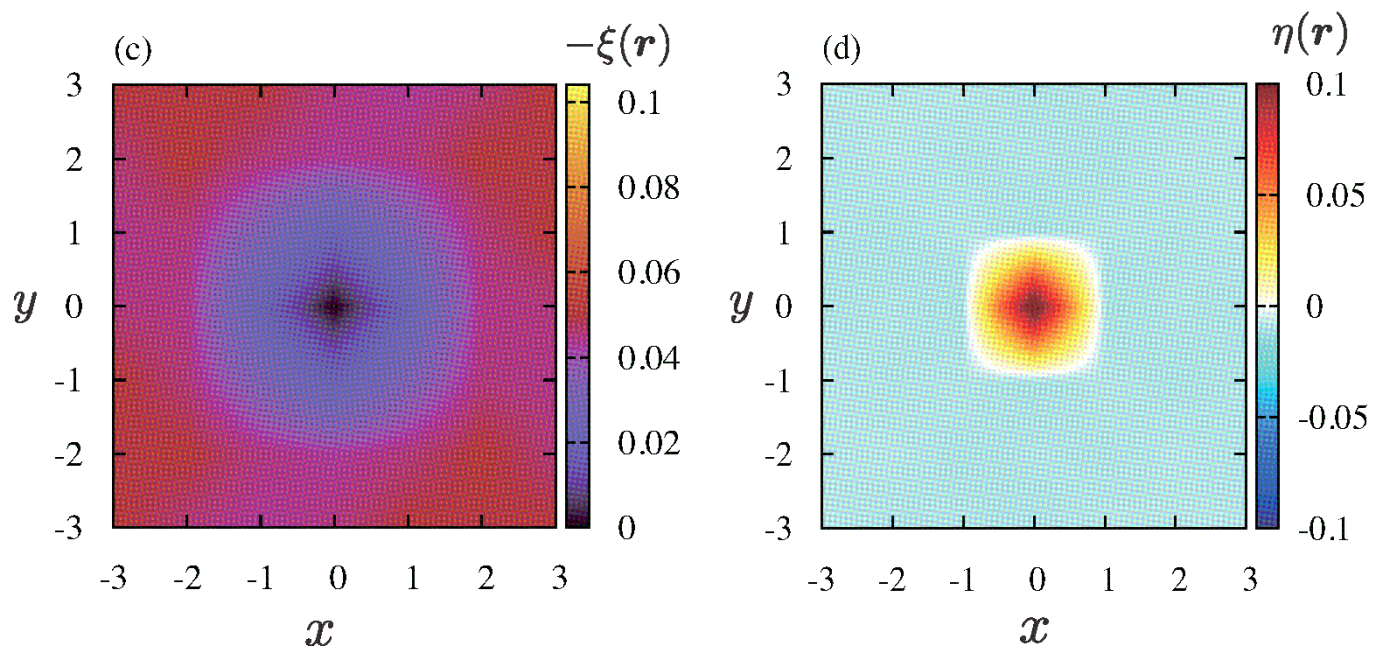
(a) $T/U = 0$



$E_k=0$

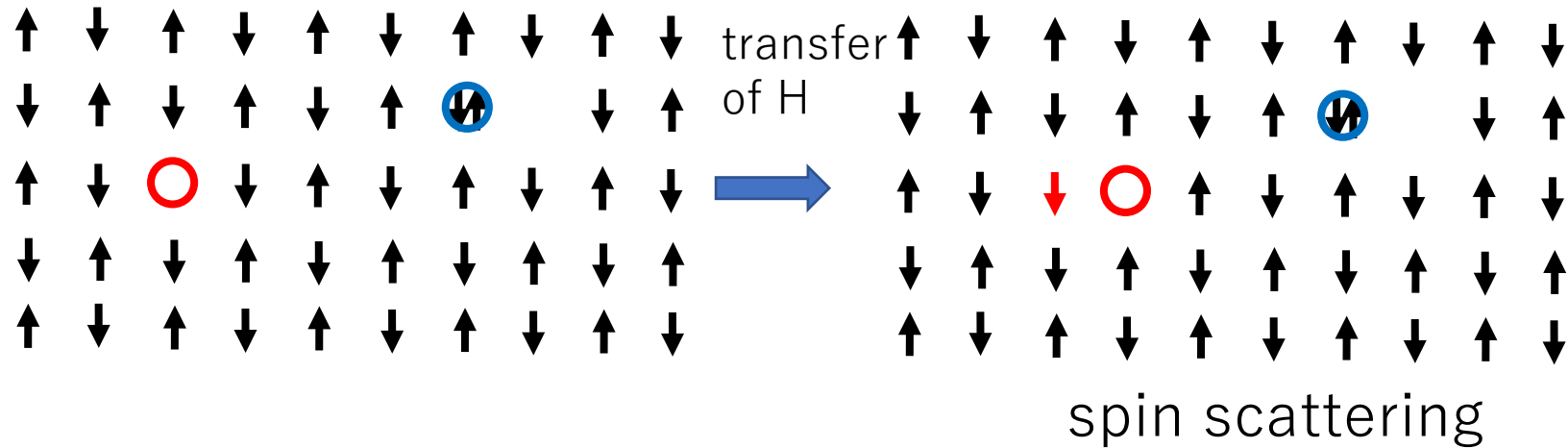


$E_k=5.85T$



free H and D states

- (i) the interaction between a H and a D induced by the the spin scattering is negligible
- (ii) the transfer of a H (D) induces only the phase shift in the spin wave function.



phase $\exp\left[i\left(q_x + \Delta q_x(s)\right)\right]$ is added.

↑
↑
 holon spin

free H and D states

H and D move freely with the constraint that H and D cannot occupy the same site. The main effect of the spin degrees of freedom on the transfer of H and D is the phase shift.

$$|\psi_{q,s}\rangle = \frac{\sqrt{2}}{N} \sum_{n,m} \sin[\mathbf{q} \cdot (\mathbf{r}_n - \mathbf{r}_m)] |f_s(n,m)\rangle$$

H at site n and D at site m

$|f_s(n,m)\rangle$: the spin wave function with H at site n and D at site m

$$E_{q,s} = -2T \left\{ \cos(q_x + \Delta q_x(s)) + \cos(q_x - \Delta q_x(s)) + \cos(q_y + \Delta q_y(s)) + \cos(q_y - \Delta q_y(s)) \right\}$$



consistent with the numerical results by SVD.

free H and D states

$$|\psi_{\mathbf{q},s}\rangle = \frac{\sqrt{2}}{N} \sum_{n,m} \sin[\mathbf{q} \cdot (\mathbf{r}_n - \mathbf{r}_m)] |f_s(n,m)\rangle$$

H at site n and D at site m

$|f_s(n,m)\rangle$: the spin wave function with H at site n and D at site m

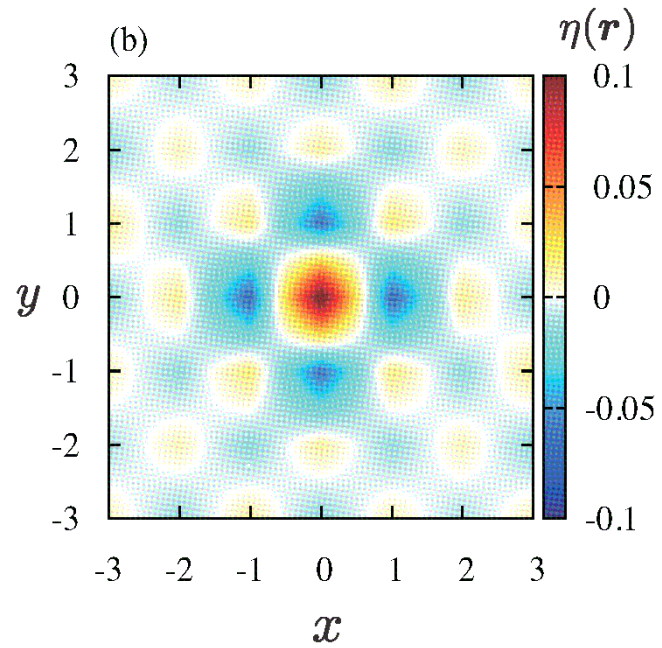
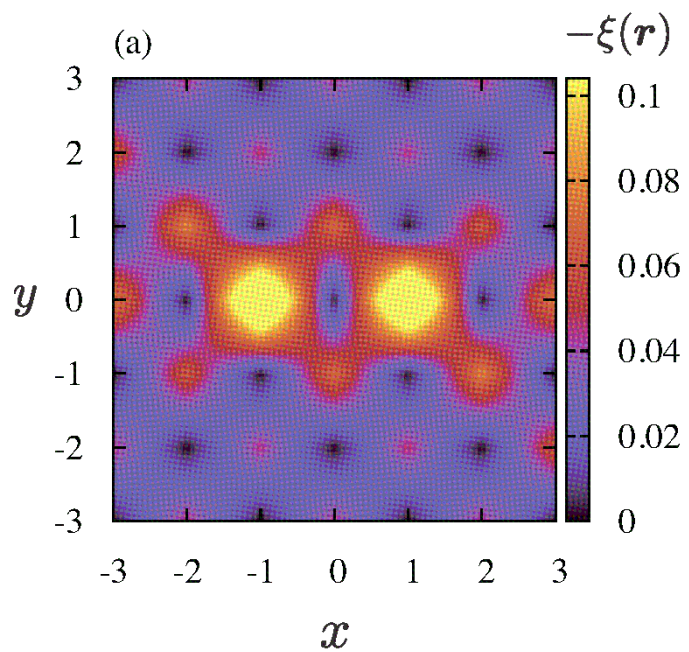
$$E_{\mathbf{q},s} = -2T \left\{ \cos(q_x + \Delta q_x(s)) + \cos(q_x - \Delta q_x(s)) + \cos(q_y + \Delta q_y(s)) + \cos(q_y - \Delta q_y(s)) \right\}$$

There are a few \mathbf{q} that are compatible with periodic boundary condition. If $\Delta\mathbf{q}(s) = 0$, light absorption spectrum consists of a few isolated peaks. Large number of spin degrees of freedom (s) \rightarrow a band is formed.

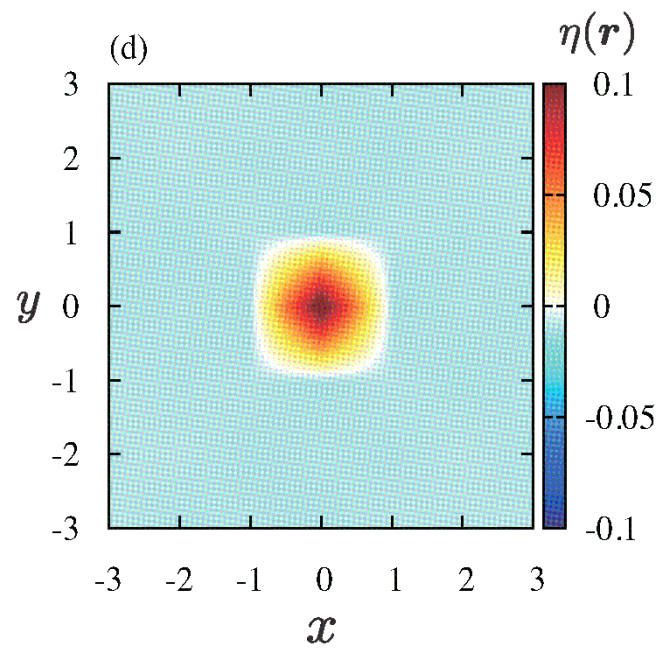
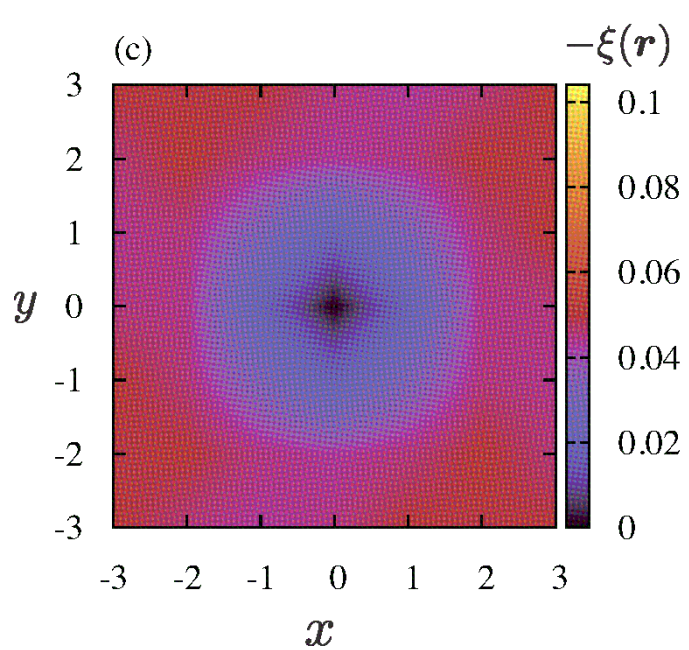
charge correlation $\xi(\mathbf{r}) = -\frac{4}{N^2} \sin^2[\mathbf{q} \cdot \mathbf{r}]$

$$E_k=0$$

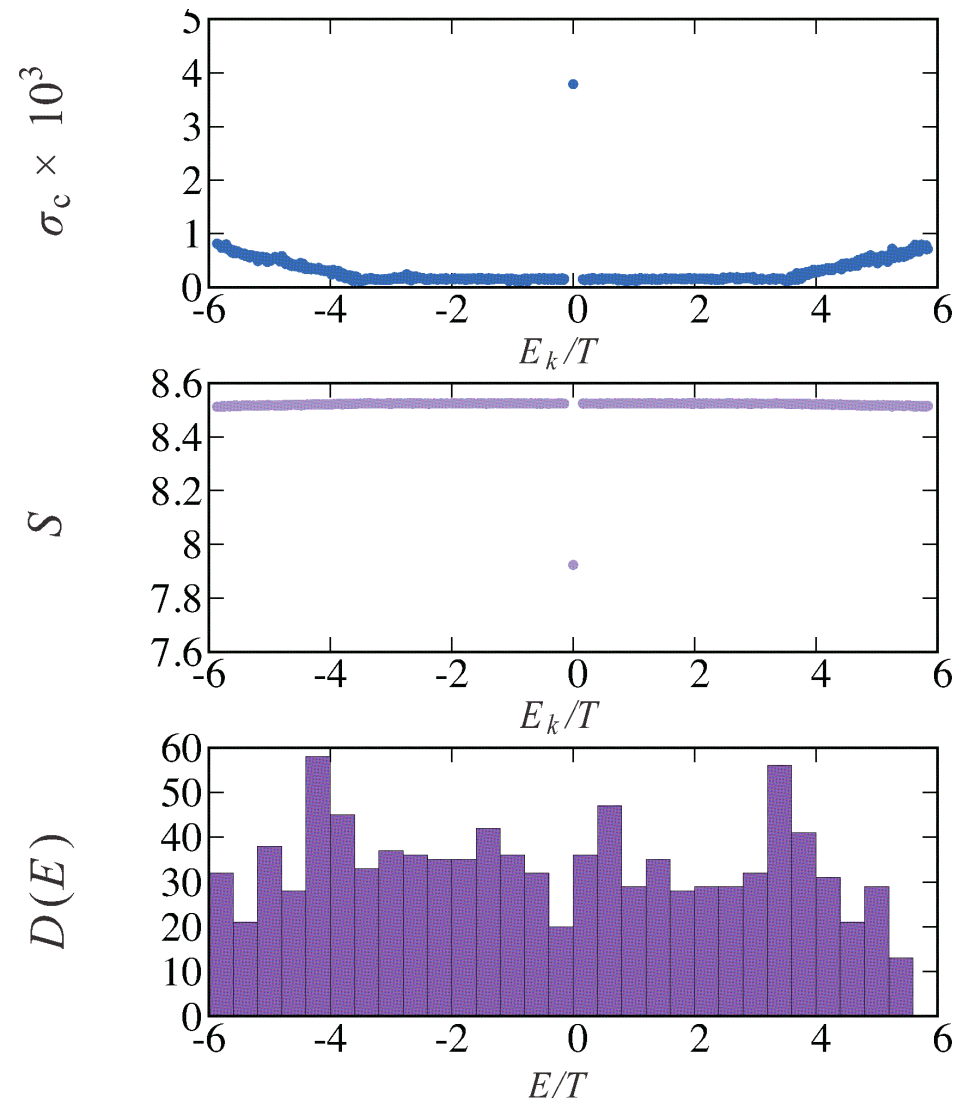
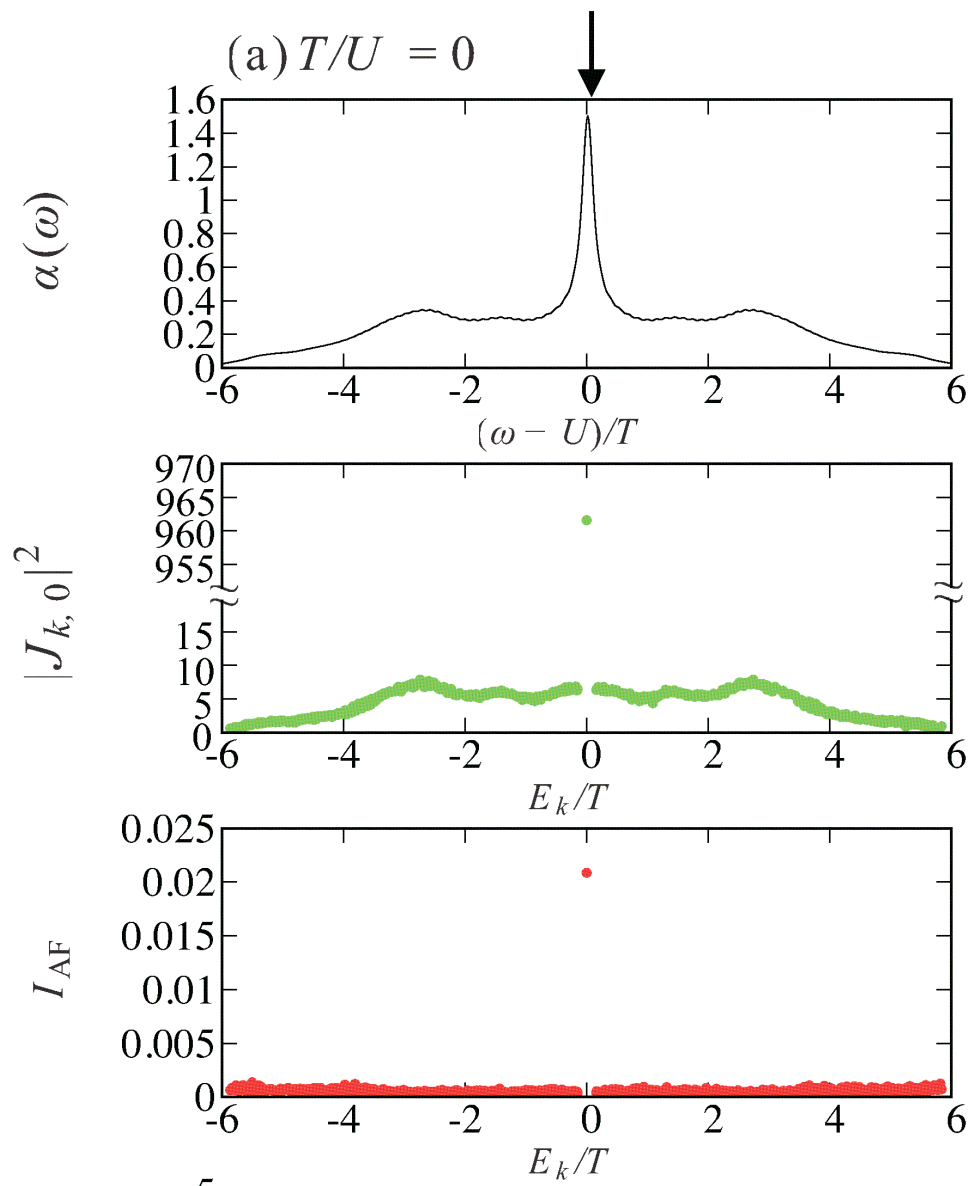
$$|q_x| = |q_y| = \frac{\pi}{2}$$



$$E_k=5.85T$$



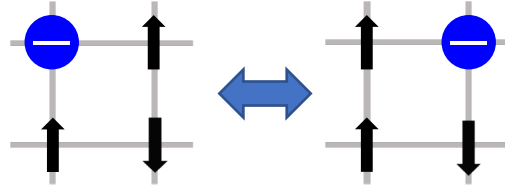
$$|q_x| = |q_y| = \frac{\pi}{2}$$



Results for the realistic magnitude of Coulomb interaction $T/U=0.1$

$$H_{\text{eff}} = P_1 \hat{K} P_1 - U^{-1} P_1 \hat{K} P_2 \hat{K} P_1 + U^{-1} P_1 \hat{K} P_0 \hat{K} P_1$$

- $P_1 \hat{K} P_1 \propto T$
the transfer of H and D to nearest neighbor spin sites

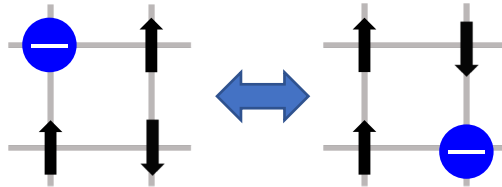


- terms $\propto J = 4T^2/U$

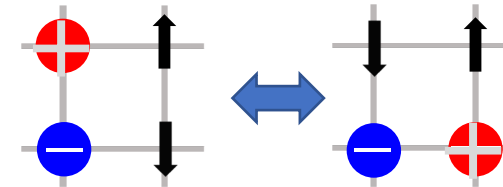
AF Heisenberg interaction

$$J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

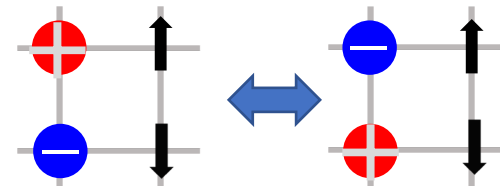
a virtual three-site transfer

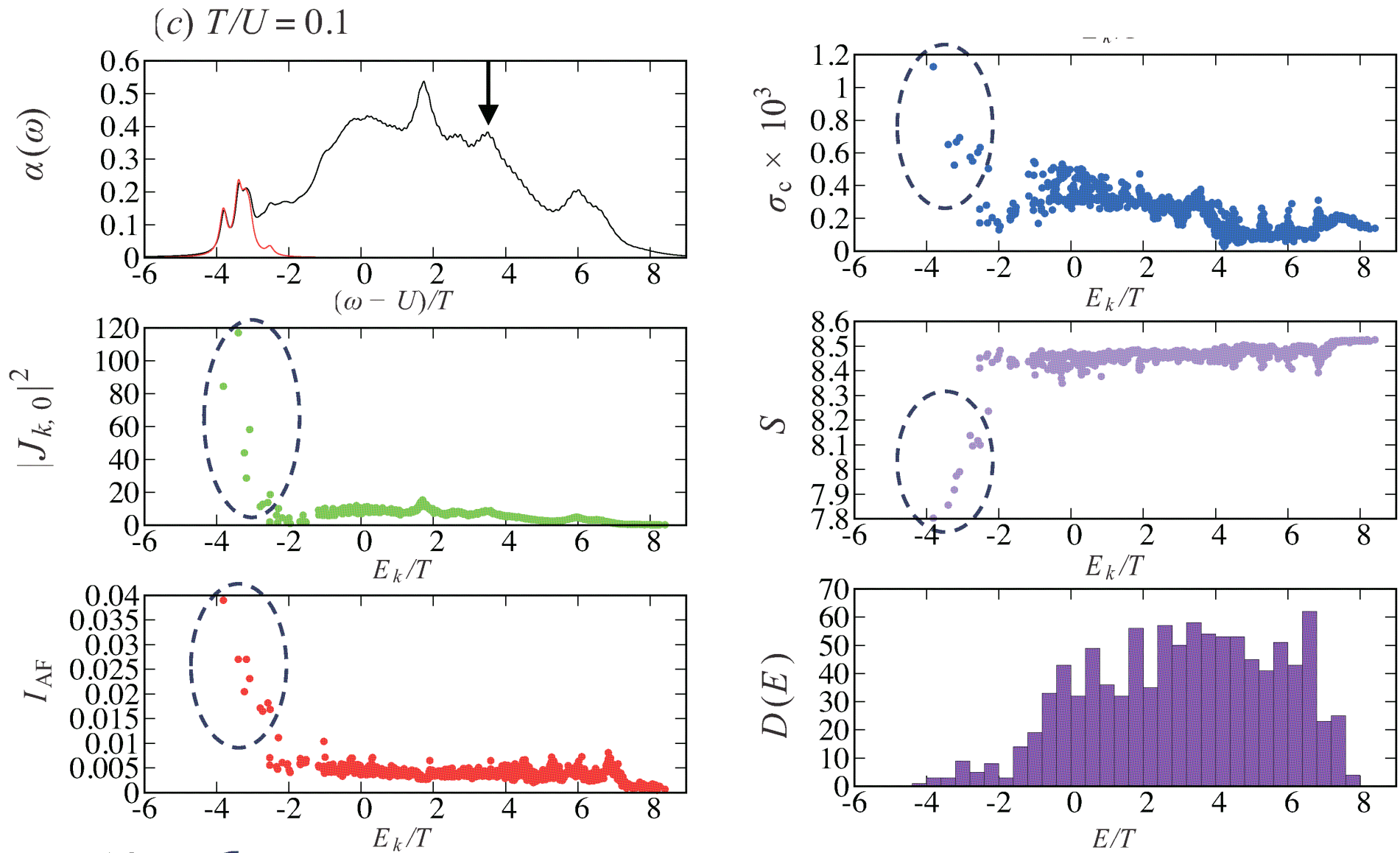


the transfer of an H-D pair on nearest-neighbor site



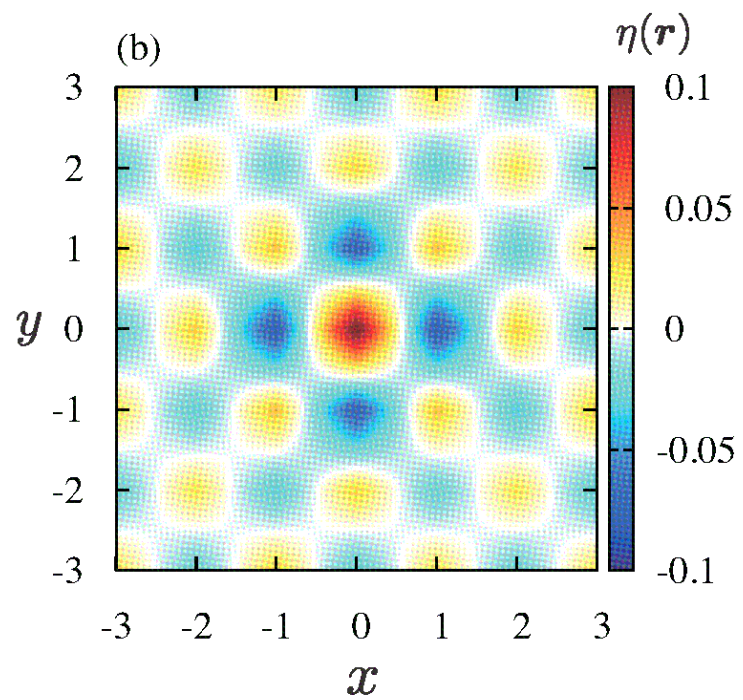
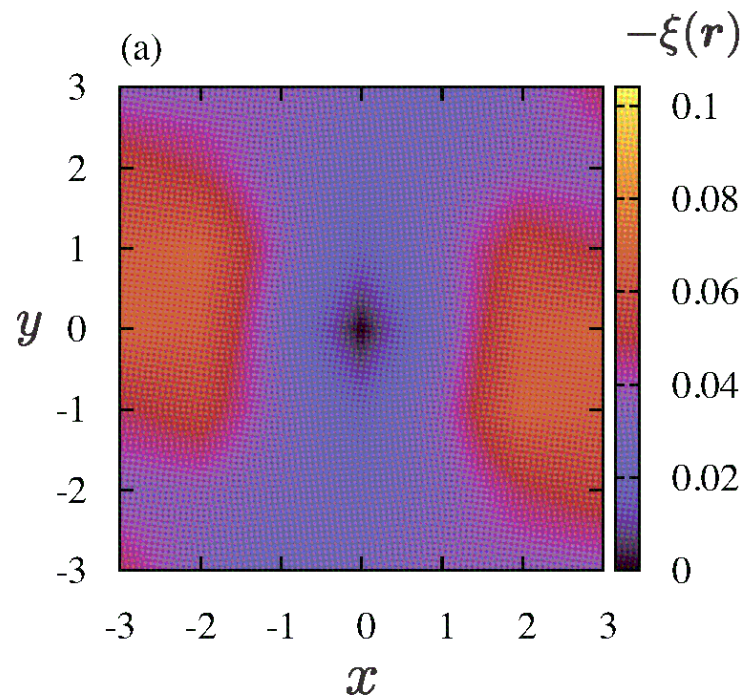
the exchange of an H and a D on nearest-neighbor sites



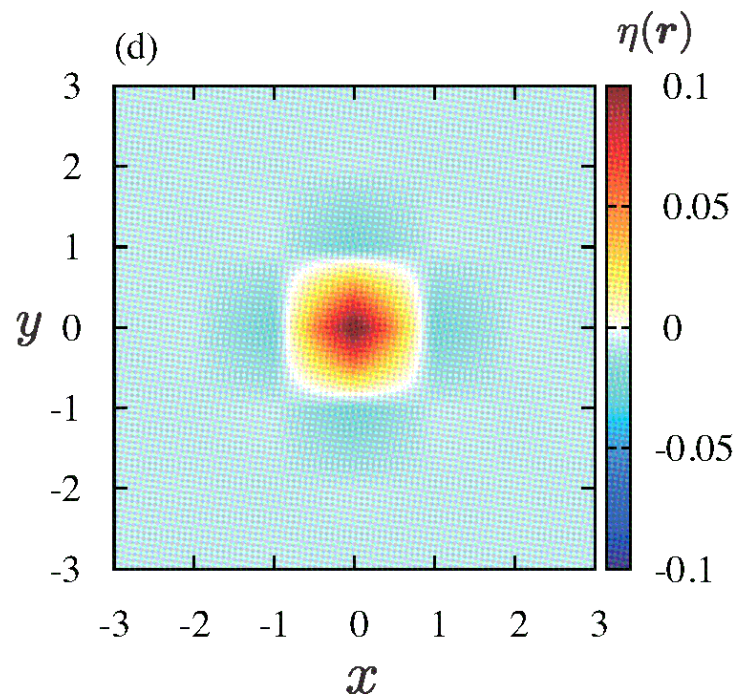
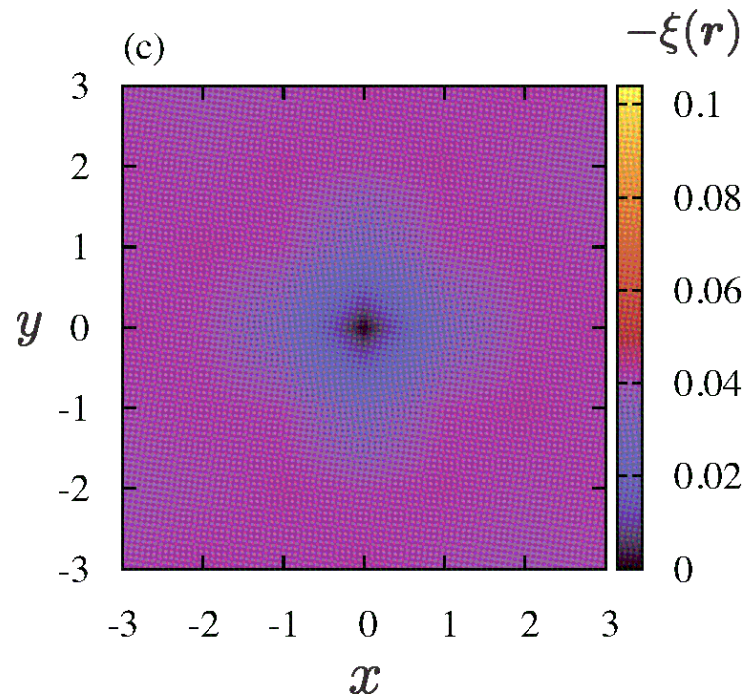


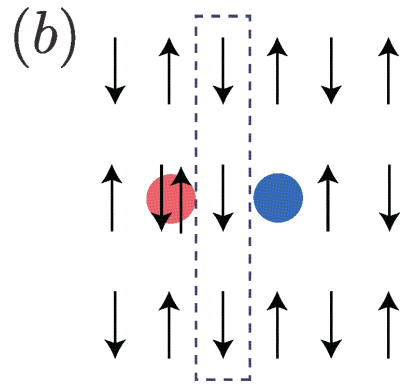
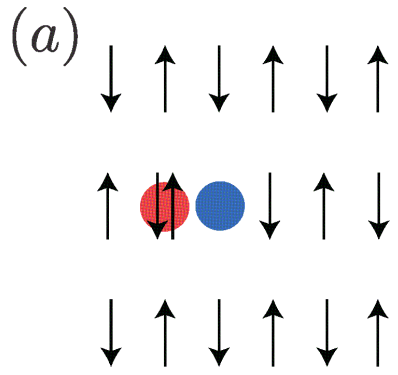
free H-D and localized H-D states

$$E_k = -3.82T$$



$$E_k = 1.69T$$



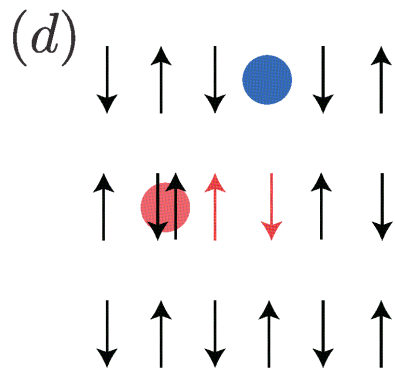
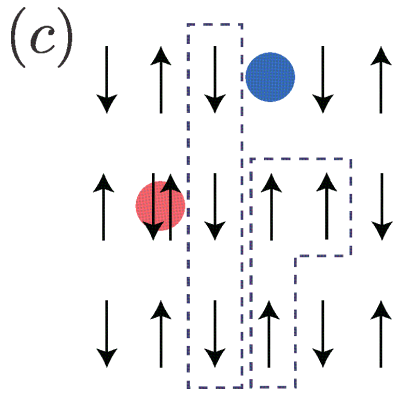


transfer of H(D) + spin flip



AF spin order is preserved when

$$r_{\text{H-D}} = 1, \sqrt{5}, \sqrt{13}, \dots$$



H-D interaction originating from spins
Local minimum of potential

localization of H and D at $r_{\text{H-D}} = 1, \sqrt{5}, \sqrt{13}, \dots$



AF spin order is not destroyed.

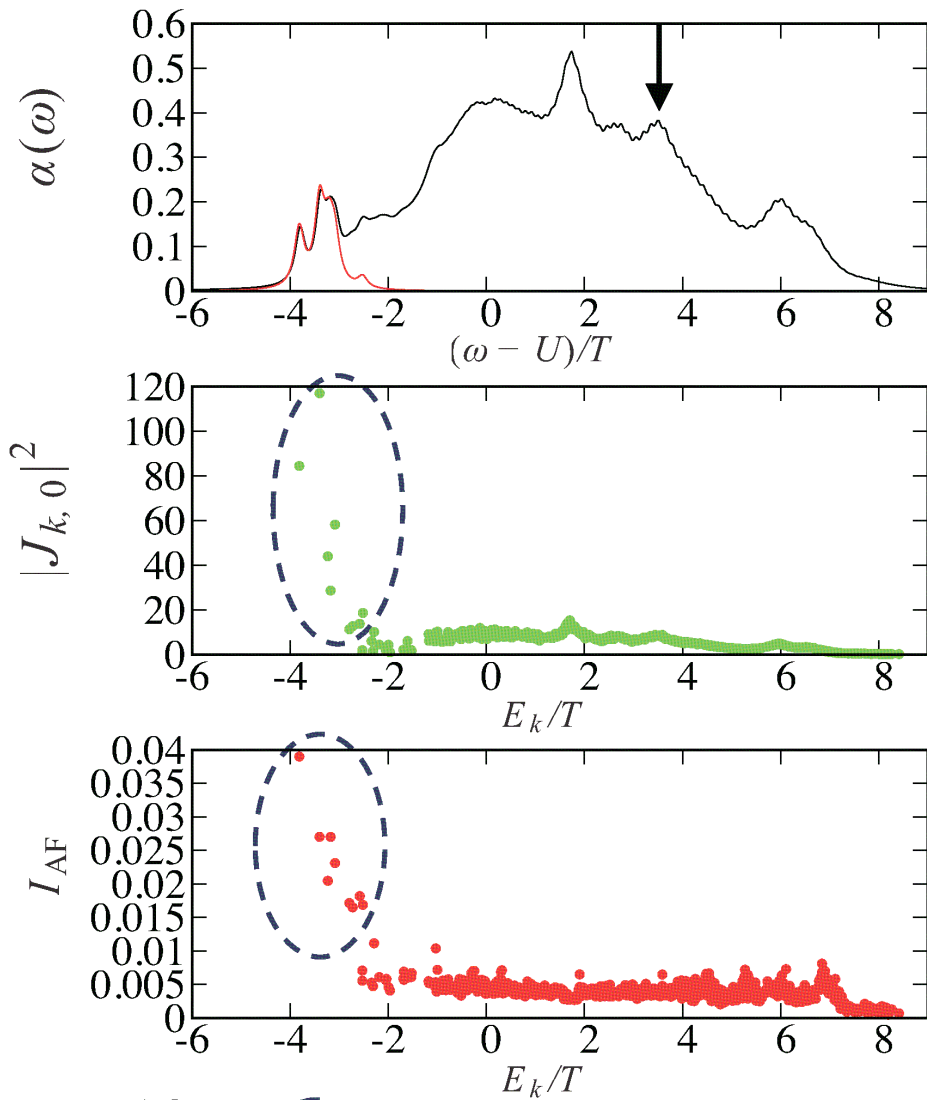
Energy gain due to the AF spin order

the attractive H-D interaction of spin origin

[Phys. Rev. B **104**, 205123 \(2021\).](#)

[Phys. Rev. B **106**, 075128 \(2022\).](#)

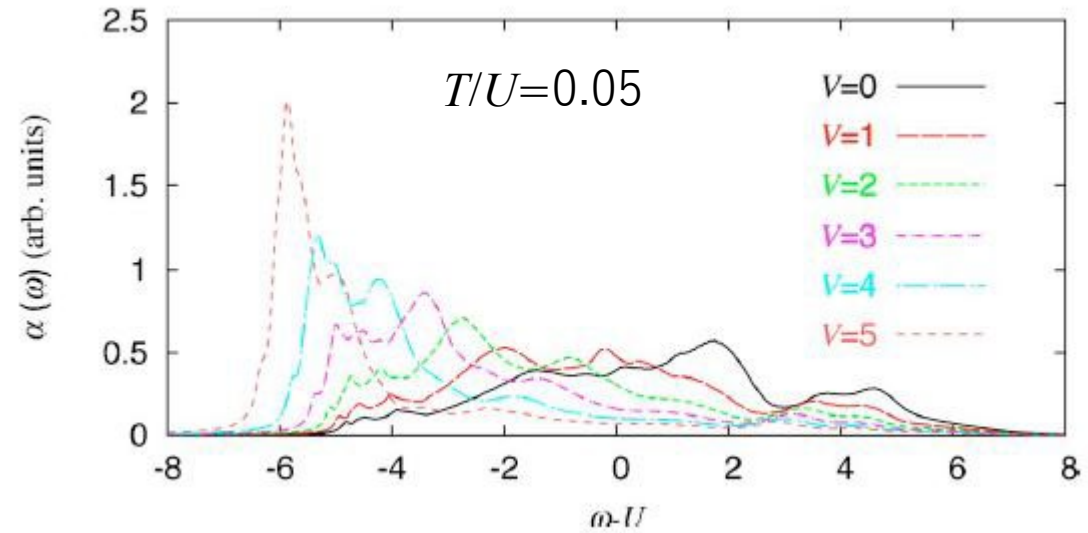
(c) $T/U = 0.1$



$$r_{\text{H-D}} = \sqrt{5} \text{ or } \sqrt{13}$$

for these localized H-D states

H-D bound state induced by V .



H-D exciton states exist for $V/T \geq 2$.
the peaks due to excitations to the
H-D exciton states are dominant

$$r_{\text{H-D}} = 1$$

conclusions

- Important degrees of freedom in the quantum dynamics induced by an external stimuli can be extracted by SVD of coefficient matrix.

In 2D Mott insulator

- Practically exact light absorption spectrum can be reproduced by as small as 1000 energy eigenstates in 1.7×10^7 -dimension Hilbert space.
- These photoactive energy eigenstates are classified into free H-D and localized H-D states.
- The localization of the charge in localized H-D states is of magnetic origin.

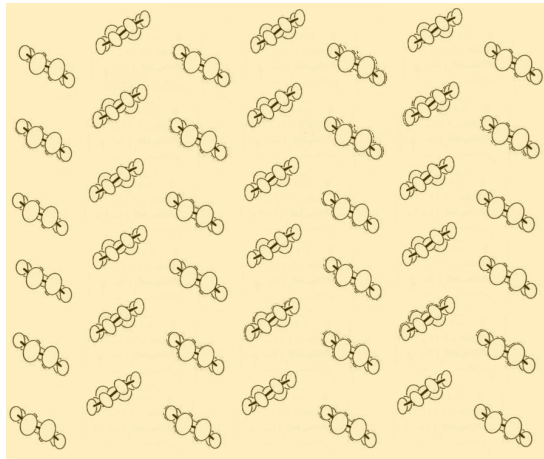
Subjects in future

- various systems, ex. phonon systems
- various stimuli, ex. magnetic field, quench
- search for unconventional photoexcited states
- strong excitation case
- photoinduced phase transitions
- analysis of transient absorption spectrum

charge ordered phase in α -(BEDT-TTF)₂I₃

2D strongly correlated electron system
 $\frac{1}{4}$ -filled, triangle lattice

metallic phase



charge ordered insulator phase



charge ordered phase in α -(BEDT-TTF)₂I₃

metallic phase

charge ordered insulator phase

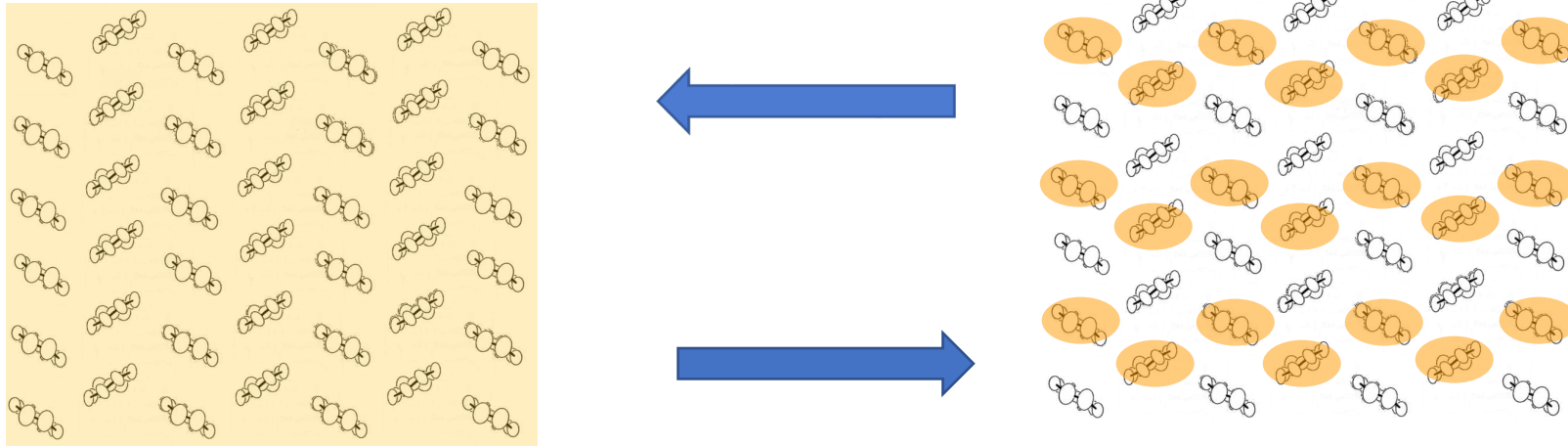
Photoinduced insulator-to-metal transition

S. Iwai, et al., PRL 98, 097402 (2007).

S. Iwai, et al., PRB 77, 125131 (2008).

H. Nakaya, et al., PRB 81, 155111(2010).

Y. Kawakami, et al., PRL 105, 246402 (2010).



a short-range charge order is induced

T. Ishikawa, et al., Nat. Commun. 5, 5528 (2014).

Y. Kawakami, et al., PRB 95, 201105 (2017).

dynamical localization?

Y. Kayanuma and K. Saito, Phys. Rev. A 77, 010101(R) (2008).

K. Nishioka and K. Yonemitsu, J. Phys. Soc. Jpn 83, 024706 (2014).

quarter-filled extended Hubbard model on 2D triangular lattice

$$H = \sum_{n,m,\sigma} \beta_{n,m} c_{m,\sigma}^\dagger c_{n,\sigma} + U \sum_n n_{n,\uparrow} n_{n,\downarrow} + \frac{1}{2} \sum_{n,m} V_{n,m} n_n n_m$$

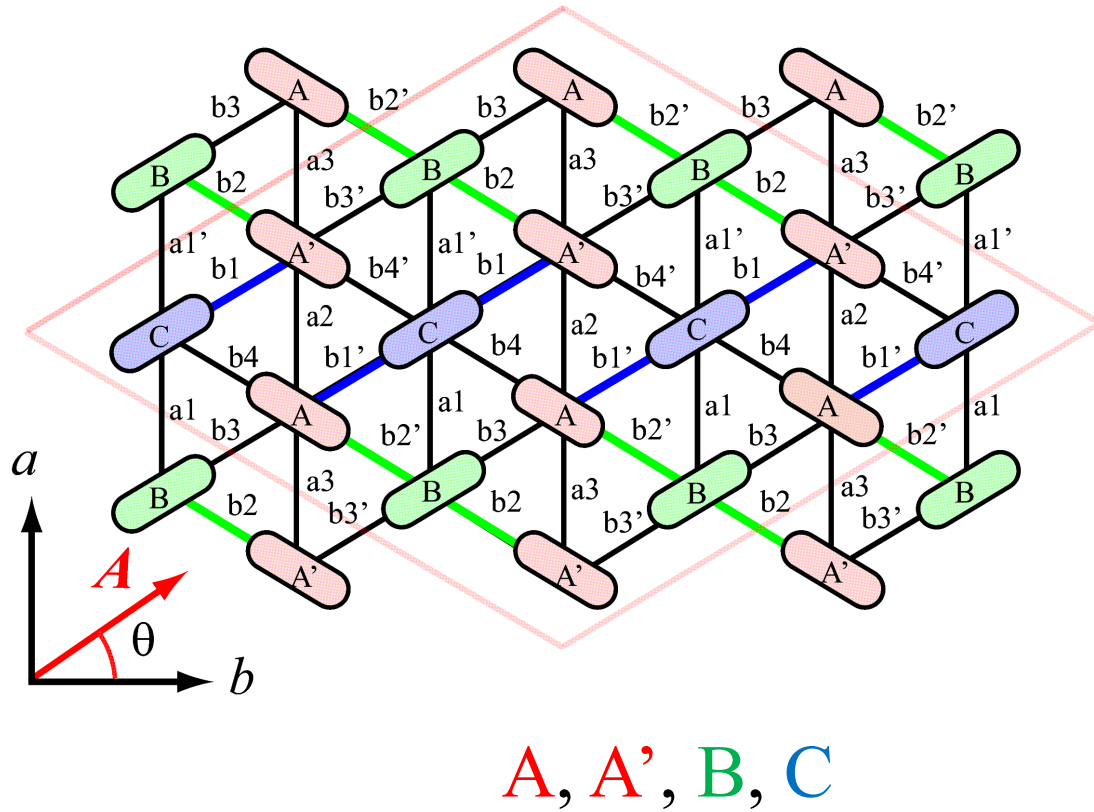
$$n_{m,\sigma} = c_{m,\sigma}^\dagger c_{m,\sigma}, \quad n_m = \sum_\sigma n_{m,\sigma}$$

$\beta_{n,m}$: transfer integral between neighboring sites

U : on-site Coulomb interaction energy

$V_{n,m}$: Coulomb interaction energy between neighboring sites

charge ordered phase

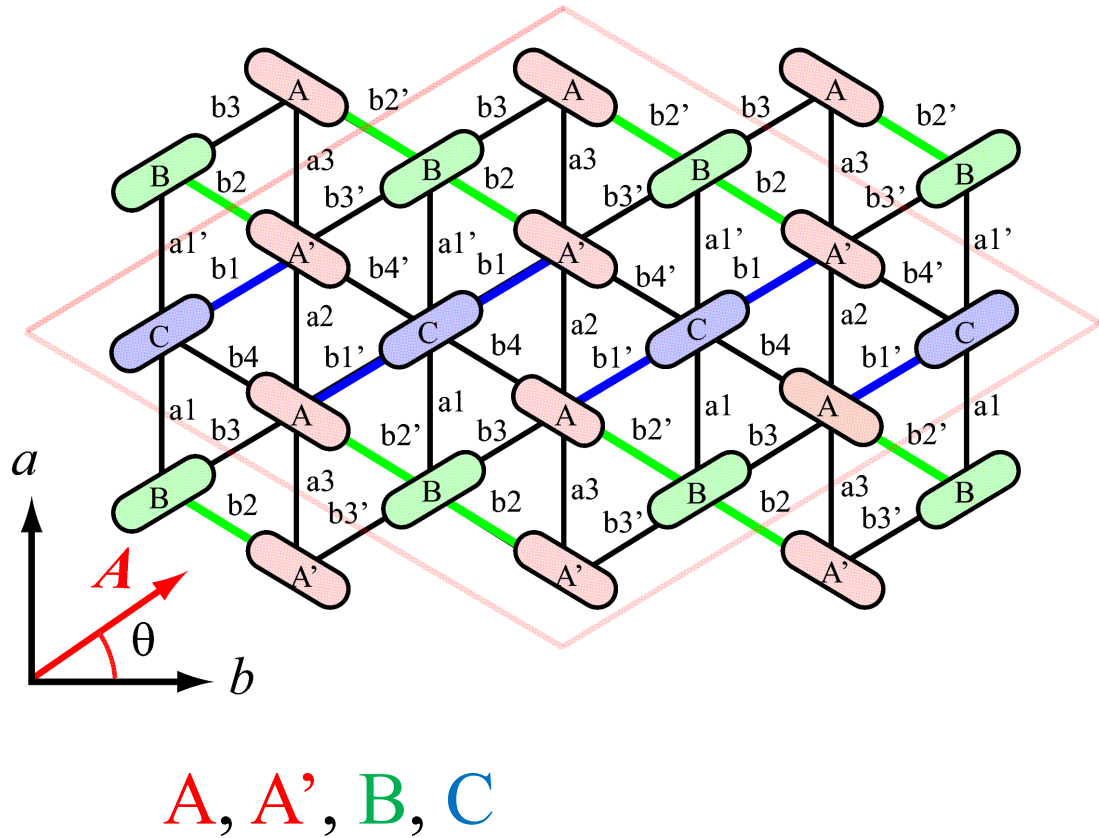


$\beta_{n,m}$ extended Huckel method

	<u>Kakiuchi</u> (Low)
a1	-0.0308
a1'	-0.0495
a2	-0.0544
a3	0.0329
b1	0.1212
b1'	0.1652
b2	0.1577
b2'	0.1773
b3	0.0673
b3'	0.0656
b4	0.0039
b4'	0.0323

T. Kakiuchi et al.,
J. Phys. Soc. Jpn. 76 113702 (2007)

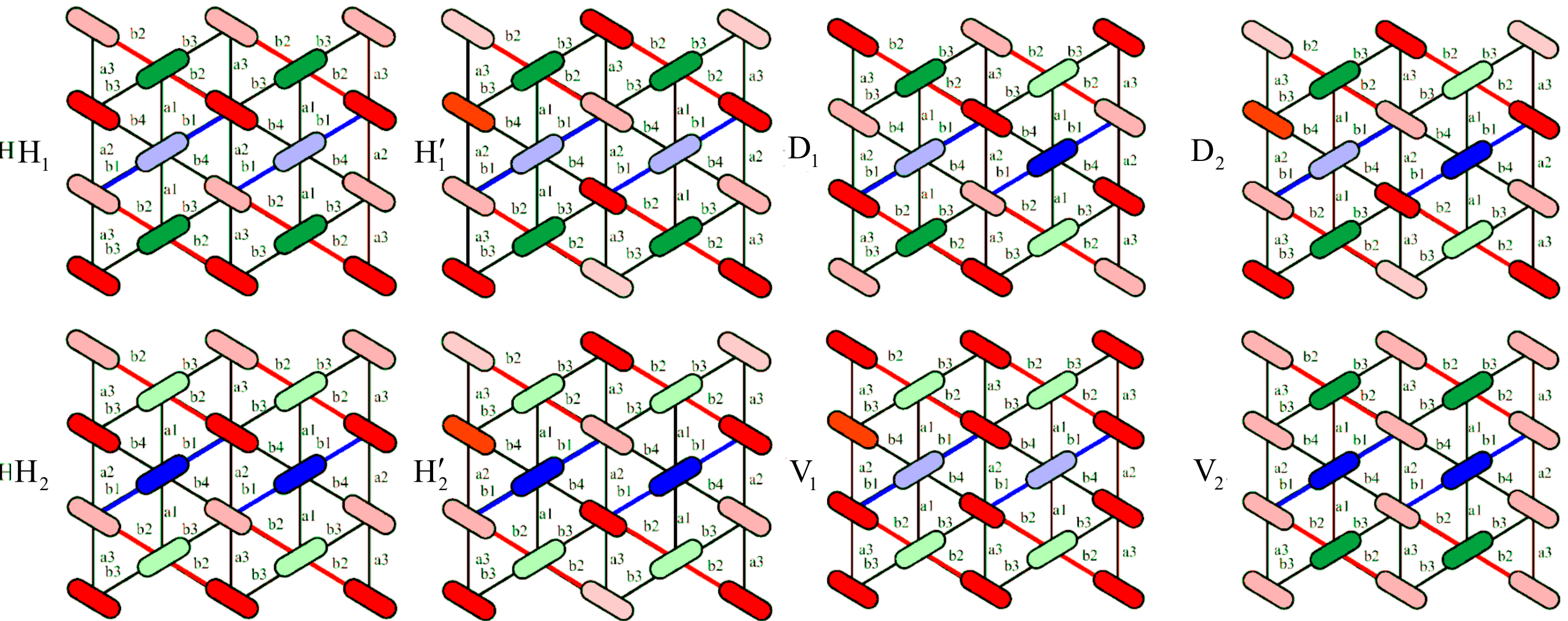
charge ordered phase



$$U = 0.9, V_p = 0.18, V_c = 0.22$$

diagonal vertical

	theory	experiment
$\bar{\rho}_A$	0.81	0.82
$\bar{\rho}_{A'}$	0.28	0.29
$\bar{\rho}_B$	0.70	0.68
$\bar{\rho}_C$	0.22	0.23



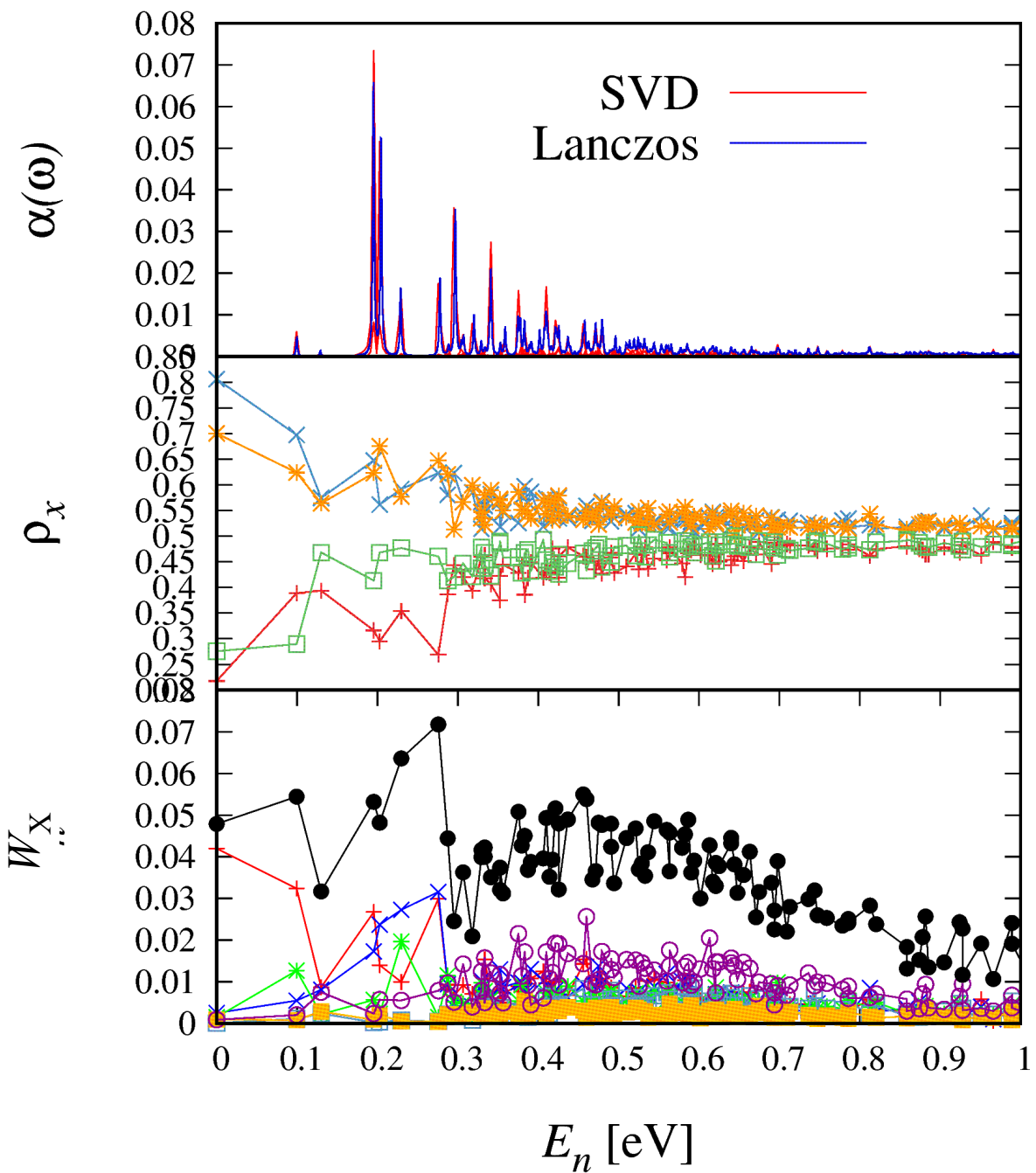
hole density $\rho_n = \langle \psi_k | \sum_{\sigma} c_{n,\sigma}^{\dagger} c_{n,\sigma} | \psi_k \rangle$

W_X : quantum weight for the state in which holes exist only at charge-rich sites of charge-ordered state X

$$X = H_1, H'_1, H_2, H'_2, V_1, V_2, D_1, D_2$$

$$W_{\text{tot}} = \sum_X W_X$$

random hole distribution	\rightarrow	$W_X \sim 10^{-5}$
		$W_{\text{tot}} \sim 10^{-4}$
charge ordered ground state	\rightarrow	$W_{\text{tot}} \sim 3 \times 10^{-2}$



low energy ($E_n \leq 0.3$) region

- W_{total} is larger than that of the ground state.
- charge disproportionation is large.
- density of states is small.



Collective motion associated with quantum fluctuations between different charge ordered states