Dynamics of photoinduced insulator-metal transition in one-dimensional Mott insulators

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Abstract. We theoretically investigate the dynamics of insulator-metal transition induced by light-pulse excitation in one-dimensional (1D) and two-dimensional (2D) Mott insulators. We adopt the Pariser-Parr-Pople model, and pump-probe signal is obtained by numerically calculating the time development of the state excited by a light pulse. In the 1D case, when photoexcitation density is below about 10%, an extremely small portion of the energy eigenstates dominates the optical process, and the spin-charge separation holds in these dominant energy eigenstates. As a result, the Mott gap and the short-range antiferromagnetic (AF) spin order are preserved, and the spin relaxation does not occur. When the density is above the value, a metallic state without the Mott gap is photogenerated, and the magnitude of the short-range AF spin order is significantly reduced by photoexcitation. This is consistent with the experimentally observed photoinduced Mott transition. Furthermore, the spin relaxation occurs in the metallic state. This photoinduced Mott transition is a manifestation of the spin-charge coupling in the intensely photoexcited states. In the 2D case, the spin and the charge degrees of freedom are coupled even in the weakly photoexcited states, and the characteristic crossover associated with the excitation density is not observed.

1. Introduction
Iwai et al. [1] and Okamoto et al. [2] investigated pump-probe reflection spectroscopy in one-dimensional (1D) Mott insulators: halogen-bridged Ni-chain compounds and ET-F$_2$TCNQ. When the photoexcitation density is below about 10%, the original absorption peak due to the Mott-gap transition and the Drude component coexist. This result can be understood by the concept of the spin-charge separation [2-4]. The Drude component is due to the photogenerated free charges just like the case of photoconductivity in conventional semiconductors. When the photoexcitation density is above the critical value, the Mott gap is destroyed and the electronic state is drastically changed by intense photoexcitation. However, the physical mechanism of this photoinduced transition is still unsolved. Furthermore, they found that the transition to the metallic state occurs within the time resolution of a few hundreds of femtoseconds, and the original Mott insulator recovers within a few picoseconds. However, the origins of these fast dynamics are also still unknown. It is also noted that only optical methods enable us to see the ultrafast dynamics of Mott transition induced by the carrier density control. The investigation of the dynamics will provide us with a new view point to clarify the origin of the Mott transition. Considering these points, we theoretically analyze the time evolution of the photogenerated excited state in 1D and two-dimensional (2D) Mott insulators. Some of the results in the present paper have been published in reference 5.
2. Method of calculation

We consider the 1D and 2D PPP Hamiltonian given by.

\[
H = -\sum_{n,i,\sigma} c_{n+1,i,\sigma}^\dagger c_{n,i,\sigma} e^{-i\mathcal{A}_i} + U \sum_n c_{n,\uparrow}^\dagger c_{n,\downarrow}^\dagger c_{n,\uparrow} c_{n,\downarrow} + \frac{V}{2} \sum_{n,m} |n-m|^{-1} e^{-\kappa |n-m|^{-1}} n_m n_m^\dagger,
\]

where \( c_{n,\sigma} \) is the annihilation operator for an electron of spin \( \sigma \) at site \( n \), \( l \) is the displacement vector to the nearest-neighbor sites, \( A(t) = \mathcal{A}(t) \mathcal{U} \mathcal{A}(t) \mathcal{U}^{-1} \), \( \mathcal{U} \) is the on-site Coulomb repulsion energy, \( n_m = \sum_{\sigma} c_{n,\sigma}^\dagger c_{n,\sigma} \), \( \kappa \) is the screening parameter, and \( V \) is the nearest-neighbor interaction energy. We use the unit of energy where the magnitude of the transfer integral between nearest-neighbor sites is 1.

We consider the two pulse excitation given by the vector potential

\[
A(t) = A_{\text{pump}} e_i \left[ \frac{1}{2} \left( \frac{t}{T_{\text{pump}}} \right)^2 \cos \omega_{\text{pump}} t \right] + A_{\text{probe}} e_i \left[ \frac{1}{2} \left( \frac{t - \tau_s}{T_{\text{probe}}} \right)^2 \cos \left[ \omega_{\text{pump}} (t - \tau_s) \right] \right],
\]

where \( \tau_s \) is the delay time, \( A_i \) is the maximum amplitude, \( e_i \) is the unit vector of the polarization direction, \( T_i \) is the duration time, \( \omega_i \) is the center frequency, and \( i= \text{pump or probe} \). The pump-probe signal \( I_{p-p} \) is proportional to the probability that the photoexcited state \( |\psi(t)\rangle \) by the pump pulse absorbs a probe photon irrespective of absorption time [5].

3. Results and discussion

We consider the periodic 1D chain with the system size \( N=14 \) and the periodic 2D square lattice with \( N=16 \) in the half-filled case. We have obtained the qualitatively the same results with \( N=12, \) and \( 16 \) in the 1D case, and with \( N=10 \) in the 2D case. We use the Coulomb parameters for the 1D Mott insulators where charge binding effect is significant [5]: \( U=10, V=2.5, \) and \( \kappa=0.5 \) in the 1D case. We use the Coulomb parameters \( U=10, V=0 \) in the 2D case. The pump and probe light pulses are assumed to be polarized in the chain direction in the 1D case, and in the direction of a side of the square in the 2D case. The pulse duration is set to \( T_{\text{pump}} = T_{\text{probe}} = 7 \). The pulse duration corresponds to 21 fs with using the parameter for halogen-bridged Ni-chain compound. Although this value is shorter than pulse duration in the experiments, it is adopted to see ultrafast response more clearly.

**Figure 1.** The pump-probe spectra \( I_{p-p} \) for various values of \( \tau_s \) at (a) \( A_{\text{pump}} = 0.05 \), and (b) \( A_{\text{pump}} = 1 \) in the 1D case.

**Figure 2.** The pump-probe spectra \( I_{p-p} \) for various values of \( \tau_s \) at (a) \( A_{\text{pump}} = 0.1 \), and (b) \( A_{\text{pump}} = 0.5 \) in the 2D case.
We show the transient pump-probe spectra for various values of delay time \( \tau \) in the 1D (2D) case in Fig. 1 (Fig. 2). We set \( \omega_{\text{pump}} \) to the peak frequency of the largest peak in the absorption spectrum. The attention should be paid to the fact that the pump-probe spectra for \( A_{\text{pump}} \leq 0.1 \) are essentially different from those for \( A_{\text{pump}} \geq 0.5 \) in the 1D case. On the contrary, the characteristic crossover of the spectrum associated with the excitation density is not observed in the 2D case.

### 3.1. Below threshold excitation density in 1D case

We first consider the case where photoexcitation density \( n_{\text{ph}}(\infty) \leq 10\% \) in the 1D case, where \( n_{\text{ph}}(\tau) = \Delta E(\tau)/(N\omega_{\text{pump}}) \) and \( \Delta E(\tau) \) is the increase of the electronic energy by the pump-light excitation. We mainly discuss the case of \( A_{\text{pump}} = 0.05 \) (\( n_{\text{ph}}(\infty) = 9\% \)) here, but the same conclusion can be obtained below the threshold excitation density. As seen from Fig. 1 (a), \( I_{\text{p-p}} \) spectrum mainly consists of a few peaks, and is clearly separated into higher \( (\omega_{\text{probe}}>6) \) and lower \( (\omega_{\text{probe}}<3.5) \) energy parts. The former part is dominant for any values of \( \tau \). The modified spin correlation function is defined by \( \tilde{\eta}(r, \tau) = \langle \psi(\tau) \rangle \sum S_r \cdot S_l \langle \hat{P}_1 \psi(\tau) \rangle \), where \( S_r \) is the spin operator at site \( r \), \( \hat{P}_1 \) is the projection operator onto the subspace where there are \( l \) doubly occupied or empty sites between the \( 0 \)-th and \( r + l \)-th sites, and it is the spin correlation function for the separated spin wave function in spin-charge separated case [5,6]. The function \( \tilde{\eta}(r, \tau) \) slightly changes with time, and therefore the short-range AF spin order of the ground state is not destroyed by pump-pulse excitation.

These characteristic features below the threshold excitation density come from the spin-charge separation. We have numerically calculated the absorption spectrum from the ground state \( |\psi_n\rangle \), and induced absorption spectrum from the energy eigenstate with \( n \) bound holon-doublon pairs \( |\psi_n\rangle \) (see below). We have found that the absorption and the induced absorption spectrum from \( |\psi_n\rangle \) with \( n = 0 \), consist of a few major peaks and each of these peaks arises from excitation to a single energy eigenstate, and the largest peak is due to excitation from \( |\psi_n\rangle \) to \( |\psi_{n+1}\rangle \). An extremely small portion of energy eigenstates dominates the transition moment from these energy eigenstates. Since the spin-charge separation holds for \( |\psi_n\rangle \) with \( n = 0 \), only the energy eigenstates with one (two) specific spin wave function(s) have transition moment from \( |\psi_n\rangle \) ( \( |\psi_n\rangle \) with \( n = 1, 2 \) ) [5]. The characteristic absorption and induced absorption spectrum originates from the spin-charge separation.

The total quantum weight \( W = \sum_{n=1}^{2} |c_n|^2 \approx 1 \), where \( c_n(\tau) = \langle \psi_n | \psi(\tau) \rangle \), and the spin-charge separated energy eigenstates \( |\psi_n\rangle \) with \( n = 0 \) dominate the quantum weight at any times. The energy eigenstates \( |\psi_n\rangle \) with \( n = 1, 2 \) have the short-range AF spin order, and the magnitudes of the order are slightly weaker than that of the ground state. Furthermore, these states have the Mott gap. As a result of these properties originating from the spin-charge separation, the short-range AF spin order is slightly weakened, and the Mott gap is not destroyed by the pump-pulse excitation even at the high photoexcitation density of 9%. The spin-charge separation strongly hinders the cooperative effects among the photogenerated charges.

### 3.2. Above threshold excitation density in 1D case

We consider the case where \( n_{\text{ph}}(\infty) > 10\% \) in the 1D case in this subsection. We mainly discuss the results at \( A_{\text{pump}} = 1 \), and the same conclusion can be obtained above the threshold excitation density.

We show \( \tilde{\eta}(r, \tau) \), \( n_{\text{ph}}(\tau) \) and \( W(\tau) \) in Fig. 3. As seen from this figure, \( n_{\text{ph}}(\tau) \leq 10\% \) for \( \tau \geq -10 \), the characteristic features below the threshold excitation density are seen. The total weight \( W(\tau) \) is nearly equal to 1, and the three spin-charge separated states dominate the quantum weight. As a result,
\( \tilde{\eta}(r, \tau) \) for \( |\psi(\tau)\rangle \) slightly changes with \( \tau \) as seen from Fig.3 (b), and the Mott gap remains in the corresponding region \( r_s \leq -10 \) as seen from Fig. 1 (b). Around \( \tau = -10 \), \( n_{ph}(\tau) \) exceeds the threshold value, and the characteristic features above the threshold excitation density appear after the time as will be mentioned below. For \(-10 \leq \tau \leq 0\), \( W(\tau) \) decreases to about 0.15, and the magnitude of \( \tilde{\eta}(r, \tau) \) for \( |\psi(\tau)\rangle \) significantly decreases with increasing \( \tau \). In the corresponding region \(-10 \leq r_s \leq 0\), \( I_{p-p} \) for \( \omega_{\text{probe}} \leq 6 \) significantly increases with increasing \( r_s \). Around \( r_s = 0 \), \( I_{p-p} \) distributes broadly below and above the Mott gap, and the Mott gap transition peak becomes much weaker. This shows that the Mott gap is destroyed by strong light excitation. In this way, the characteristic properties of the spin-charge separated states disappear in this time region.

We have also calculated the time dependence of the nearest-neighbor spin-spin interaction energy \( E_{ss}(\tau) \) for \( |\psi(\tau)\rangle \). The net component of \( E_{ss}(\tau) \) monotonously increases with \( \tau \), even after the pump light has gone off. This shows that the energy transfer from the charge degrees of freedom to the spin degrees of freedom occurs, and the spin relaxation occurs.

![Figure 3](image)

**Figure 3.**
The time dependence of (a) the modified spin correlation function \( \tilde{\eta} \), (b) the quantum weights \( |c_n|^2 \) and the total quantum weight \( W = \sum_{n=0}^{2} |c_n|^2 \), and (c) photoexcitation density \( n_{ph}(\tau) \) at \( A_{\text{pump}} = 1 \) in the 1D case.

### 3.3. 2D case

We consider the 2D case in this subsection. As seen from Fig. 2, the characteristic crossover associated with the excitation density of the 1D case is not observed even \( A \) is increased upto 0.5. Furthermore, the short-range AF spin order for the photoexcited state also becomes smaller gradually as the density is increased. We have numerically calculated the induced absorption spectrum from \( |\psi_1\rangle \), which has the largest transition moment from \( |\psi_0\rangle \). The induced absorption spectrum from \( |\psi_1\rangle \) have quite broad background and many peaks in side the Mott gap. Neither \( |\psi_0\rangle \) nor \( |\psi_1\rangle \) do not have the characteristic optical properties of the spin-charge separated states. As a result, \( |\psi(\tau)\rangle \) does not have them even in the weakly excited case.

### References

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