Disorder effects on relaxation of photoinduced metallic phase in one-dimensional Mott insulators

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Abstract. We theoretically investigate the effect of static disorder on the dynamics of photoexcited states in one-dimensional Mott insulators. We numerically calculate the time development of the state excited by a light pulse in the Pariser-Parr-Pople model, where disorder is introduced in the site energy and the transfer integral. In the weaker excitation case, the spin-charge coupling is induced by the disorder in the transfer integral. The effects of the disorder in the site energy on the spin-charge coupling are much weaker than those in the transfer integral. In the stronger excitation case, the spin and the charge degrees of freedom are coupled in the photoexcited states even in regular systems. The spin relaxations are enhanced by the disorders, and there are no significant differences in the effects of disorders on the spin relaxations between these two kinds of disorders.

1. Introduction
Ultrafast relaxation of photoexcited states have been observed in various one-dimensional (1D) Mott insulators [1-5], and it is considered that this is a characteristic commonly observed in the 1D Mott insulators. However, the origin of the ultrafast relaxation is not yet understood well. It is proposed that the relaxation is due to the spin degrees of freedom, which play a crucial role in their physical properties [1,2]. However, it has been shown that the spin relaxation does not occur in the weakly photoexcited states as a result of the separation between the spin and charge degrees of freedom as far as the regular Hubbard and Pariser-Parr-Pople (PPP) models are concerned[6-8]. We therefore investigate the effect of the disorder on the relaxation of photoexcited states in 1D Mott insulators.

2. Method of calculation
We consider the 1D PPP Hamiltonian including disorder in both the site energy and the electron transfer terms. It is given by

\[ H = \sum_n \alpha_n n_n + \sum_{n,\sigma} \beta_n \left( c_{n,\sigma}^\dagger c_{n+1,\sigma} + \text{h.c.} \right) + U \sum_n c_{n,\uparrow}^\dagger c_{n,\uparrow} c_{n,\downarrow}^\dagger c_{n,\downarrow} + \frac{V}{2} \sum_{n,m} |n-m|^{-1} e^{-\kappa |n-m|} n_n n_m, \]

where \( c_{n,\sigma} \) is the annihilation operator for an electron of spin \( \sigma \) at site \( n \), \( A(t) \) is the component of vector potential parallel to the 1D direction at time \( t \), \( U \) is the on-site Coulomb repulsion energy, \( n_n = \sum_\sigma c_{n,\sigma}^\dagger c_{n,\sigma} \), \( \kappa \) is a screening parameter, and \( V \) is the nearest-neighbor interaction energy. Using
random numbers $\tilde{\alpha}_n$ and $\tilde{\beta}_n$ that satisfy the conditions $\langle \tilde{\alpha}_n \rangle = 0$ and $\langle \tilde{\beta}_n \rangle = 0$, where $\langle \tilde{\alpha}_n \rangle = N^{-1} \sum_{n=1}^{N} \tilde{\alpha}_n \ (N$ is the system size), the site energy $\alpha_n$ and the transfer integral between the nearest-neighbor sites $\beta_n$ are given by $\alpha_n = \tilde{\alpha}_n$ and $\beta_n = -1 + \tilde{\beta}_n$, respectively. We adopt the units of energy with $\langle \beta_n \rangle = -1$. The averages $\langle \tilde{\alpha}_n \rangle$ and $\langle \tilde{\beta}_n \rangle$ are the measures of two kind of disorders.

We consider the optical pulse excitation given by the vector potential $A(t) = A \exp(-t^2/(2\omega_0))^2 \cos(\omega_0 t)$, where $A$ is the maximum amplitude, $T$ is the pulse duration, and $\omega_0$ is the center frequency. The photoexcited state $|\psi(t)\rangle$ by the excitation light pulse is calculated by numerically solving the time-dependent Schrodinger equation. We consider the time dependence of the nearest-neighbor spin-spin interaction energy $E_{ss}(t) = \sum_{n=1}^{N} J_n \langle \psi(t)|S_n \cdot S_{n+1}|\psi(t)\rangle$, the on-site Coulomb interaction energy $E_U(t) = U \langle \psi(t)|\sum_{n=1}^{N} c_{n+1}^\dagger c_n c_n^\dagger c_{n+1}|\psi(t)\rangle$ for the photoexcited state, where $J_n = 4J_n^2/U$ is the Heisenberg spin-spin coupling constant, and $S_n$ is the spin operator at site $n$.

3. Results and discussion

We consider the periodic 1D chain with the system size $N=14$, and the half-filled case is investigated in the present paper. To check the finite size effects, we have done some calculations with using the system sizes $N=12$ and $16$, and the qualitatively same results are obtained. We use the Coulomb parameters $U=10$, $V=2.5$, and $\kappa = 0.5$. These values are appropriate for the 1D Mott insulators where the charge binding effect is significant [8]. We have done some calculations also for the Hubbard model (in the case of $V=0$). The pulse duration is set to $T=10$. The largest peak in the optical absorption spectrum corresponds to the excitation to the single exciton like charge bound state [9-12], and it is the lowest energy one among the major peaks. Since we are not interested in the time resolution shorter than the pulse duration, we consider the averaged $\bar{E}_{ss}$ defined by

$$\bar{E}_{ss}(t) = \frac{1}{T}\int_{-T/2}^{T/2} E_{ss}(\tau) d\tau , \quad \text{and} \quad \bar{E}_U(t) \text{ is defined in the same way.}$$

3.1. Regular case

We show the results in the regular lattice case in this subsection. We have calculated the time dependence of $\bar{E}_{ss}(t)$ and $\bar{E}_U(t)$ for $A = 0.01$, 0.03, 0.1 and 0.5. For $A = 0.01$, both $\bar{E}_{ss}(t)$ and $\bar{E}_U(t)$ are almost time independent. The energy transfer to the spin degrees of freedom does not occur as a result of the spin-charge separation. For $0.03 \leq A \leq 0.1$, the net component of $\bar{E}_{ss}(t)$ slowly increases with $t$, and simple oscillations are observed in the dependence of $\bar{E}_{ss}(t)$, showing the weak spin-charge coupling in the photoexcited state. The excitation region $0.03 \leq A \leq 0.1$ is not in the linear response region, and a few multiphoton excited states have significant quantum weight in the photoexcited states [8]. The weak spin-charge coupling is due to the spin-charge separated nature of these excited states [8]. Furthermore, $\bar{E}_U(t)$ decreases and the decay rate is much larger than that of $\bar{E}_{ss}(t)$. The energy transfer within the charge degrees of freedom is much larger than that between spin and charge degrees of freedom. The energy transfer within the charge degrees of freedom is due to the Auger like process, and this will be discussed elsewhere. Actually, in the Hubbard model, where the Auger like process is strongly suppressed, both $\bar{E}_{ss}(t)$ and $\bar{E}_U(t)$ are almost time independent. On the other hand, for $0.5 \leq A$, the net component of $\bar{E}_{ss}(t)$ increases with $t$, and the increase rate is much larger than those for $A \leq 0.1$. Furthermore, the complicated oscillations are observed unlike the case of $A \leq 0.1$. These results come from the spin-charge coupling in the strongly photoexcited states.
3.2. Weaker excitation case
We show the results of the disorder effects in the weaker excitation case of $A \leq 0.1$ in this subsection. As seen from figure 1, when disorder exists only in the transfer integral, $\bar{E}_{ss}(t)$ oscillates with time, and the average of $\bar{E}_{ss}(t)$ increases with $t$. These results show that energy transfer between the charge and the spin degrees of freedom occurs, and the spin-charge coupling is induced by the disorder. On the contrary, in the case of site-energy disorder, the time dependence of $\bar{E}_{ss}(t)$ is changed slightly, and the characteristic oscillation in $\bar{E}_{ss}(t)$ is not observed. Therefore, the site-energy disorder has only minor effects on the spin-charge coupling. The difference between these two kinds of disorders can be attributed to the fact that the disorder in the $J_n$ is directly induced by the disorder in $\beta_n$.

![Figure 1](image1.png)  
**Figure 1.** The time dependence of spin-spin interaction energy at $A=0.01$ in the cases of $\langle \tilde{\alpha}_n \rangle = \langle \tilde{\beta}_n \rangle = 0$, $\langle \tilde{\alpha}_n \rangle = 0$ and $\langle \tilde{\beta}_n \rangle = 0.1$, and $\langle \tilde{\alpha}_n \rangle = 0.1$ and $\langle \tilde{\beta}_n \rangle = 0$, are shown by the thick solid, dotted and thin solid lines, respectively.

![Figure 2](image2.png)  
**Figure 2.** The time dependence of spin-spin interaction energy at $A=0.5$ in the cases of $\langle \tilde{\alpha}_n \rangle = \langle \tilde{\beta}_n \rangle = 0$, $\langle \tilde{\alpha}_n \rangle = 0$ and $\langle \tilde{\beta}_n \rangle = 0.1$, and $\langle \tilde{\alpha}_n \rangle = 0.1$ and $\langle \tilde{\beta}_n \rangle = 0$, are shown by the thick solid, dotted and thin solid lines, respectively.

3.3. Stronger excitation case
We show the results in the stronger excitation case of $A \geq 0.5$ in this subsection. As seen from figure 2, the increase rate of $\bar{E}_{ss}(t)$ for $t > 250$ becomes larger both for two kinds of disorders. In contrast to the weaker excitation case, there are no significant differences between the two kinds of disorders in the effect on the energy transfer from the charge to the spin degrees of freedom.

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