Transient spectra of photo-excited states in double exchange model

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Abstract. We investigate photo-induced effect in strongly correlated electron systems where conduction electrons couple with localized spins. In particular, the photo-induced phenomena in a charge-ordered insulator associated with antiferromagnetic order are examined. Transient excitation spectra are calculated by the exact diagonalization method in one-dimensional extended double exchange model. We find that in the photo-excited states, finite spectral weights in the one-particle excitation spectra appear inside the insulating gap and its band width increases with increasing time. This change of the electronic structure is correlated with the time development of the localized spin correlation. These results indicate that a strong coupling between spin and charge governs the photo-induced phenomena in this system.

1. Introduction

It is well known that in strongly correlated electron systems, electronic states near the phase boundary are changed dramatically by applying weak external perturbations. Strong correlation among the multi-degrees of freedom of electron, i.e. charge, spin and orbital, plays a key role on this characteristic phenomenon. One of the familiar correlated electron systems is the perovskite manganites which show the colossal magneto-resistance (CMR) effect, insulator-to-metal transition by hole doping and so on. Around the quarter-filling for the eg electrons of the manganese 3d orbitals, there is competition between a charge-ordered phase associated with the antiferromagnetic (AF) order and a ferromagnetic (FM) metallic phase, originating from the double exchange (DE) interaction [1]. Strong coupling between the itinerant electrons and the localized spins brings about correlation between transport and magnetic properties. Photo-irradiation collapses a balance of the stability of these two competing states. Recently, in the charge-ordered insulator (COI) associated with the AF order, changes in the electronic and magnetic properties by the photo-irradiation are reported experimentally. Both of the transient optical reflectivity and the magnet-optic Kerr rotation are measured in a few picoseconds after the photo-irradiation in Nd₀.₅Sr₀.₅MnO₃ [2]. These experimental results indicate the photo-induced FM metallic states caused by a strong coupling between the charge and spin degrees of freedom in the photo-excited states.

The purpose of this paper is to clarify the charge and spin dynamics in photo-excited states in a strongly correlated system described by the DE model. In particular, changes in the electronic structure by the photo-irradiation and its time evolution are focused on. We calculate the transient one-particle excitation spectra and the correlation function of the localized spins in the extended DE model.
These are calculated by the recursion and Lanczos methods in a finite size cluster. We find that finite spectral weights in the one-particle excitation spectra appear inside the charge-order gap, and increasing of the band width is correlated with change in the localized spin correlation.

2. Model and formulation

We introduce the one dimensional extended DE model to describe both the COI state associated with AF order and the FM metallic one. For simplicity, the \( t_{2g} \) electrons are treated as localized spins with magnitude of \( \frac{1}{2} \), and one of the degenerated \( e_{g} \) orbitals is considered. The Hamiltonian is given by

\[
\mathcal{H} = -t \sum_{\langle ij \rangle, \sigma} c_{i, \sigma}^\dagger c_{j, \sigma} + U \sum_i n_{i, \uparrow} n_{i, \downarrow} + V \sum_{\langle ij \rangle} n_{i} n_{j} - J_H \sum_i \mathbf{S}_i \cdot \mathbf{s}_i + J_S \sum_i \mathbf{S}_i \cdot \mathbf{S}_j.
\]

Here, \( c_{i, \sigma} \) is the annihilation operator for the conduction electron with spin \( \sigma \) at site \( i \). We define the number operator \( n_{i, \sigma} = \sum_{\sigma} c_{i, \sigma}^\dagger c_{i, \sigma} \), and the spin operator \( \mathbf{s}_i = \frac{1}{2} \sum_{\sigma, \sigma'} c_{i, \sigma}^\dagger \sigma_{\sigma, \sigma'} c_{i, \sigma} \) with the Pauli matrices \( \sigma_{\sigma, \sigma'} \), and the localized spin operator \( \mathbf{S}_i \) at site \( i \). We consider the electron hopping \( t \), the on-site Coulomb repulsion \( U \), and the nearest-neighbor one \( V \) for the conduction electrons. The AF superexchange interaction between the localized spins \( J_S \) and the Hund's coupling between the conduction electron and the localized spin \( J_H \) are also introduced.

Next we define the photo-excited state and the transient excitation spectra [3]. We assume that the vector potential of the pumping light at time \( \tau \) is given by a form \( A_{\text{pump}} \exp[-|\tau| \gamma_0 + i \omega_0 \tau] \) where \( \omega_0 \) is a center of the energy and \( \gamma_0 \) corresponds to a width of the photon energy distribution. At finite time \( \tau \geq 0 \), the photo-excited state \( |\tau\rangle \) and the transient one-particle excitation spectra are defined by

\[
|\tau\rangle = \frac{1}{N} \exp[-i \mathcal{H}_0 \tau] \text{Im} \left[ \frac{1}{\omega - \mathcal{H} + E_j + i \gamma_0} \right]^j |0\rangle,
\]

\[
A_p(\omega, \tau) = -\frac{1}{\pi} \sum_{\mathbf{q}, \sigma} \text{Im} \left[ \langle \mathbf{q}, \sigma | c_{\mathbf{q}, \sigma} \frac{1}{\omega - \mathcal{H} + E_1 + i \gamma} c_{\mathbf{q}, \sigma}^\dagger |\tau\rangle \right],
\]

\[
A_h(\omega, \tau) = -\frac{1}{\pi} \sum_{\mathbf{q}, \sigma} \text{Im} \left[ \langle \mathbf{q}, \sigma | c_{\mathbf{q}, \sigma} \frac{1}{\omega - \mathcal{H} + E_1 + i \gamma} c_{\mathbf{q}, \sigma}^\dagger |\tau\rangle \right],
\]

respectively, where \(|0\rangle\) describes the ground state with the energy \( E_{\text{g}, j} \) is the current operator, and \( N \) is a renormalization factor. We consider the probe photon with the energy \( \omega \) and the width of the energy \( \gamma \). We also define that \( E_j \) is the expectation value of Hamiltonian in the photo-excited state, and \( c_{\mathbf{q}, \sigma} \) is the Fourier transform of \( c_{i, \sigma} \). We adopt the Lanczos method to calculate the time evolution of the photo-excited states, and the recursion method for the excitation spectra.

The parameters value are chosen to be \( U=20t \), \( V=5t \), \( J_H=15t \), and \( J_S=0.3t \). We use an open boundary condition in the numerical calculation. The number of the lattice sites \( L \) and the electron number are chosen to be 9 and 5, respectively. The z-component of the total spin-quantum number is set to be zero. For the pumping photon, we take \( \omega_0=3.8t \) which corresponds to the lowest peak in the optical absorption spectrum, and \( \gamma_0=0.4t \).

3. Results and discussion

First, we mention the ground state properties. From the calculation of the charge correlation function \( N(q) \) and the spin correlation one for the localized spins \( S(q) \), we confirm that the ground state is the charge ordered state with the AF spin order. The optical absorption spectrum shows a gap, which indicates the system is an insulating state.

In Fig. 1 (a), we show the one-particle excitation spectra \( A_p(\omega) \) and \( A_h(\omega) \), before the pumping, and the transient ones \( A'_p(\omega, \tau) \) and \( A'_h(\omega, \tau) \) for \( \tau=0, 5/t, 10/t \) with \( \gamma=0.2t \). The spectra before pumping show a gap between -2t and 4t reflecting the insulating state. After the photo-irradiation, finite spectral weights appear inside the gap. The width of the in-gap state increase as time develops. We interpret that the in-gap state originates from the photo-carriers. Here we compare the obtained results with those in the spinless fermion \( V-t \) model which is the model consisting of the \( V \) and \( t \) terms.
in Eq. (1). In this model, a similar photo-carrier band appears by the photo-irradiation in the COI state. However, the band width of the in-gap state does not show remarkable time dependence. This difference between the two results is attributed to the existence of the localized spins in the DE model. To clarify this point furthermore, we calculate the correlation function of the localized spins defined by

\[ E(\tau) = \sum_i <\tau| S_i \cdot S_{i+1}| \tau>/(L-1) \]

and compare it with the band width of the in-gap state. The latter is estimated from the second order moment \( \Delta(\tau) \) with respect to the energy \( \omega \) for the transient one-particle excitation spectra \( A(\omega, \tau) = A'_p(\omega, \tau) + A'_h(\omega, \tau) \) between \( \omega = -2.7t \) and \( 5t \). The results are shown in Fig. 1 (b). With increasing time, \( E(\tau) \) increases and is saturated around \( \tau = 10/t \). A similar behavior is seen in \( \Delta(\tau) \). Thus, there is a strong correlation between \( E(\tau) \) and \( \Delta(\tau) \) in the DE model. In the photo-excited states, the DE interaction caused by the motion of photo-carriers aligns the localized spins ferromagnetically. As a result, the effective hopping of the photo-carriers increases, and the system becomes more metallic. In addition, we confirm by the numerical calculation that in the transient optical absorption spectrum, finite spectral weights appear inside the insulating gap and their intensities grow up with increasing time. These results indicate that in the photo-excited states, the charge and spin degrees of freedom are strongly coupled with each other.

![Figure 1](image_url)

**Figure 1:** (a) The transient one-particle excitation spectra at various times. The curves in the lowest panel are for the spectra before photo-irradiation. (b) The time dependence of the localized spin correlation function \( E(\tau) \) and that of the band width for the in-gap state estimated from the second order moment \( \Delta(\tau) \) of the one-particle excitation spectra between \( \omega = -2.7t \) and \( 5t \).

4. Conclusion

We have examined the photo-induced effect in correlated electron systems described by the DE model. We have calculated the time dependence of the one-particle excitation spectra in the one dimensional extended DE model after photo-irradiation in the COI state. By photo-excitation, the photo-carrier band appears inside the gap, and its band width increases with increasing time. This change of the one-particle excitation spectra is correlated with development of the FM correlation in the localized spins. These results indicate that in the photo-excited state, the charge and spin degrees of freedom are strongly correlated with each other.

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