Photo-induced phenomena in correlated electron system with multi-degree of freedom

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Abstract. We present a theoretical study of photo-induced dynamics in a correlated electron system where electronic charge couples with spin and lattice. This is motivated by the recent experiments in perovskite manganites and related compounds. The generalized double exchange model is analyzed by utilizing the two complementary methods, the exact diagonalization and inhomogeneous Hartree-Fock methods. Time evolutions of the optical absorption spectra, spin correlation, and charge correlation are calculated. It is shown that photo-induced dynamics in this system is governed by strong coupling between itinerant electrons and localized spins.

1. Introduction

Photo-induced phenomena in correlated electron system are one of the attractive themes in recent solid state physics. Here, strong Coulomb interactions between electrons and the multi-degrees of freedom, i.e. charge, spin, orbital and lattice, play key roles on the photo-induced phenomena in correlated electron systems. Because of these characteristics, several electronic phases appear and compete with each other. As results, by applying the weak external perturbations, a subtle balance of these phases collapse and a gigantic response is brought about.

One of the well known examples are manganites with the perovskite crystal structure, $A_{x-y}B_xMnO_3$ (A: rare-earth ion, B: alkaline earth ion). Electron configuration of a Mn ion at concentration $x$ is $(e_g)^3(t_{2g})^3$, where the $t_{2g}$ electrons are recognized as a localized spin because of its small electron transfer, and the $e_g$ electrons are as itinerant electrons. Around $x=0.5$, the charge ordered insulating state associated with the antiferromagnetic (AFM) long-range order appears. This phase strongly competes with the ferromagnetic (FM) metallic phase which is caused by the double-exchange interaction. By applying the magnetic field, an energy balance between the two phases is changed and the colossal magneto-resistance effect is brought about.

Recently, photo-induced phenomena in manganites around $x=0.5$ are examined experimentally [1-3]. After introduction of the pump photon into the charge-ordered insulating phase, dramatic change in the optical reflectivity is observed. At the same time, a finite value in the optical Kerr rotation appears. These results imply that the charge and magnetic structures are changed cooperatively by the photo-irradiation. Instead of these developments in the experimental investigations, theoretical understanding in the photo-induced phenomena in manganites and related compounds are not satisfactory.
In this paper, we present a theoretical study of the photo-induced dynamics in a strongly correlated electron system where electronic charges and spins are strongly coupled with each other. In particular, charge and spin dynamics in manganites are focused on. We analyze the extended double exchange model by utilizing the numerical exact diagonalization method and the inhomogeneous Hartree-Fock one.

2. Model and method

Let us set up the model Hamiltonian to describe the electronic structure and photo-induced phenomena in manganites. We start from the extended double-exchange model defined by

\[ H_{\text{ele}} = -t \sum_{\langle ij \rangle \sigma} c_{i \sigma}^\dagger c_{j \sigma} + U \sum_i n_i^\dagger n_i + V \sum_i n_i n_j - J_H \sum_i S_i \cdot s_i + J_s \sum_{\langle ij \rangle} S_i \cdot S_j, \]  

(1)

where we introduce the annihilation operator \( c_{i \sigma} \) with spin \( \sigma \) at site \( i \), and the spin operator \( S_i \) for the localized spin with magnitude of \( S \). We define the electron number operator \( n_i = \sum_\sigma c_{i \sigma}^\dagger c_{i \sigma} \), and the spin one \( s_i = (1/2) \sum_{\alpha \beta} c_{i \alpha} \sigma_{\alpha \beta}^\dagger c_{i \beta} \) with the Pauli matrices \( \sigma \) for the conduction electron. For simplicity, the orbital degree of freedom in Mn 3d orbital is not taken into account. We consider the electron transfer \( t \), the on-site Coulomb interaction \( U \), the inter-site one \( V \) between the nearest neighbour (NN) sites, the Hund coupling \( J_H \) between the itinerant and localized spins, and the antiferromagnetic (AFM) superexchange interaction \( J_s \) between the NN localized spins. It is noted that the double exchange interaction caused by the \( t \) and \( J_H \) terms in Eq. (1) favour the FM metallic state, and the \( V \) and \( J_s \) terms stabilize the AFM charge ordered phase.

We also consider the lattice degree of freedom and the electron-lattice interaction. Here, motions of the oxygen ions along the Mn-O bond direction are considered, and the interaction between the electronic charge and the breathing-type lattice distortion is introduced. An explicit form of this Hamilton is given by

\[ H_{\text{lat}} = \sum_{il} \left( \frac{1}{2m} p_{i+1/2}^2 + Ku_{i+1/2}^2 \right) + g \sum_{i \delta = \pm} n_i u_{i+1/2 \delta}, \]  

(2)

where \( u_{i+1/2} \) is the displacement of an oxygen ion at site \( i+1/2 \) in a perovskite crystal with the Mn-O direction \( l \), and \( p_{i+1/2} \) is the canonical momentum. We introduce the electron-lattice coupling \( g \), the ionic mass \( m \), and the spring constant \( K \).

We analyze the above Hamiltonians by using the two complementary calculation methods, the exact diagonalization method by utilizing the Lanczos methods in an one-dimensional lattice[4], and the inhomogeneous Hartree-Fock (HF) one[5] in the two-dimensional square lattice. In the exact diagonalization method, the electronic part of the Hamiltonian \( H_{\text{ele}} \) defined in Eq. (1) is only examined, and a magnitude of the localized spin is assumed to be 1/2, for simplicity. The wave function for the photo-excited state at time \( \tau \) after the photo-irradiation is defined by

\[ |\tau\rangle = N^{-1} e^{-iH_{\text{ele}} \tau} \text{Im} \left[ \frac{1}{\omega_0 - H_{\text{ele}} + E_0 + i\gamma_0} \right] j^\dagger |0\rangle, \]  

(3)

where \( j \) is the current operator coupled with the pump photon, and \( E_0 \) is the ground state energy. We consider the pump photon with energy \( \omega_0 \) and its width \( \gamma_0 \). To obtain this formula, we assume that one photon is absorbed in the electronic system, and \( \tau >> 1/\gamma_0 \). The transient optical absorption spectra based on the linear-response theory are defined by
\[ \alpha(\omega, \tau) = -\frac{1}{\pi} \text{Im} \left\{ \tau \left[ \frac{1}{\omega + E - H + i\gamma} j^\dagger \right] \right\}, \quad (4) \]

where the probe photon with energy \( \omega \) and width \( \gamma \) is considered. We introduce the current operator \( j \) induced by the probe photon, and the energy expectation value \( E \) at time \( \tau \).

In the HF method, we introduce the mean-field decoupling in the \( U \) and \( V \) terms in Eq. (1), and the localized spin and lattice are treated as classical objects. Photo-irradiation is simulated by the excitation of an electron from the highest occupied HF level to the unoccupied one. Time evolution in the electronic state is represented by

\[ \left| \psi^{(i)}(\tau + \Delta\tau) \right> = T \exp \left\{ \int_{\tau}^{\tau + \Delta\tau} \text{d}\tau' H^{HF}(\tau') \right\} \left| \psi^{(i)}(\tau) \right>, \quad (5) \]

where \( |\psi^{(i)}(\tau)\rangle \) is the \( i \)-th HF wave function at time \( \tau \), and \( H^{HF} \) is the HF Hamiltonian derived by Eqs. (1) and (2). The time evolution of the localized spins is described by the Bloch equation where the damping factor for the spin angular momentum is introduced. The ionic motions are represented by the Newtonian equations with the Hellmann-Feynman force. The three coupled equations are solved numerically.

3. Numerical Results

3.1 Results in exact diagonalization method

First we show the numerical results obtained by the exact diagonalization method in a one-dimensional cluster. In Fig. 1(a), the transient optical absorption spectra are presented. Numbers of the sites and electrons are 9 and 5, respectively, and the open boundary condition is adopted. The ground state before the photo irradiation is the charge ordered insulator with the AFM correlation between the localized spins. The optical absorption spectra without the photo-irradiation show the insulating gap being about \( 3t \). By introducing the pump photon, the peak structure appears around \( 1.5t \) and grows up with time evolution. It is shown by the detailed analyses that the lowest component of the new peak structure inside the gap corresponds to the Drude component in the limit of the infinite cluster size. In Fig. 1(b), the integrated spectral weight, \( D(\tau) \), inside of the insulating gap (filled circles) is plotted as a function of time. We also show the time dependence of the spin correlation function, defined by \( P(\tau) = \text{L}^{-1} \langle \Sigma_{ij} \cdot \Sigma_{ij} \rangle \rangle_{\text{g.s.}} \rangle \), in the same figure. The spectral weight increases with time and is saturated around \( \tau = 10/t \). Almost similar behaviour is seen in the localized spin correlation function. Thus, in the photo excited state, spin and charge dynamics are strongly correlated with each other. This increase of the spin correlation function implies that the energy flows from the conduction electron system to the localized spin one.
Figure 1: (a) Transient optical absorption spectra for several times. A dotted curve shows spectra before pumping. (b) The time dependence of the integrated spectral intensity between 0.5t and 4t (filled circles), and the correlation function of the localized spins (open circles). Parameter values are chosen to be \( U=20, J_{II}=15, V=5, J_s S^2=0.075, \omega_0=3.8, \) and \( \gamma_0=0.4 \) as a unit of \( t \).

3.2 Results in Hartree-Fock method

Next, we present the results obtained by the inhomogeneous HF method. The calculations are done in a two dimensional 10 \times 10 site cluster \( (L=100) \) with the periodic boundary condition. In the ground state before pumping, we confirm that the system is in the charge ordered phase with the ferrimagnetic spin order. In Fig. 2(a), we present the time dependence of the charge transfer gap and the staggered type charge correlation function defined by \( N(\pi,\pi)=L^{-1}\sum_{\langle ij\rangle}\langle n_i n_j\rangle \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i \rangle_i 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