Photoinduced dynamics in the one-dimensional two-orbital degenerate Hubbard model

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Abstract. We study photoinduced dynamical properties of the one-dimensional two-orbital degenerate Hubbard model. By using the Lanczos diagonalization method, we calculate optical conductivity spectra of the lowest photoexcited state. The optical conductivity has the Drude-like low-energy component caused by photogenerated carriers, a holon and a triplet doublon. We have also found an additional low-energy component in the spectra, which demonstrates a transition from the triplet doublon to singlet doublons.

1. Introduction

A Perovskite vanadium oxide LaVO\textsubscript{3} shows a series of phase transitions and spin/orbital orders [1]. At 143K, a magnetic phase transition occurs and the C-type antiferro(AF)-magnetic spin order appears, and a first-order structural phase transition occurs at 141K (=\(T_o\)). Below \(T_o\), the G-type AF orbital order with an alternating \(d_{yz}\)/\(d_{zx}\) electron configuration is theoretically suggested to appear. [2, 3]

A recent experiment has demonstrated that LaVO\textsubscript{3} may show a “photoinduced phase transition (PIPT)”, a macroscopic change of physical properties induced by photoirradiation [4]. The femtosecond time-resolved reflection spectroscopy exhibits a photoinduced enhancement of low-energy component immediately after the photoirradiation. This phenomenon is considered to be the evidence of generation of photoinduced carriers and the melting of the orbital and/or spin order in multi-orbital degenerate strongly correlated electron systems, which is not as well understood as the single-orbital systems [5].

In this study, we investigate physical properties of the photoexcited state of the one-dimensional (1D) two-orbital degenerate Hubbard model. Here we use the 1D model, taking account of the strong one-dimensional anisotropy of LaVO\textsubscript{3} [6]. We assume that the two highest orbitals, \(d_{yz}\) and \(d_{zx}\), are degenerate and the lowest orbital \(d_{xy}\) does not play a key role [3]. We calculate optical conductivity spectra of the lowest photoexcited state of the model by using the Lanczos diagonalization method. Our results show that the photoexcited state is metallic with a large Drude-like component. By contrast, the spectral weight above the optical gap is remarkably reduced. It is also found that an additional low-energy component appears, which shows a transition between the triplet doublon and singlet doublons.
2. Model and method

In this work, we use the 1D two-orbital degenerate Hubbard model. The Hamiltonian is given by

\[
\mathcal{H} = -t \sum_{l} \sum_{m=1,2} \sum_{\sigma=\uparrow,\downarrow} (c_{lm\sigma}^\dagger c_{l+1m\sigma} + H.c.) + U \sum_{lm} n_{lm\uparrow} n_{lm\downarrow} + U' \sum_{l} n_{l1} n_{l2} \\
- J \sum_{l} (n_{l1\uparrow} n_{l2\downarrow} + n_{l1\downarrow} n_{l2\uparrow}) + J' \sum_{l} (c_{l1\uparrow}^\dagger c_{l2\downarrow}^\dagger c_{l1\downarrow} c_{l2\uparrow} + H.c.),
\]

where \( c_{lm\sigma}^\dagger \) (\( c_{lm\sigma} \)) is the creation (annihilation) operator of an electron with spin \( \sigma \) at orbital \( m \) of site \( l \), \( n_{lm\sigma} = c_{lm\sigma}^\dagger c_{lm\sigma} \), and \( n_{lm} = n_{lm\uparrow} + n_{lm\downarrow} \). The transfer integral \( t \) is finite only between the same orbitals of neighboring sites. In the interaction terms, \( U, U', J, \) and \( J' \) denote intraorbital Coulomb, interorbital Coulomb, interorbital spin exchange, and interorbital pair hopping interactions, respectively. We also note that the following relations \( U = U' + 2J \) and \( J' = J \) hold [7]. For the real material, these Coulomb parameters satisfy \( U > U' > J \) [6, 8].

In order to clarify the photoinduced optical properties, we calculate the optical conductivity spectrum of the optically allowed, first excited state \( |\psi_{opt}\rangle \) with energy \( E_{opt} \) given by

\[
\sigma_{opt}(\omega) \equiv -\frac{1}{N\omega} \text{Im} \left[ \langle \psi_{opt} | \frac{1}{\omega + i\epsilon + E_{opt} - \mathcal{H}} \hat{j} | \psi_{opt} \rangle \right],
\]

where \( \hat{j} \) is the current operator defined as \( \hat{j} \equiv it \sum_{lm\sigma} (c_{l+1m\sigma}^\dagger c_{lm\sigma} - c_{lm\sigma}^\dagger c_{l+1m\sigma}) \) [9, 10], and the finite broadening factor \( \epsilon \) is set to \( t \) in our calculations. The lowest photoexcited state \( |\psi_{opt}\rangle \) is obtained by a preliminary calculation of the optical conductivity \( \sigma_{0}(\omega) \) of the ground state \( |\psi_0\rangle \) defined by

\[
\sigma_{0}(\omega) = -\frac{1}{N\omega} \text{Im} \left[ \langle \psi_0 | \frac{1}{\omega + i\epsilon + E_0 - \mathcal{H}} \hat{j} | \psi_0 \rangle \right].
\]

We treat the quarter-filled \( N \)-site chain with the system size \( N = 8 \) and impose the open boundary condition (OBC).

3. Results

Figure 1 shows the optical conductivity \( \sigma_{0}(\omega) \) of the ground state of this model with \( U'/t = 10 \). We note that, at \( U'/t = 10 \), the ground state for \( J < J_c \sim 4.6t \) is in the paramagnetic phase, and for \( J > J_c \) in the ferromagnetic phase [11], as shown in the inset of figure 1. In the paramagnetic phase, \( \sigma_{0}(\omega) \) has the lowest main peak and two higher-energy peaks with smaller spectral intensities. These three peaks are caused by the multiplet of the doubly occupied site. The photoexcited state of this model has a single empty site and a doubly occupied site. Following preceding studies of the single-band Hubbard model [12, 13], we here call the empty site “holon” and the doubly occupied site “doublon”. The Hund’s coupling splits the doublon into the lowest triplet and two higher-energy singlets, which are the origins of the three peaks in the paramagnetic phase. In the ferromagnetic phase, there is only a single peak caused by the triplet doublon. The schematic picture of the multiplet and the excitation gaps \( \Delta_{opt} \) is depicted in figure 2.

Figure 3 shows the optical conductivity \( \sigma_{opt}(\omega) \) of the lowest photoexcited state \( |\psi_{opt}\rangle \). We note that \( |\psi_{opt}\rangle \) corresponds to the lowest peak of \( \sigma_{0}(\omega) \) and thus involves the holon and the triplet doublon in both phases. The most pronounced character of \( \sigma_{opt}(\omega) \) is the low-energy component around \( \omega \sim t \). This is considered to be the Drude “precursor”, the analog of the
Figure 1. Optical conductivity spectra $\sigma_0(\omega)$ for $U'/t = 10$. The dotted lines show the excitation gaps of the multiplet (see figure 2). The inset shows the ground-state phase diagram of the model [11].

Drude weight of OBC [14]. Hence, the photoexcited state has a metallic character because of the photoinduced carriers. Higher energy components caused by the charge transfer excitation are depressed, which is similar to the photoinduced propriety of the single-band Hubbard model [10].

Here we focus on the low-energy component, and then calculate integrated spectral intensities defined by

$$I_{\text{opt}} \equiv \int_0^{\omega_l} d\omega \sigma_{\text{opt}}(\omega),$$

where the cutoff $\omega_l$ is introduced to reflect only the metallic low-energy component. For comparison, the low-energy components of doped systems are also evaluated in the same manner;

$$I_{1h(1d)} \equiv \int_0^{\omega_l} d\omega \sigma_{1h(1d)}(\omega),$$

where $\sigma_{1h(1d)}(\omega)$ is the optical conductivity of the ground state of the 1-hole (1-electron) doped system.

Figure 4 shows the $J$-dependence of the integrated intensities for the photoexcited state and the doped systems. The intensity $I_{\text{opt}}$ has non-monotonic $J$-dependence. For small $J$, $I_{\text{opt}}$ decreases with increasing $J$ and has a minimum around $J \sim 4t$. Then $I_{\text{opt}}$ increases. Comparison with the doped systems clarifies that this non-monotonic $J$-dependence is caused by the triplet doublon. The spectral intensity $I_{1d}$ has the similar $J$-dependence while $I_{1h}$ is nearly constant for small $J$.

This non-monotonic dependence of $I_{\text{opt}}$ and $I_{1d}$ stems from the transition from the triplet doublon to the singlet doublon. The charge transfer by the optical excitation can generate the singlet doublon from the triplet doublon [15]. For small $J$, where the splitting between the triplet and the singlet is small, the related spectral component appears below the optical gap. As $J$ increases, this weight merges into that of the charge-transfer excitation and disappears.

4. Summary and discussion

In this study, we have investigated dynamical properties of the photoexcited state of the 1D two-orbital degenerate Hubbard model. The lowest photoexcited state $|\psi_{\text{opt}}\rangle$ consists of a
photogenerated holon and a triplet doublon. The optical conductivity \( \sigma_{\text{opt}}(\omega) \) has the metallic low-energy component caused by these photogenerated carriers, while the spectral intensity of the charge-transfer excitation is largely depressed. These results explain the photoinduced change of the reflectance spectrum of LaVO\(_3\), and are similar to those of the photoexcited state of the single-orbital Hubbard model.

The difference between the single-orbital model and the two-orbital model is in the low-energy spectrum associated with the transition from the triplet doublon to the singlet doublon in the two-orbital model. For small \( J \), we can observe this component below the optical gap because the splitting between them is small.

For LaVO\(_3\), this low-energy component would be hard to observe experimentally. This is because the LaVO\(_3\) has the ferromagnetic spin configuration along the c-axis. Thus the excitation by a pulse laser polarized along the c-axis generates only the triplet doublon. Although the conductivity spectrum polarized perpendicular to the c-axis may detect the low-energy component, the signal would be small because of the strong one-dimensional anisotropy of LaVO\(_3\) [4, 6]. For YVO\(_3\), by contrast, this low-energy component can be observed more easily. This material also shows the anisotropy [6], and the spin configuration along the c-axis is antiferromagnetic (the G-type AF magnetic phase). Hence the created triplet doublon can change into the singlet doublon.

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