Multipole correlations of $t_{2g}$-orbital Hubbard model with spin-orbit coupling

Hiroaki Onishi*

Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, JAPAN

We investigate the ground-state properties of a one-dimensional $t_{2g}$-orbital Hubbard model including an atomic spin-orbit coupling by using numerical methods, such as Lanczos diagonalization and density-matrix renormalization group. As the spin-orbit coupling increases, we find a ground-state transition from a paramagnetic state to a ferromagnetic state. In the ferromagnetic state, since the spin-orbit coupling mixes spin and orbital states with complex number coefficients, an anti-ferro-orbital state with complex orbitals appears. According to the appearance of the complex orbital state, we observe an enhancement of $\Gamma_{2u}$ octupole correlations.

KEYWORDS: $t_{2g}$ orbitals, spin-orbit coupling, multipole, density-matrix renormalization group

The competition and cooperation between spin and orbital degrees of freedom in strongly correlated electron systems manifest itself in the emergence of various types of spin-orbital ordered and quantum liquid phases.\textsuperscript{1–3} In general, among competing interactions involving spin and orbital, the spin-orbit coupling is supposed to be weak in 3$d$ transition-metal oxides such as cuprates and manganites, while as we move to 4$d$ and 5$d$ electrons, the spin-orbit coupling becomes strong and responsible for magnetic, transport, and optical properties. When the spin-orbit coupling is dominant, spin and orbital are not independent, but instead the total angular momentum gives a good description of the many-body state. In fact, it has been suggested that Sr$_2$IrO$_4$, in which Ir$^{4+}$ ions have five electrons in triply degenerate $t_{2g}$ orbitals, exhibits a novel Mott-insulating state with an effective total angular momentum $J_{\text{eff}}$=1/2 due to a strong spin-orbit coupling.\textsuperscript{4–7}

In the limit of strong spin-orbit coupling, the ground-state Kramers doublet at a local ion can be described by an isospin $J$, and the truncation error is estimated to be $10^{-5}$. We attempt to clarify multipole properties of $t_{2g}$-orbital Hubbard model including the spin-orbit coupling by numerical methods. With increasing the spin-orbit coupling, the ground state changes from a paramagnetic state to a ferromagnetic state in terms of the magnitude of the total spin. In the ferromagnetic phase, antiferro-dipole correlations develop even when the spin state is ferromagnetic due to the orbital contribution. On the other hand, the spin-orbit coupling induces a complex orbital state, in which real $xy$, $yz$, and $zx$ orbitals are mixed with complex number coefficients. According to the complex orbital state, the $\Gamma_{2u}$ octupole correlations are enhanced.

Let us consider triply degenerate $t_{2g}$ orbitals on a one-dimensional chain along the $x$ direction with five electrons per site. The one-dimensional $t_{2g}$-orbital Hubbard model with the spin-orbit coupling is described by

$$
H = \sum_{i,\tau,\sigma} t_{i\tau\sigma}(d_{i\tau\sigma}^\dagger d_{i+1\tau\sigma} + \text{h.c.}) + \lambda \sum_{i} \mathbf{L}_i \cdot \mathbf{S}_i + U \sum_{i,\tau} \rho_{i\tau\uparrow} \rho_{i\tau\downarrow} + \left(\frac{U'}{2}\right) \sum_{i,\tau,\tau' \neq \tau} \rho_{i\tau\sigma} \rho_{i\tau'\sigma} + \left(\frac{J}{2}\right) \sum_{i,\tau,\sigma,\tau' \neq \tau} d_{i\tau\sigma}^\dagger d_{i\tau'\sigma} d_{i\tau'\sigma}^\dagger d_{i\tau\sigma} + \left(\frac{J'}{2}\right) \sum_{i,\tau,\sigma,\tau' \neq \tau} d_{i\tau\sigma}^\dagger d_{i\tau'\sigma} d_{i\tau'\sigma}^\dagger d_{i\tau\sigma},
$$

where $d_{i\tau\sigma}$ ($d_{i\tau\sigma}^\dagger$) is an annihilation (creation) operator for an electron with spin $\sigma (\uparrow, \downarrow)$ in orbital $\tau (xy, yz, zx)$ at site $i$, and $\rho_{i\tau\sigma} = d_{i\tau\sigma}^\dagger d_{i\tau\sigma}$. The hopping amplitude is given by $t_{xy,xy}=t_{zx,zx}=t$ and zero for other combinations of orbitals. Hereafter, $t$ is taken as the energy unit. $\mathbf{L}_i$ and $\mathbf{S}_i$ represent orbital and spin angular momentum operators, respectively, and $\lambda$ is the spin-orbit coupling. $U$, $U'$, $J$, and $J'$ denote intra-orbital Coulomb, inter-orbital Coulomb, exchange, and pair-hopping interactions, respectively. We assume $U=U'+J+J'$ due to the rotation symmetry in the local orbital space and $J'=J$ due to the reality of the orbital function.\textsuperscript{12} Throughout this paper, we set $\hbar=\kappa_B=1$.

We investigate the ground-state properties of the model (1) by exploiting a finite-system density-matrix renormalization group (DMRG) method with open boundary conditions.\textsuperscript{13} The number of states kept for each block is up to $m=120$, and the truncation error is estimated to be $10^{-4}$~$10^{-5}$. We remark that due to the three orbitals in one site, the number of bases for the single site is 64, and the size of the superblock

*E-mail address: onishi.hiroaki@jaea.go.jp
Hilbert space grows as $m^2 \times 64^2$. To reduce the size of the Hilbert space, we usually decompose the Hilbert space into a block-diagonal form by using symmetries of the Hamiltonian. In the present case, however, the spin-orbit coupling breaks the spin SU(2) symmetry, so that we cannot utilize $S_{\text{tot}}^z$ as a good quantum number, where $S_{\text{tot}}^z$ is the $z$ component of the total spin. Since we ignore $e_g$ orbitals among $d$ orbitals, the total angular momentum is not a conserved quantity. The total number of electrons can be used as a good quantum number.

Thus, since DMRG calculations consume much CPU times, we supplementally use a Lanczos diagonalization method for the analysis of a four-site periodic chain to accumulate results with relatively short CPU times.

Let us first look at Lanczos results for the four-site periodic chain. In Fig. 1(a), we show the phase diagram in the $(J, \lambda)$ plane for $U' = 10$. The phase boundary is determined by the magnitude of the total spin $S_{\text{tot}}^2$. As shown in Fig. 1(b), $S_{\text{tot}}^2$ is almost zero for small $\lambda$, indicating a spin-singlet ground state. As $\lambda$ increases, we find a transition to a ferromagnetic state with finite $S_{\text{tot}}^2$. Note that even in the limit of large $\lambda$, $S_{\text{tot}}^2$ does not approach the maximum value $2(2+1)=6$, since the spin-orbit coupling mixes spin up and down states and the complete ferromagnetic state is disturbed. In Fig. 1(c), we plot the correlation between spin and orbital in the local site $L_{\text{loc}} \cdot S_{\text{loc}}$. At $\lambda = 0$, there is no correlation between spin and orbital. As $\lambda$ increases, the spin-orbital correlation develops and approaches one in the limit of large $\lambda$, indicating totally parallel spin and orbital angular momenta. In Fig. 1(d), the magnitude of the total angular momentum in the single site $J_{\text{loc}}^2$ is shown. At the transition point, $J_{\text{loc}}^2$ exhibits a sudden increase, since the spin-orbit coupling stabilizes a large total angular momentum state at every local site. Note again that $J_{\text{loc}}^2$ does not reach the maximum value $2(2+1) = 6$ in the limit of large $\lambda$, since the total angular momentum is not a conserved quantity. Regarding the orbital state, we show the charge density in each orbital in Fig. 1(e). Due to the spatial anisotropy of orbitals, one hole is preferably accommodated in itinerant $xy$ or $zx$ orbitals in each site, while localized $yz$ orbitals are doubly occupied. Measuring charge correlations, we find that holes occupy real $xy$ or $zx$ orbital alternately for small $\lambda$ (not shown). Namely, the ground state is a real orbital state. For large $\lambda$, however, $xy$, $yz$, and $zx$ orbitals are mixed with complex number coefficients by the spin-orbit coupling, leading to a complex orbital state.

Now we move on to the analysis of multipole properties to clarify the ground-state properties from the viewpoint of multipole. We measure multipole correlation functions

$$\chi_{\Gamma, \gamma}(q) = \sum_{j,k} (X_{j\Gamma, \gamma} X_{k\Gamma, \gamma}) e^{iq(j-k)}/N,$$  

where $X_{i\Gamma, \gamma}$ is a multipole operator with the symbol $X$ of multipole for the irreducible representation $\Gamma$ in the cubic symmetry at site $i$. Here, we consider 15 types of multipoles including three dipoles ($X=J$), five quadrupoles ($X=Q$), and seven octupoles ($X=T$), as listed in Table I. By evaluating the multipole correlation functions by DMRG calculations with chains of 16 sites.

Figure 2 shows DMRG results of the multipole correlation functions at $U' = 10$, $J = 2$, and $\lambda = 0$ for the paramagnetic phase. Regarding dipoles, as shown in Fig. 2(a), the $J_x$ correlation has a peak at $q = \pi$, which signals an antiferromagnetic state. Here, we notice that each of the dipole correlations exhibits a peak at $q = \pi/2$, while the kink corresponds to a peak for the $J_y$ and $J_z$ correlations. This kink structure originates in the spin-orbital SU(4) symmetry which realizes at a special point $J = \lambda = 0.15-19$. At the SU(4) symmetric point, correlations of spin $S_i$ and orbital pseudospin $T_i := \frac{1}{2} \sum_{\sigma} t_{i\sigma} \sigma_{\tau\tau'} d_{i\tau\sigma}^\dagger \sigma_{\tau'\sigma}$, where $\sigma$ are Pauli matrices, coincide with each other and have a peak at $q = \pi/2$. With increasing $J$, the spin correlation of $q = \pi/2$ grows and the peak of the spin correlation remains at $q = \pi$.
The spin state is ferromagnetic. Moreover, the complex orbital state appears, since the spin-orbit coupling yields the linear combinations of spin and orbital states with complex number coefficients. Accordingly, we observe an enhancement of the \( \Gamma_{4u} \) octupole correlations. It is an interesting issue to explore possible multipole ordering in 5d-electron Ir compounds with strong spin-orbit coupling.

The author thanks G. Khalilullin, S. Maekawa, and M. Mori for useful discussions. This work was supported by Grant-in-Aid for Scientific Research of Ministry of Education, Culture, Sports, Science, and Technology of Japan.

1) Proc. Int. Conf. Strongly Correlated Electrons with Orbital Degrees of Freedom (ORBITAL2001), J. Phys. Soc. Jpn. 71 (2002) Suppl.
2) Y. Tokura and N. Nagaosa: Science 288 (2000) 462.
3) T. Hotta: Rep. Prog. Phys. 69 (2006) 2061.
4) B. J. Kim, H. Jin, S. J. Moon, J.-Y. Kim, B.-G. Park, C. S. Leem, J. Yu, T. W. Noh, C. Kim, S.-J. Oh, J.-H. Park, V. Durairaj, G. Cao, and E. Rotenberg: Phys. Rev. Lett. 101 (2008) 076402.
5) B. J. Kim, H. Ohsumi, T. Komesu, S. Sakai, T. Morita, H. Takagi, and T. Arima: Science 323 (2009) 1329.
6) S. Chikara, O. Korneta, W. P. Crummett, L. E. DeLong, P. Schlottmann, and G. Cao: Phys. Rev. B 80 (2009) 140407.
7) S. J. Moon, H. Jin, W. S. Choi, J. S. Lee, S. S. A. Seo, J. Yu, G. Cao, T. W. Noh, and Y. S. Lee: Phys. Rev. B 80 (2009) 195110.
8) H. Jin, H. Jeong, T. Ozaki, and J. Yu: Phys. Rev. B 80 (2009) 075112.
9) G. Jackeli and G. Khalilullin: Phys. Rev. Lett. 102 (2009) 017205.
10) Y. Kuramoto, H. Kusunose, and A. Kiss: J. Phys. Soc. Jpn. 78 (2009) 072001.
11) H. Onishi and T. Hotta: J. Phys. Soc. Jpn. 75 Suppl. (2006) 266.
12) D. Dagotto, T. Hotta, and A. Moreo: Phys. Rep. 344 (2001) 1.
13) S. White: Phys. Rev. Lett. 93 (1992) 2863.
14) R. Shiina, H. Shiba, and P. Thalmeier: J. Phys. Soc. Jpn. 66 (1997) 1741.
15) Y. Yamashita, N. Shibata, and K. Ueda: Phys. Rev. B 58 (1998) 9114.
16) H. C. Lee, P. Azaria, and E. Boulat: Phys. Rev. B 69 (2004) 155109.
17) J. C. Xavier, H. Onishi, T. Hotta, and D. Dagotto: Phys. Rev. B 73 (2006) 014405.
18) H. Onishi and T. Hotta: J. Magn. Magn. Mater. 310 (2007) 790.
19) H. Onishi: Phys. Rev. B 76 (2007) 014441.