Variational Monte Carlo study of ferromagnetism in the two-orbital Hubbard model on a square lattice

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To understand the effects of orbital degeneracy on magnetism, in particular the effects of Hund's rule coupling, we study the two-orbital Hubbard model on a square lattice by a variational Monte Carlo method. As a variational wave function, we consider a Gutzwiller-projected wave function for a staggered spin- and/or orbital-ordered state. We find a ferromagnetic phase with staggered orbital order around quarter filling, i.e., electron number $n=1$ per site, and an antiferromagnetic phase without orbital order around half filling $n=2$. In addition, we find that another ferromagnetic phase without orbital order is realized in a wide filling region for large Hund's rule coupling. These two ferromagnetic states are metallic except for quarter filling. We show that orbital degeneracy and strong correlation effects stabilize the ferromagnetic states.

The mechanism of itinerant ferromagnetism is a long-standing problem in physics of condensed matter. As a simple model for itinerant ferromagnetism, the single-orbital Hubbard model has been studied intensively, but it has been revealed that it is difficult to stabilize a ferromagnetic state in the Hubbard model with only nearest-neighbor hopping on simple lattices such as a square lattice.

One possible improvement to the Hubbard model for ferromagnetism is a modification of the band structure. Since the early stage of the study of ferromagnetism, a large density of states around Fermi level, as in an fcc lattice with appropriate filling, has been suggested to stabilize a ferromagnetic state.[1–3] Indeed, realization of ferromagnetic ground states is proven for some flatband systems[4,5] and nearly flatband systems.[6] It is also shown that ferromagnetism occurs for an fcc-like infinite-dimensional lattice and for an fcc lattice by using a dynamical mean-field theory,[7] while a dynamical mean-field theory for a hypercubic lattice does not show ferromagnetism.[8] For finite dimensions, it has been shown that ferromagnetism can occur by including next-nearest hopping, which induces Van Hove singularity, for a chain[9] and for a square lattice.[10]

Another possible improvement is the inclusion of orbital degree of freedom, which may be important in dealing with realistic situations in transition metals. For orbitally degenerate systems, it has been suggested that intra-atomic Hund's rule coupling can stabilize ferromagnetism.[11–13] The simplest extended model of the single-orbital Hubbard model including orbital degree of freedom is the two-orbital Hubbard model. This model shows ferromagnetism with antiferro-orbital order at quarter filling, i.e., electron number $n=1$ per site, in the strong Coulomb interaction limit.[14]

This ferromagnetic state at $n=1$ is insulating. Thus, it is an interesting problem as to what extent is the ferromagnetic state stable against doping of electron or hole which makes the system metallic. It is found that the ferromagnetic state is stable to some extent against doping in one dimension[15] and in infinite dimensions.[16,17] For other finite dimensions, there are few studies on doping effects on magnetism of the two-orbital Hubbard model. Sakai *et al.*[18] studied the two-orbital

Hubbard model on an fcc lattice by a dynamical mean-field theory and stressed the importance of the lattice structure and Hund's rule coupling for ferromagnetism.

To understand the magnetism of the two-orbital Hubbard model deeply, we have to investigate the model for many parameter sets since the two-orbital Hubbard model has a parameter for Hund's rule coupling in addition to that for the Coulomb interaction. However, such an extensive study is difficult for a two-orbital model beyond the Hartree-Fock approximation since there are as many as 16 electron configurations at each site, and thus it is hard for numerical calculations.

In this Rapid Communication, to overcome such difficulty, we apply a variational Monte Carlo method[19] to the two-orbital Hubbard model on a square lattice. We use a Gutzwiller-projected wave function as a variational wave function. This wave function is simple enough but includes correlation effects, and we can evaluate energy for several states and for various parameters. In particular, we can construct a phase diagram by varying the value of Hund's rule coupling and filling n. Thus, we can investigate the overall feature of the two-orbital Hubbard model. At $n=1$, a similar model without considering orbital order[20] and the two-orbital Hubbard model considering the possibility of orbital order[21] have been studied by the variational Monte Carlo method, but the effect of doping has not been investigated by these studies, which is a main topic of the present Rapid Communication.

The two-orbital Hubbard model is given by

$$H = \sum_{\mathbf{k},\tau,\sigma} \epsilon_{\mathbf{k}} c^{\dagger}_{\mathbf{k}\tau\sigma} c_{\mathbf{k}\tau\sigma} + U \sum_{i,\tau} n_{i\tau\uparrow} n_{i\tau\downarrow} + U' \sum_{i} n_{i1} n_{i2}$$
$$+ J \sum_{i,\sigma,\sigma'} c^{\dagger}_{i1\sigma} c^{\dagger}_{i2\sigma'} c_{i1\sigma'} c_{i2\sigma} + J' \sum_{i,\tau \neq \tau'} c^{\dagger}_{i\tau\uparrow} c^{\dagger}_{i\tau\downarrow} c_{i\tau'\downarrow} c_{i\tau'\uparrow},$$

$$(1)$$

where $c_{i\tau\sigma}$ is the annihilation operator of the electron at site i with orbital τ (=1 or 2) and spin σ (=$\uparrow$ or $\downarrow$), $c_{\mathbf{k}\tau\sigma}$ is the Fourier transform of it, $n_{i\tau\sigma}=c^{\dagger}_{i\tau\sigma} c_{i\tau\sigma}$, and $n_{i\tau}=\Sigma_{\sigma} n_{i\tau\sigma}$. The

coupling constants U, U', J, and J' denote the intraorbital Coulomb, interorbital Coulomb, exchange, and pair-hopping interactions, respectively. We use the relation $U=U'+J+J'$, which is satisfied in several orbital-degenerate models such as a model for p orbitals, a model for e_g orbitals, and a model for t_{2g} orbitals.[22] We also use the relation $J=J'$, which holds if we can choose the wave functions of orbitals to be real.[22] We consider only a nearest-neighbor hopping integral t for both orbitals, and the kinetic energy is given by $\epsilon_{\mathbf{k}} =2t(\cos k_x+\cos k_y)$. Here we have set the lattice constant to unity.

We consider the variational wave function given by

$$|\Psi\rangle = P_G|\Phi\rangle = \prod_{i\gamma}[1-(1-g_\gamma)|i\gamma\rangle\langle i\gamma|]|\Phi\rangle, \quad (2)$$

where P_G is the Gutzwiller projection operator for onsite density correlation.[23–25] $|i\gamma\rangle\langle i\gamma|$ denotes the projection onto the state γ at site i and g_γ is the variational parameter controlling the probability of state γ. There are 16 states at each site in the present two-orbital model. The Hartree-Fock-type wave function $|\Phi\rangle$, which describes charge, spin, orbital, and spin-orbital coupled ordered states, is given by

$$|\Phi\rangle = \prod_{\mathbf{k}a\tau\sigma} b^{(a)\dagger}_{\mathbf{k}\tau\sigma}|0\rangle, \quad (3)$$

where $|0\rangle$ is the vacuum. The quasiparticles occupy N_σ states for each spin σ from the lowest quasiparticle energy state, where N_σ is the number of electrons with spin σ. The energy of the quasiparticle in the ordered state is given by

$$\lambda^{(a)}_{\mathbf{k}\tau\sigma} = a\sqrt{\Delta^2_{\tau\sigma}+\epsilon^2_{\mathbf{k}}}. \quad (4)$$

The creation operators of quasiparticles are given by

$$b^{(-)\dagger}_{\mathbf{k}\tau\sigma} = u_{\mathbf{k}\tau\sigma}c^{\dagger}_{\mathbf{k}\tau\sigma} + \text{sgn}(\Delta_{\tau\sigma})v_{\mathbf{k}\tau\sigma}c^{\dagger}_{\mathbf{k}+\mathbf{Q}\tau\sigma}, \quad (5)$$

$$b^{(+)\dagger}_{\mathbf{k}\tau\sigma} = -\text{sgn}(\Delta_{\tau\sigma})v_{\mathbf{k}\tau\sigma}c^{\dagger}_{\mathbf{k}\tau\sigma} + u_{\mathbf{k}\tau\sigma}c^{\dagger}_{\mathbf{k}+\mathbf{Q}\tau\sigma}, \quad (6)$$

where $\mathbf{Q}=(\pi,\pi)$ is the ordering vector considered in this study and

$$u_{\mathbf{k}\tau\sigma} = [(1-\epsilon_{\mathbf{k}}/\sqrt{\Delta^2_{\tau\sigma}+\epsilon^2_{\mathbf{k}}})/2]^{1/2}, \quad (7)$$

$$v_{\mathbf{k}\tau\sigma} = [(1+\epsilon_{\mathbf{k}}/\sqrt{\Delta^2_{\tau\sigma}+\epsilon^2_{\mathbf{k}}})/2]^{1/2}. \quad (8)$$

The quasiparticle gap in the ordered state is given by

$$\Delta_{\tau\sigma} = \Delta_c + \Delta_s(\delta_{\sigma\uparrow}-\delta_{\sigma\downarrow}) + \Delta_o(\delta_{\tau1}-\delta_{\tau2})$$
$$+ \Delta_{so}(\delta_{\sigma\uparrow}-\delta_{\sigma\downarrow})(\delta_{\tau1}-\delta_{\tau2}), \quad (9)$$

where Δ_c, Δ_s, Δ_o, and Δ_{so} denote the gaps for charge, spin, orbital, and spin-orbital ordered states, respectively, and we take them as variational parameters.

Here, we have chosen the z component of spin for the ordered state. We can choose x or y component, but they are equivalent due to the rotational symmetry in spin space. On the other hand, there is rotational symmetry only in the z-x plane in the orbital space. For orbital order, in addition to the z component as in Eq. (9), we have also investigated the possibility of y-component order. The model Hamiltonian (1) can be rewritten in terms of basis states of the y component
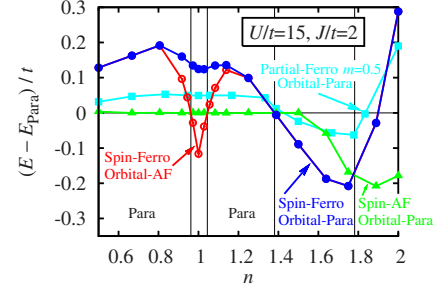

FIG. 1. (Color online) Filling dependence of energy for several states measured from that of the paramagnetic state: spin-antiferro (AF) orbital-para ($m=0$, $\Delta_s\neq0$, $\Delta_o=0$, and solid triangles), partial-ferro $m=0.5$ orbital-para ($\Delta_s=0$, $\Delta_o=0$, and solid squares), spin-ferro orbital-para ($m=1$, $\Delta_s=0$, $\Delta_o=0$, and solid circles), and spin-ferro orbital-AF ($m=1$, $\Delta_s=0$, $\Delta_o\neq0$, and open circles).

of orbital by replacing interaction parameters with tilde: $\tilde{U} =(U+U'+J-J')/2$, $\tilde{U}'=(U+U'-J+J')/2$, $\tilde{J}=(U-U'+J +J')/2$, and $\tilde{J}'=(-U+U'+J+J')/2$. Thus we can study the y-component orbital order with the same form for the variational function by simply changing interaction parameters.

We evaluate the expectation value of energy for the variational wave function by using the Monte Carlo method, and we optimize these gap parameters and the Gutzwiller parameters to minimize energy. For the optimization we use a fixed sampling method.[26,27] The number of parameters in the Gutzwiller projection can be reduced from 16 to 10 by considering equivalence of the two orbitals.[25] We can further reduce the number of the Gutzwiller parameters to seven when we consider the conservation of spin, i.e., when we fix N_σ. Thus the total number of the variational parameters is 11. Note that, in this study, we take partial-ferromagnetic states into consideration and spin $\uparrow$ and $\downarrow$ are not equivalent for these states. Thus we cannot reduce the number of the parameters further by considering spin states. We can also evaluate energy by fixing some parameters, for example, we set all the gap parameters to zero for a paramagnetic state. The calculations have been done for a 12×12 lattice with periodic-antiperiodic boundary conditions.

Figure 1 shows the energy E in ordered states per site measured from the energy E_{para} per site in the paramagnetic state as functions of n for $U/t=15$ and $J/t=2$ as an example. Statistical errors are much smaller than the symbol sizes. In the low filling region, the paramagnetic state is stable. Around quarter filling, $n=1$, the ferromagnetic phase with antiferro-orbital order appears. In the filling region, $1.38 \lesssim n \lesssim 1.78$, the ferromagnetic phase without orbital order appears. Around half filling, $n=2$, antiferromagnetic phase appears due to nesting of the Fermi surface. While we have calculated energy of partial-ferromagnetic states, for example, for $m=(N_\uparrow-N_\downarrow)/(N_\uparrow+N_\downarrow)=0.5$ shown in Fig. 1, these states do not become the ground state.

Here we comment on the ground states at $n=1$ and 2. The chemical potential μ can be obtained from $\mu=dE/dn$ in the ground state, and we also obtain the relation
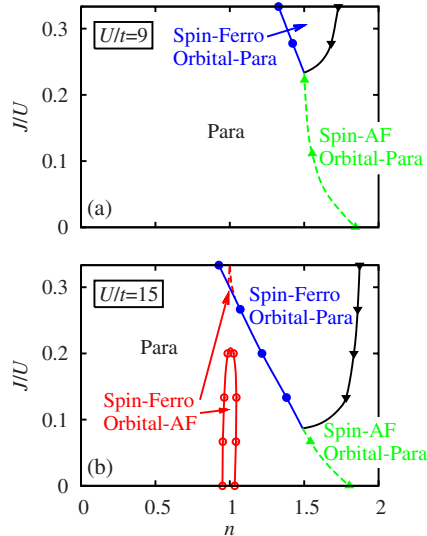
FIG. 2. (Color online) Phase diagrams for (a) $U/t=9$ and for (b) $U/t=15$. Solid lines denote first-order transitions and dashed lines denote second-order transitions.

$$\frac{dn}{d\mu}\frac{d^2E}{dn^2}=1. \qquad (10)$$

Thus, in an insulating state, i.e., $dn/d\mu=0$, the second derivative of energy E with respect to n should diverge and vice versa. At $n=1$, the ground-state energy has a cusp as shown in Fig. 1 and the ground state is insulating. Note that we can obtain energy for $n>2$ from the present data by using electron-hole symmetry of the model, and we find a cusp in the ground-state energy also at $n=2$.

Figure 2 shows the phase diagrams for $U/t=9$ and 15. First, we explain the phase diagram [Fig. 2(b)] for $U/t=15$. The ferromagnetic phase with antiferro-orbital order appears around quarter filling as is expected from the effective Hamiltonian for the strong-coupling limit. However, it is found that this ferromagnetic phase is easily destabilized by doping. To stabilize this ferromagnetic phase in a wider filling region, a much larger value of Coulomb interaction is necessary.

This ferromagnetic phase is also destabilized by increasing Hund's rule coupling J. Hund's rule coupling is usually expected to stabilize magnetically ordered states, but the effective interaction $U_{\text{eff}}=U'-J$ between different orbitals is reduced by Hund's rule coupling. Thus, Hund's rule coupling destabilizes the orbital order, and as a result, the ferromagnetic state supported by the orbital order is also destabilized. Note that in the ferromagnetic phase, there is rotational symmetry in the orbital space, and orbital order for the x, y, and z components are equivalent.

At higher filling region, another ferromagnetic phase without orbital order appears in a large Hund's rule coupling region. The ferromagnetic phase extends in a wide parameter region for $n\gtrsim1$, while it does not for $n\lesssim1$. This finding is in agreement with the statement that double-exchange-like mechanism works well for $n\gtrsim1$ since the probability of double occupancy is high, but it is less effective for $n\lesssim1$.[17]

In this ferromagnetic phase at $n\simeq1$ and $J/U\simeq0.3$, we expect an orbital-antiferro order, since in the ferromagnetic state the model is reduced to the single-orbital Hubbard model with effective interaction U_{eff} if we regard spin in the single-orbital Hubbard model as orbital, and around $n=1$ an orbital-antiferro state should occur. However, it is difficult to distinguish a small energy difference between orbital-para and orbital-antiferro states around there due to the small value of U_{eff}. Thus, the phase boundary between spin-ferro orbital-para and spin-ferro orbital-antiferro in Fig. 2(b) is merely a guide to the eyes.

Around half filling, the antiferromagnetic phase appears as expected from the nesting of Fermi surface. The phase transition from the paramagnetic phase to the antiferromagnetic phase is of the second order. We have checked that the energy difference between these phases is proportional to $(n-n_c)^2$ for $n\gtrsim n_c$, where n_c is the critical filling. Note that the spin-ferro orbital-antiferro state at $n=1$ and the spin-antiferro orbital-para state at $n=2$ are insulating, and other ground states are metallic.

By reducing the Coulomb interaction U, the regions of the ordered phases become narrower as shown in Fig. 2(a) for $U/t=9$. In particular the spin-ferro orbital-antiferro state disappeared. At $n=1$, the ferromagnetic state with orbital order disappears at $U/t\lesssim10$.[21] The other ferromagnetic phase without orbital order is also easily destabilized by reducing the Coulomb interaction. This fact indicates that realization of ferromagnetism is a strong correlation effect. On the other hand, the antiferromagnetic phase around half filling, which is stabilized by the nesting of the Fermi surface and can be obtained with a weak-coupling theory, is realized in a wide region even at $U/t=9$.

Note that we have also calculated the energy of the single-orbital Hubbard model within the Gutzwiller wave function, and we have found that a much larger value of Coulomb interaction $U/t\gtrsim23$ is necessary to stabilize a ferromagnetic phase. Thus, the orbital degeneracy and Hund's rule coupling are important ingredients for the realization of ferromagnetism with a moderate value of the Coulomb interaction.

To summarize, we have studied the two-orbital Hubbard model on a square lattice by a variational Monte Carlo method. We have considered charge, spin, orbital, and spin-orbital coupled ordered states for the variational wave function. Then, we have constructed phase diagrams for the ground states. We find a narrow region of the ferromagnetic state with orbital-antiferro order around quarter filling and a wide region of ferromagnetic phase without orbital order at large Hund's rule coupling for $U/t=15$. The ferromagnetic phase with orbital order is easily destabilized by doping and by reducing the Coulomb interaction. The ferromagnetic phase without orbital order is also destabilized strongly by reducing the Coulomb interaction. Thus, realization of ferromagnetic states is a strong correlation effect. Investigation of the effects of realistic anisotropic hopping integral depending on orbital and further improvement of the variational wave function are important future problems.

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