Stability of the Nagaoka-type ferromagnetic state in a t_{2g} orbital system on a cubic lattice

Eric Bobrow and Yi Li
Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218, USA

(Received 10 January 2018; revised manuscript received 9 March 2018; published 16 April 2018)

We generalize the previous exact results of the Nagaoka-type itinerant ferromagnetic states in a three-dimensional t_{2g} orbital system to allow for multiple holes. The system is a simple cubic lattice with each site possessing d_{xy}, d_{yz}, and d_{xz} orbitals, which allow two-dimensional hopping within each orbital plane. In the strong-coupling limit of $U \to \infty$, the orbital-generalized Nagaoka ferromagnetic states are proved to be degenerate with the ground state in the thermodynamic limit when the hole number per orbital layer scales slower than $L^{\frac{1}{2}}$. This result is valid for arbitrary values of the ferromagnetic Hund's coupling $J > 0$ and interorbital repulsion $V \geqslant 0$. The stability of the Nagaoka-type state at finite electron densities with respect to a single spin flip is investigated. These results provide helpful guidance for studying the mechanism of itinerant ferromagnetism for the t_{2g} orbital materials.

DOI: 10.1103/PhysRevB.97.155132

I. INTRODUCTION

Itinerant ferromagnetism, i.e., ferromagnetism of metallic states with Fermi surfaces, remains a challenging problem of condensed-matter physics. Since the Coulomb interaction is spin independent, it cannot directly give rise to electron spin polarizations at the classic level, and the mechanism of itinerant ferromagnetism is hence fundamentally quantum mechanical. As first described through Stoner's criterion [1], itinerant ferromagnetism arises from the direct exchange interaction among electrons with the same spin. However, this criterion overlooks correlation effects among electrons with opposite spins. In fact, even in the presence of very strong interactions, electrons can remain unpolarized and build up highly correlated wave functions to reduce repulsive interactions. Due to the intrinsic strong-correlation nature of the problem, it is usually difficult to obtain a precise description based on perturbative approaches, and thus, nonperturbative results have played important roles in the study of itinerant ferromagnetism [2–14]. The previously known exact results of ferromagnetism are largely classified into two categories: Nagaoka ferromagnetism [3,15] for single-band Hubbard systems and flat-band ferromagnetism [7,8].

A key result in the study of itinerant ferromagnetism is Nagaoka's theorem for the Hubbard model, which proves the ground state is fully spin polarized for exactly one hole away from half filling in the $U \to \infty$ limit [3]. The underlying physics is that the fully polarized state maximally facilitates the hole's coherent hopping to reduce the kinetic energy. Tasaki [15] simplified the proof of Nagaoka's theorem by using the Perron-Frobenius theorem [2]. However, this method typically breaks down for fermionic systems with multiple holes in two or higher dimensions because of the fermion sign originating from the antisymmetry under exchange. For the case of a single-band Hubbard model in the $U \to \infty$ limit, the Nagaoka-type ferromagnetic state with multiple holes was proven to be degenerate with the ground state under the following conditions [16,17]: The hole number $N_h \sim L^{\alpha}$ for $0 \leqslant \alpha < 1$ on an $L \times L$ square lattice in the limit of $L \to \infty$. This stability was investigated using a squeeze theorem argument in which a variational upper bound to the ground-state energy was shown to be equal to a lower bound for suitably low N_h in the thermodynamic limit. The variational trial state used was the fully spin polarized Nagaoka-type state. The lower bound was given by the Gershgorin circle theorem from linear algebra, which is explained in the Appendix.

In contrast to single-band Hubbard models, most itinerant ferromagnetic metals are orbital active. In such systems, the multiorbital structure together with Hund's interaction plays an important role in the onset of itinerant ferromagnetism despite Hund's interaction being local and typically polarizing only spins on the same site. Recently, exact results of itinerant ferromagnetism in strongly correlated multiorbital systems were proven, providing a sufficient condition for ferromagnetism driven by Hund's coupling [18]. Different from the Nagaoka theorem, an entire phase of itinerant ferromagnetism is set up with a wide range of electron filling. Furthermore, since such systems are free of the fermion sign problem of quantum Monte Carlo simulations [18], the Curie-Weiss metal phase and critical scalings of the ferromagnetic phase transitions have been simulated by quantum Monte Carlo to high numerical precision [19]. However, the proof of the ground-state ferromagnetism is based on the Perron-Frobenius theorem, which requires one-dimensional bands to avoid issues related to fermionic exchange. In order to generalize itinerant ferromagnetism to orbital-active systems with a quasi-two-dimensional band structure, a t_{2g} orbital system was previously studied [20] with the number of holes restricted to exactly one away from half filling in each orbital plane. In this case, a multiorbital generalization of the Nagaoka-like ferromagnetic state was proven to be the unique ground state up to spin degeneracy. Nevertheless, its stability has not been previously studied.

In this paper, we extend the multiorbital Nagaoka-type ferromagnetic states proven in Ref. [20] to the multihole case and investigate its stability. The analyses are done in the presence of two- and three-dimensional band structures in the strong-correlation regime. By generalizing the method used

in Refs. [16,17] to include Hund's coupling and interorbital repulsion, we show that in the strong-interaction limit, the generalized Nagaoka ferromagnetic state with the t_{2g} orbitals remains degenerate with the ground state in the multihole case in the thermodynamic limit. Although the hole density remains zero, the hole number can go to infinity, scaling as a finite power of the system size. An analysis of the stability of the generalized Nagaoka state against flipping a single spin is also performed, which shows that the region of instability shrinks in the presence of Hund's coupling.

The remainder of this paper is organized as follows: In Sec. II, we introduce the multiorbital Hubbard model for a three-dimensional (3D) t_{2g} orbital system with a two-dimensional (2D) band in a cubic lattice. In Sec. III, we analyze the stability of the ferromagnetic ground state in the presence of multiple holes for the multiorbital model presented in Sec. II. Then, by analyzing the change in energy due to a single flipped spin [21], a region in which the fully polarized state is no longer a ground state is identified. The Nagaoka-type ferromagnetic state and its stability in the presence of a 3D band structure is studied in Sec. IV. Conclusions and discussion are presented in Sec. V.

II. THE t_{2g} ORBITAL SYSTEM WITH MULTIORBITAL INTERACTIONS

In this section, we present the band structure of the 3D t_{2g} orbital system and the on-site multiorbital Hubbard interactions.

The system to be studied is a 3D multiorbital Hubbard model on an $L \times L \times L$ simple cubic lattice with d_{xy}, d_{yz}, and d_{xz} orbitals at each site with on-site multiorbital interactions. The Hamiltonian can be written as

$$H = H^K + H^U + H^V + H^J, \tag{1}$$

where H^K, H^U, H^V, and H^J represent the kinetic energy, the intraorbital Hubbard interaction, the interorbital Hubbard interaction, and the interorbital Hund's coupling, respectively. Since the t_{2g} orbitals are planar, the hopping term for each orbital is anisotropic in general. For the system with quasi-2D band structure studied here and in Sec. III, transverse hopping perpendicular to the orbital plane is much weaker than intraplane hopping and hence will be neglected. A fully 3D band structure including transverse perpendicular hopping terms will be considered in Sec. IV. With intraplane hopping neglected, the kinetic term corresponding to the d_{xy} orbital takes the form

$$H^K_{xy} = t \sum_{\mathbf{r},\sigma} [d^\dagger_{xy,\sigma}(\mathbf{r}) d_{xy,\sigma}(\mathbf{r}+\hat{x}) + d^\dagger_{xy,\sigma}(\mathbf{r}) d_{xy,\sigma}(\mathbf{r}+\hat{y}) + \text{H.c.}], \tag{2}$$

where the lattice constant is taken to be 1 and t is the hopping integral. The hopping terms for the other orbital planes have the same form with the directional indices replaced as necessary, and the full kinetic term H^K is a sum of hopping terms for the d_{xy}, d_{yz}, and d_{xz} orbital planes.

For a simple cubic lattice with negligible transverse hopping, Eq. (2) and its yz and xz counterparts constitute all nearest-neighbor hoppings allowed by symmetry. Different orbitals do not mix at this level due to the cubic symmetry of the system, which can be seen as follows. Without loss of generality, consider an x bond between sites $\mathbf{r}$ and $\mathbf{r}+\hat{x}$. Since this bond is invariant under reflections with respect to both the xy plane and the xz plane, hopping along this bond should respect these symmetries. The d_{xy} orbital is even, while d_{xz} and d_{yz} orbitals are odd under the former reflection. Thus, d_{xy} does not mix with either d_{xz} or d_{yz} through this hopping. Furthermore, d_{xz} is even, while d_{yz} is odd under the latter reflection, and thus, they do not mix either.

The on-site multiorbital Hubbard interactions consist of intraorbital and interorbital terms. The intraorbital interaction H^U is expressed as

$$H^U = U \sum_{\mathbf{r},a} n_{a,\uparrow}(\mathbf{r}) n_{a,\downarrow}(\mathbf{r}), \tag{3}$$

where a is the orbital index and $n_{a,\sigma}(\mathbf{r}) = d^\dagger_{a,\sigma}(\mathbf{r}) d_{a,\sigma}(\mathbf{r})$. The interorbital interaction takes the form

$$H^V = V \sum_{\mathbf{r},a>b} [1 - n_a(\mathbf{r})][1 - n_b(\mathbf{r})], \tag{4}$$

where $n_a(\mathbf{r}) = n_{a,\uparrow}(\mathbf{r}) + n_{a,\downarrow}(\mathbf{r})$. H^V is expressed in terms of the hole number occupation, which is equivalent to the corresponding electron number form up to an overall constant. Since we will explore the stability of the Nagaoka state in which nearly every orbital on every site is filled, the hole representation will be more convenient.

The final interaction in the model, the on-site interorbital Hund's coupling, reads

$$H^J = -J \sum_{\mathbf{r},a>b} \left[\mathbf{S}_a(\mathbf{r}) \cdot \mathbf{S}_b(\mathbf{r}) - \frac{1}{4} n_a(\mathbf{r}) n_b(\mathbf{r}) \right]. \tag{5}$$

For any two orbitals a and b on site $\mathbf{r}$, the energy from the Hund's coupling is non-negative if $J > 0$. The energy contribution is J if both orbitals are filled and form a spin singlet and zero otherwise.

Below, we consider the limit of $U \to \infty$, in which no individual orbital can hold two electrons. Instead, individual sites can hold up to three electrons, all in different orbitals, with their interaction determined by H^V and H^J. We also consider only the case of $V \geqslant 0$, i.e., with repulsive interorbit interaction, and $J > 0$, i.e., ferromagnetic Hund's coupling.

III. STABILITY OF THE GENERALIZED NAGAOKA-LIKE STATE

In this section, we investigate the stability of the generalized Nagaoka state in the 3D t_{2g} orbital systems with quasi-2D band structure. Such a state with a single hole per orbital layer was proven previously in Ref. [20].

Since the hopping term H^K allows only holes to hop within orbital planes, the number of holes in each orbital plane is conserved. For simplicity, assume that each orbital plane has the same number n_h of holes. Since there are L layers for each of the three orbital plane directions, the total number of holes is then $N_h = 3Ln_h$. Note that in what follows, the number of holes will always refer to the number of holes above the half-filled background or the number of electrons below half filling.

As in Ref. [20], conservation of the hole number in each orbital plane allows the overall Hilbert space to be written as a tensor product of Hilbert spaces for each layer in each orbital plane direction. For the lth layer of orbital type a, we define a reference state $|R_{a,l,\uparrow}\rangle$ in which each orbital is filled with a spin-↑ electron. $|R_{a,l,\uparrow}\rangle$ is equivalent to the state with all single-particle momentum states $\mathbf{k}$ in the 2D Brillouin zone fully filled. Now we add n_h holes by removing n_h electrons one by one from the highest filled single-particle state. The resulting many-body state, a Slater determinant state with all electron spins up, is expressed as

$$|h_{a,l,\uparrow}\rangle = \prod_{i=1}^{n_h} d_{a,l}(\mathbf{k}_i)|R_{l,a,\uparrow}\rangle, \tag{6}$$

where $d_{a,l}(\mathbf{k}_i) = \frac{1}{L}\sum_{\mathbf{r}} d_{a,l}(\mathbf{r})e^{i\mathbf{k}_i\mathbf{r}}$ and $\mathbf{k}_i$ and $\mathbf{r}$ represent 2D momentum and lattice vectors, respectively. The momenta take values $\mathbf{k}_i = (\frac{2m_1\pi}{L}, \frac{2m_2\pi}{L})$ with $m_{1,2}$ integers. We consider the limit of $n_h/L^2 \to 0$, where the single-particle spectrum becomes parabolic. From the sign convention of t in Eq. (2), m_1 and m_2 start from (0,0) and take values in ascending order of $m_1^2 + m_2^2$.

A. Estimation of the upper bound

In order to show the existence of a fully spin-polarized ground state, consider a trial state

$$|\psi_t\rangle = \bigotimes_{l=1}^{L} |h_{xy,l,\uparrow}\rangle \otimes |h_{yz,l,\uparrow}\rangle \otimes |h_{xz,l,\uparrow}\rangle, \tag{7}$$

where the constraint of n_h holes per layer is enforced by the form of the basis states. This is a fully spin polarized state with the maximum spin $S = S_z = \frac{N_s}{2}$ for N_s spins. Since the Hamiltonian possesses SU(2) symmetry, applying $S_{T,-} = S_{T,x} - iS_{T,y}$ successively on $|\psi_t\rangle$, where $\vec{S}_T$ is the total spin operator, produces a $2N_s + 1$ SU(2) multiplet.

An upper bound on the ground-state energy E_g is derived by evaluating the energy expectation value E_T of the Nagaoka-like trial state $|\psi_t\rangle$,

$$E_T = E_K + E_U + E_J + E_V, \tag{8}$$

where $E_K = \langle\psi_t|H_K|\psi_t\rangle/\langle\psi_t|\psi_t\rangle$ and expressions for E_U, E_J, and E_V can be defined similarly. $E_U = 0$ since every individual orbital is, at most, singly occupied in the $U \to \infty$ limit. Since $|\psi_t\rangle$ describes a fully polarized state, any two electrons form a spin triplet, and $E_J = 0$ as well. E_V can be evaluated easily by noting that the hole distributions on different layers are uncorrelated for this trial state, and thus,

$$E_V = V\sum_{r,a>b} \frac{\langle\psi_t|1-n_a(\mathbf{r})|\psi_t\rangle\langle\psi_t|1-n_b(\mathbf{r})|\psi_t\rangle}{\langle\psi_t|\psi_t\rangle^2} = 3\frac{n_h^2}{L}V. \tag{9}$$

The upper bound on the kinetic energy can be estimated as follows. E_K is the sum of the kinetic energies of each layer. Up to a constant, the dispersion for each band can be rewritten in terms of hole number occupation as

$$H_{xy}^K = -4t\sum_{\mathbf{k},\sigma}\left(1-\frac{\mathbf{k}^2}{4}\right)[1-n_{xy,\sigma}(\mathbf{k})], \tag{10}$$

with parabolic dispersion near $k = 0$ in the $n_h/L^2 \to 0$ limit. Expressions for H_{yz}^K and H_{zx}^K follow by changing the indices. Since the trial state corresponds to removing n_h electrons from the band maximum of the fully filled Brillouin zone, or, equivalently, adding n_h holes to the band minimum in the hole description, the kinetic energy for a single layer can be estimated as

$$-4n_ht + tL^2\int_0^{k_0}\frac{k^3dk}{2\pi} = -4n_ht + tO\left(\frac{n_h^2}{L^2}\right), \tag{11}$$

where $(L/2\pi)^2\pi k_0^2 \approx n_h$. Summing over all $3L$ layers gives

$$E_K = -12n_hLt + tO\left(\frac{n_h^2}{L}\right). \tag{12}$$

Including the E_V contribution, the total trial state energy serves as an upper bound on the ground-state energy E_g of

$$E_g \leqslant -12n_hLt + tO\left(\frac{n_h^2}{L}\right) + 3V\frac{n_h^2}{L}. \tag{13}$$

B. Estimation of the lower bound

Since both H^J and H^V are non-negative operators, their lower bounds are zero. Thus, a lower bound on H^K is also a lower bound on E_g. Since H^K is the sum of the kinetic energies of each layer, the sum of the lower bounds on the kinetic energy of each layer is also a lower bound on E_g. The lower bound on the kinetic energy of each layer is simply $-4n_ht$, which has been worked out [16,17] by applying the Gershgorin circle theorem and considering a configuration where each hole has no neighboring holes. A brief review of this result is provided in the Appendix. Summing over each layer, we arrive at the lower bound on the ground-state energy

$$-12n_hLt \leqslant E_g. \tag{14}$$

Combining the upper and lower bounds, the ground-state energy satisfies

$$-12n_hLt \leqslant E_g \leqslant -12n_hLt + tO\left(\frac{n_h^2}{L}\right) + 3V\frac{n_h^2}{L}. \tag{15}$$

So far, we have assumed the same number of holes n_h in each layer. In fact, the result of Eq. (15) can be straightforwardly generalized to the case with different number of holes in different layers as

$$-4N_ht \leqslant E_g \leqslant -4N_ht + tO\left(\frac{n_{h,m}^2}{L}\right) + 3VO\left(\frac{n_{h,m}^2}{L}\right), \tag{16}$$

where N_h is the sum of hole numbers of all layers and $n_{h,m}$ is the maximal layer hole number. As a result, the generalized Nagaoka trial state becomes degenerate with the energy of the ground state E_g in the thermodynamic limit, when the maximal layer hole number $n_{h,m}$ in each layer scales as L^α with $\alpha < \frac{1}{2}$. Hence, the total bulk hole number can increase to the order of $L^{\frac{3}{2}}$, which is higher than the single-band case in the 3D cubic lattice [16,17], in which $\alpha < \frac{6}{5}$. This is due to the combined effect of the quasi-2D band structure and Hund's coupling.

C. Instability against a single spin flip

The Shastry-Krishnamurthy-Anderson method [21,22] provides a useful way to identify a region of instability of the Nagaoka trial state. Here we generalize it for the multiorbital systems. The method considers modifying the state by removing a spin-up particle from the Fermi surface of one orbital layer, flipping its spin, and adding it back to the bottom of the spin-down band. The $U = \infty$ limit is enforced by projecting out doubly occupied orbitals in real space. This procedure takes the form

$$|\phi_{l,a}\rangle = \prod_{\mathbf{r}\in l}[1 - n_{a\uparrow}(\mathbf{r})n_{a\downarrow}(\mathbf{r})]d^{\dagger}_{a\downarrow}(\mathbf{q})d_{a\uparrow}(\mathbf{k_F})|\psi_t\rangle, \quad (17)$$

where $\mathbf{r}$, $\mathbf{q}$, and $\mathbf{k_F}$ are all in layer l and orbital type a. Here $\mathbf{k_F}$ is a Fermi wave vector, and $\mathbf{q}$ is a momentum vector at the band bottom. The energy difference between $|\phi_{l,a}\rangle$ and $|\psi_t\rangle$ as a function of the filling can be computed, and a region of instability can be identified when $|\phi_{l,a}\rangle$ has lower energy. Since a spin has been flipped in only one band relative to the fully polarized generalized Nagaoka state, the energy difference between $|\phi_{l,a}\rangle$ and $|\psi_t\rangle$ follows only from the kinetic energy change in the up and down spins in layer l and from Hund's interaction between orbital a and the other two orbital types at sites on layer l.

The kinetic energy difference due to a spin flip in a single band on a square lattice, as evaluated in Ref. [21], is

$$\Delta E_K = -\epsilon_F - \frac{E_{Nag}}{n_h} - 4t\frac{n_h}{L^2}\left[1 - \left(\frac{E_{Nag}}{4tn_h}\right)^2\right], \quad (18)$$

where $E_{Nag}/L^2 = \int_{-4t}^{\epsilon_F} \epsilon\rho_{2D}(\epsilon)d\epsilon$ and $n_h/L^2 = \int_{\epsilon_F}^{4t} \rho_{2D}(\epsilon)d\epsilon$. Here $\rho_{2D}(\epsilon) = \frac{1}{2\pi^2 t}\Theta(4t - |\epsilon|)K(1 - \epsilon^2/16t^2)$, with K being a complete elliptic integral of the first kind, is the density of states for a square lattice with nearest-neighbor hopping. E_h is the kinetic energy of a single-band Nagaoka state with n_h holes on a square lattice, corresponding to the state in Eq. (6).

Since H^V is concerned only with the number of holes, flipping a single spin does not change E_V, i.e., $\Delta E_V = 0$, as can be verified by explicit calculation. What remains is then to evaluate $\Delta E_J = \langle\phi_{l,a}|H^J|\phi_{l,a}\rangle/\langle\phi_{l,a}|\phi_{l,a}\rangle$. Expressing the spin operators in terms of d_σ and $d^{\dagger}_\sigma$, the only terms that can possibly contribute to $\langle H^J\rangle$ are those involving a down-spin operator in only orbital a,

$$\Delta E_J = \frac{J}{2}\sum_{\substack{\mathbf{r}\in l,\\ b\neq a}} \frac{\langle\phi_{l,a}|n_{a\downarrow}(\mathbf{r})n_{b\uparrow}(\mathbf{r})|\phi_{l,a}\rangle}{\langle\phi_{l,a}|\phi_{l,a}\rangle}. \quad (19)$$

Since $|\phi_{l,a}\rangle$ is a direct product of wave functions of each layer, $n_{a\downarrow}$ and $n_{b\uparrow}$ are uncorrelated, in spite of the strong intralayer correlations. Then we have

$$\Delta E_J = \frac{J}{2}\sum_{\substack{\mathbf{r}\in l,\\ b\neq a}} \frac{\langle\phi_{l,a}|n_{a\downarrow}(\mathbf{r})|\phi_{l,a}\rangle\langle\phi_{l,a}|n_{b\uparrow}(\mathbf{r})|\phi_{l,a}\rangle}{|\langle\phi_{l,a}|\phi_{l,a}\rangle|^2}$$

$$= \frac{J}{2}L^2\sum_{b\neq a}\bar{n}_{la\downarrow}\bar{n}_{lb\uparrow}, \quad (20)$$

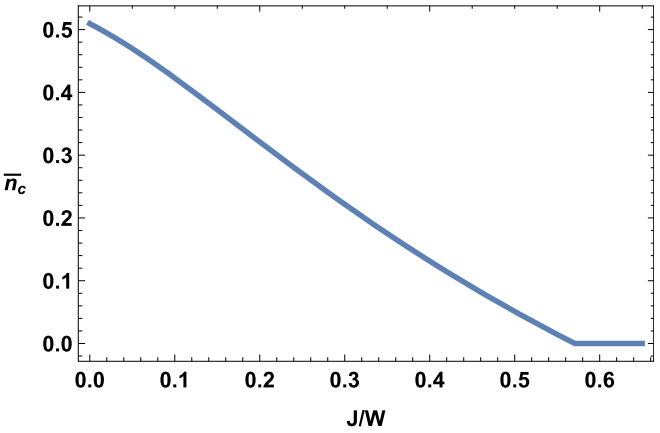

FIG. 1. The electron density $\bar{n}_c$ below which the generalized Nagaoka state becomes unstable to a single spin flip. $W = 8t$ is the bandwidth for a 2D square band. When J/W is larger than a critical value around 0.57, a single spin flip is not sufficient to destabilize the state for any electron density.

where $\bar{n}_{la\downarrow}$ and $\bar{n}_{lb\uparrow}$ are independent of $\mathbf{r}$ since $|\phi_{l,a}\rangle$ is a momentum eigenstate. It is easy to evaluate that $\bar{n}_{la\downarrow} = 1/L^2$ and $\bar{n}_{lb\uparrow} = 1 - n_h/L^2$; hence,

$$\Delta E_J = J\bar{n}, \quad (21)$$

where $\bar{n} = 1 - \frac{n_h}{L^2}$ is the electron density per site in the orbital plane.

Combining the Hund's interaction energy change with the kinetic energy change from Ref. [21], we have

$$\Delta E(n)/t = (\Delta E_K + \Delta E_V + \Delta E_J)/t$$
$$= -w - 4(1 - \bar{n}) + \frac{wy}{1 - \bar{n}}\left(\frac{wy}{4} - 1\right) + \frac{J}{t}\bar{n}, \quad (22)$$

where $w = \epsilon_F/t$, $y = \int_{-4/w}^{1} x\rho_{2D}(x\epsilon_F)dx$, and $w(\bar{n})$ is determined through the relation of $\bar{n} = \int_{-4/w}^{1}\rho_{2D}(x\epsilon_F)dx$. The electron density $\bar{n}_c$ below which the generalized Nagaoka state becomes unstable to a single spin flip can be solved by requiring $\Delta E(\bar{n}_c) = 0$. The critical density $\bar{n}_c(J/W)$ is plotted in Fig. 1, where $W = 8t$ is the bandwidth. As J increases, the ferromagnetic ground state becomes more and more stable. There exists a value of $J/W \approx 0.57$, beyond which the Nagoka state is stable against a single spin flip at any electron density.

IV. THE STABILITY OF THE t_{2g} NAGAOKA STATE WITH 3D BAND STRUCTURE

In this section, we consider the stability of the 3D Nagaoka state with t_{2g} orbitals and a 3D band structure.

Consider a Hamiltonian

$$H = H^K + H^U + H^V + H^J \quad (23)$$

as before, but where the kinetic terms now allow for perpendicular hopping within the same d orbital. Electrons now hop along the cube, remaining in the same orbital type, and the system is now composed of three cubic orbital bands. Explicitly, the perpendicular d_{xy} orbital hopping modifies

H_{xy}^K to

$$H_{xy}^K = t\sum_{\mathbf{r},\sigma}[d_{xy,\sigma}^\dagger(\mathbf{r})d_{xy,\sigma}(\mathbf{r}+\hat{x}) + d_{xy,\sigma}^\dagger(\mathbf{r})d_{xy,\sigma}(\mathbf{r}+\hat{y}) + d_{xy,\sigma}^\dagger(\mathbf{r})d_{xy,\sigma}(\mathbf{r}+\hat{z}) + \text{H.c.}]. \quad (24)$$

The hopping Hamiltonians of the d_{yz} and d_{xz} orbital bands can be similarly modified.

We can prove in the case where each orbital band has exactly one hole that the Nagaoka state is the unique ground state, up to trivial spin degeneracy. This can be done through Perron-Frobenius methods used in Refs. [15,20] as follows. Since the off-diagonal matrix elements, the hopping terms and spin-flipping Hund's coupling terms, all have negative matrix elements in the basis used in Ref. [20], the nonpositivity condition is satisfied. Now let us check the transitivity condition. Within each orbital band, it is satisfied since any two spins can be exchanged by repeatedly exchanging neighboring spins by cycling the hole around the square plackets. Spins in different orbitals can be exchanged by moving the spins to the same site, exchanging them using the Hund's coupling, and returning the spins to their original positions following the method presented in Ref. [20]. Since both the connectivity and nonpositivity conditions of the Perron-Frobenius theorem are satisfied, the ground state must be a positive-weight superposition of all basis elements. Since the maximum total-spin state is symmetric under the exchange of any two spins, it has nonzero overlap with this positive-weight superposition, and thus, the positive-weight superposition must be a maximum total-spin state due to the SU(2) symmetry.

With the Nagaoka-like state established as the ground state when there is a single hole in each of the three orbital bands, the stability of this state can be analyzed in the presence of multiple holes as in the case of 2D band structure studied above. We define the reference state $|R_{a,\uparrow}\rangle$ where all momentum states $\mathbf{k}$ in the 3D Brillouin zone are filled. In this case, there is no need for a layer index. Adding n_h holes to each band then takes the form of Eq. (6) with no layer index. The trial state of interest is then

$$|\psi_t\rangle = |h_{xy,\uparrow}\rangle \otimes |h_{yz,\uparrow}\rangle \otimes |h_{xz,\uparrow}\rangle, \quad (25)$$

which corresponds again to filling holes up to their Fermi energy in each band.

Now let us calculate the energy expectation value of the trial state $|\psi_t\rangle$. $E_U = 0$ since no orbital is doubly occupied, and $E_J = 0$ since the trial state is fully spin polarized. In this case,

$$E_V = V\sum_{r,a>b}\frac{\langle\psi_t|1-n_a(\mathbf{r})|\psi_t\rangle\langle\psi_t|1-n_b(\mathbf{r})|\psi_t\rangle}{\langle\psi_t|\psi_t\rangle^2} = 3\frac{n_h^2}{L^3}V \quad (26)$$

since the n_h holes in each band are now distributed over L^3 sites. The kinetic energy E_K can now be evaluated for each band as

$$-6n_ht + tL^3\int_0^{k_0}\frac{k^4dk}{4\pi^2} = -6n_ht + tO\left(\frac{n_h^{5/3}}{L^2}\right), \quad (27)$$

where $6L^3/\pi^2k_0^3 \approx n_h$. Including all three bands gives a factor of 3, and the resulting upper bound for the ground-state energy is

$$E_g \leqslant -18n_ht + tO\left(\frac{n_h^{5/3}}{L^2}\right) + 3V\frac{n_h^2}{L^3}. \quad (28)$$

The lower bound follows like in the t_{2g} case. In the case of 3D bands, the lower bound to the kinetic energy follows from maximizing the number of possible hole hoppings, which allows each hole to hop to six neighboring sites. Thus, each of the three bands contributes $-6n_ht$ to the lower bound, and the ground-state energy is bounded by

$$-18n_ht \leqslant E_g \leqslant -18n_ht + tO\left(\frac{n_h^{5/3}}{L^2}\right) + 3V\frac{n_h^2}{L^3}. \quad (29)$$

This Nagaoka-type trial state will be degenerate with the ground state in the thermodynamic limit when n_h scales as L^α, where $\alpha < \frac{6}{5}$.

An instability analysis of the ferromagnetic state for the case with 3D band structure can be performed as in Sec. III C. The resulting kinetic energy change is [21]

$$\Delta E_K = -\epsilon_F - \frac{E_{Nag}}{n_h} - 6t\frac{n_h}{L^3}\left[1-\left(\frac{E_{Nag}}{6tn_h}\right)^2\right], \quad (30)$$

where $E_{Nag}/L^3 = \int_{-6t}^{\epsilon_F}\epsilon\rho_{3D}(\epsilon)d\epsilon$ and $n_h/L^3 = \int_{\epsilon_F}^{6t}\rho_{3D}(\epsilon)d\epsilon$, with $\rho_{3D}(\epsilon)$ being the density of states on a 3D simple cubic lattice. The energy change due to H^V and H^J can be shown to take the same form as in the case of 2D bands with $\Delta E_V = 0$ and $\Delta E_J = J\bar{n}$, where $\bar{n}$ is the electron density in each orbital band defined as $\bar{n} = 1 - n_h/L^3$. Then the total energy change can be expressed as

$$\frac{\Delta E(\bar{n})}{t} = -w - 6(1-\bar{n}) + \frac{wy}{1-\bar{n}}\left(\frac{wy}{6}-1\right) + \frac{J}{t}\bar{n}, \quad (31)$$

where $w = \epsilon_F/t$, $y = \int_{-6/w}^1 x\rho_{3D}(x\epsilon_f)dx$, and $w(\bar{n})$ is determined by $\bar{n} = \int_{-6/w}^1\rho_{3D}(x\epsilon_F)dx$.

Again, the critical density $\bar{n}_c(J/W)$ below which the Nagaoka-like state becomes unstable to a single spin flip is solved and shown in Fig. 2, where $W = 12t$ is the bandwidth. At $J = 0$, the value of $\bar{n}_c = 0.68$ is consistent with previous results in Ref. [21] for a model with a single 3D band. The Hund's coupling further stabilizes the Nagaoka-like state, which is similar to the case with 2D band structure in Sec. III C. However, a significant difference is that $\bar{n}_c$ does not drop to zero even at large values of J/W.

The different behavior of $\bar{n}_c$ for 2D and 3D bands is due to the different scalings of the density of states at low energy. It is easy to check that in the low-density limit, the energy costs of a single spin flip in Eqs. (22) and (31) can be expanded to the leading order as

$$\Delta E_d(\bar{n}_d) \approx -(\epsilon_F - \epsilon_b) + J\bar{n}_d, \quad (32)$$

where ϵ_b is the band bottom energy and $\bar{n}_d$ is the particle density for d-dimensional bands. In the low-density limit, $\epsilon_F - \epsilon_b \propto (\bar{n}_d)^{2/d}$. In three dimensions, the kinetic energy

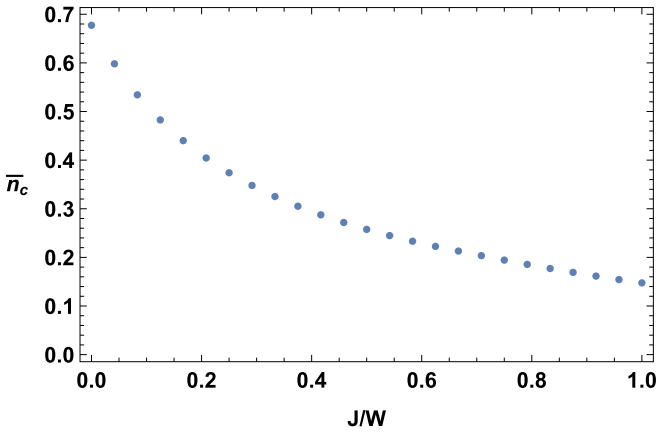

FIG. 2. The critical value of electron density $\bar{n}_c$ below which the Nagaoka-like state $|\psi_t^{3D}\rangle$ becomes unstable to a single spin flip. $W = 12t$ is the bandwidth for the 3D cubic bands. Unlike in the case of 2D bands, $\bar{n}_c$ does not approach zero for finite J/W because the 3D density of states vanishes at the low-density limit.

change in Eq. (32) dominates the Hund's coupling energy cost, allowing a single spin flip to lower the total energy and destabilize the Nagaoka state. By contrast, both terms in Eq. (32) scale the same in two dimensions, and a single spin flip costs energy when J is large enough.

V. CONCLUSIONS AND DISCUSSION

We have studied the stability of the generalized Nagaoka ferromagnetic state in a 3D cubic lattice with t_{2g} orbitals. Applying the bounding method of Refs. [16,17], for a cubic lattice with size $L \times L \times L$ and a quasi-2D t_{2g} orbital band structure, the fully polarized Nagaoka state becomes degenerate with the ground state as $L \to \infty$ when the number of holes in each orbital plane scales slower than $L^{\frac{1}{2}}$ or the total hole number scales slower than $L^{\frac{3}{2}}$. For the case with 3D band structure, we have generalized the Nagaoka theorem to the case that each orbital has a single hole. Again, for the multihole case, the fully polarized Nagaoka ferromagnetic state remains degenerate with the ground state at $L \to \infty$ when the hole number scales slower than $L^{\frac{6}{5}}$. These results apply in the limit of $U \to \infty$ and arbitrary ferromagnetic Hund's coupling $J > 0$ and interorbital repulsion $V \geqslant 0$. We have also examined the stability of the orbital-generalized Nagaoka states against a single spin flip for both quasi-2D and 3D band structures. In both cases, the instability region shrinks as Hund's coupling increases.

The above bounding estimation proves only the degeneracy of the Nagaoka-type ferromagnetic state with the ground state but does not prove the uniqueness of the ground state. Hence, even within the above bounds, the above results actually do not prove the ground-state ferromagnetism for the multihole case. Nevertheless, the stability of the ground-state ferromagnetism is still conceivable. The above analysis does not imply that the fully polarized state must break down when the hole number exceeds the above bounds. Recent numerical calculations based on the density-matrix renormalization-group method have shown evidence of the stability of the Nagaoka state at finite hole densities for the 2D single-band case [23], although exact proof remains an open question.

The above study is not just of academic interest. In fact, itinerant ferromagnetism has been discovered in the t_{2g} orbital active material $SrRuO_3$, which is a weak ferromagnet with partial polarization and Curie temperature $T_c \approx 160$ K [24–26]. Its electronic structure can be modeled by the multiorbital Hubbard model with the quasi-2D band structure and the prominent Hund's coupling. Certainly, the filling is 4/3 electron per orbital on each site and thus significantly far from the half filling, which corresponds to one electron per orbital. The real system of $SrRuO_3$ implies that the Hund's-rule-facilitated itinerant ferromagnetism may remain stable at finite values of U and away from half filling. Our work provides useful guidance for studying itinerant ferromagnetism in this class of materials.

ACKNOWLEDGMENTS

E.B. and Y.L. are supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, Grant No. DE-FG02-08ER46544.

APPENDIX: STABILITY OF THE ONE-BAND NAGAOKA STATE

The method of applying the Gershgorin circle theorem to study the stability of the Nagaoka state was used for a one-band Hubbard model on a square lattice in Ref. [16], where the model used corresponds to $H = H^U + H^K_{xy}$. The method, as generalized to a d-dimensional hypercubic lattice in Ref. [17], is reviewed here for completeness. For a single-band Hubbard model, the fully polarized Slater determinant state $|h\rangle$ in Eq. (6) can be taken as a variational trial state. In 2D bands on a simple square lattice, this trial state corresponds to a single orbital plane a,l. The trial state can be generalized to d-dimensional bands on a hypercubic lattice by taking $\mathbf{r}$ and $\mathbf{k}$ to be d-dimensional lattice and momentum vectors, respectively.

The energy E_K of this trial state was evaluated by considering the dispersion for a d-dimensional hypercubic lattice. In terms of the hole number occupation picture,

$$H^K = -2dt \sum_{\mathbf{k},\sigma} \left(1 - \frac{\mathbf{k}^2}{2d}\right)[1 - n_\sigma(\mathbf{k})]. \tag{A1}$$

This parabolic dispersion holds in the limit $n_h/L^d \to 0$. The energy of the trial state can then be evaluated as

$$E_K = -2dtn_h + tL^d \int_0^{k_0} \frac{\Omega_d k^{d+1} dk}{(2\pi)^d}, \tag{A2}$$

where Ω_d is the surface area of a unit d sphere and $(L/2\pi)^d V_d k_0^d \approx n_h$, with V_d being the volume of a unit d sphere. The upper bound on the ground-state energy is then

$$E_g \leqslant -2dtn_h + tO\left(\frac{n_h^{\frac{d+2}{d}}}{L^2}\right). \tag{A3}$$

The lower bound follows from the Gershgorin circle theorem, which states that for any eigenvalue λ of a square matrix H,

there exists a row i such that

$$|\lambda - H_{ii}| \leqslant \sum_{j\neq i} |H_{ij}|. \tag{A4}$$

It follows that a lower bound on the ground-state energy is given by

$$\min_i \left\{ H_{ii} - \sum_{j\neq i} |H_{ij}| \right\} \leqslant E_g. \tag{A5}$$

Intuitively, the lower bound is the configuration that minimizes the energy of an analogous bosonic system, where sign changes from hopping are neglected. For the 2D square lattice, this configuration corresponds to placing all holes on either the even or odd sublattice, allowing each hole to hop to four neighboring sites. Thus,

$$-4n_h t \leqslant E_g. \tag{A6}$$

The lower and upper bounds on the ground-state energy coincide in the thermodynamic limit as long as $n_h \sim L^\alpha$, where $0 \leqslant \alpha < \frac{2d}{d+2}$. Thus, the fully spin polarized trial state is a ground state that remains stable for a number of holes that grows sufficiently slowly.

[1] E. C. Stoner, Proc. R. Soc. London, Ser. A **169**, 339 (1939).
[2] E. H. Lieb and D. Mattis, Phys. Rev. **125**, 164 (1962).
[3] Y. Nagaoka, Phys. Rev. **147**, 392 (1966).
[4] E. H. Lieb, Phys. Rev. Lett. **62**, 1201 (1989).
[5] A. Mielke, J. Phys. A **24**, L73 (1991).
[6] A. Mielke, J. Phys. A **24**, 3311 (1991).
[7] A. Mielke, J. Phys. A **25**, 4335 (1992).
[8] H. Tasaki, Phys. Rev. Lett. **69**, 1608 (1992).
[9] A. Mielke and H. Tasaki, Commun. Math. Phys. **158**, 341 (1993).
[10] H. Tasaki, Commun. Math. Phys. **242**, 445 (2003).
[11] E. H. Lieb, in *XIth International Congress of Mathematical Physics*, edited by D. Iagolnitzer (International Press of Boston, Boston, 1995), pp. 392–412.
[12] J. E. Hirsch, Phys. Rev. B **40**, 2354 (1989).
[13] A. Tanaka and H. Tasaki, Phys. Rev. Lett. **98**, 116402 (2007).
[14] D.-P. Zhang, G.-S. Tian, and H.-Q. Lin, Commun. Theor. Phys. **63**, 261 (2015).
[15] H. Tasaki, Phys. Rev. B **40**, 9192 (1989).
[16] G.-S. Tian, Phys. Rev. B **44**, 4444 (1991).
[17] S.-Q. Shen, Z.-M. Qiu, and G.-S. Tian, Phys. Lett. A **178**, 426 (1993).
[18] Y. Li, E. H. Lieb, and C. Wu, Phys. Rev. Lett. **112**, 217201 (2014).
[19] S. Xu, Y. Li, and C. Wu, Phys. Rev. X **5**, 021032 (2015).
[20] Y. Li, Phys. Rev. B **91**, 115122 (2015).
[21] B. S. Shastry, H. R. Krishnamurthy, and P. W. Anderson, Phys. Rev. B **41**, 2375 (1990).
[22] P. Fazekas, *Lecture Notes on Electron Correlation and Magnetism* (World Scientific, Singapore, 1999), pp. 729–732.
[23] L. Liu, H. Yao, E. Berg, S. R. White, and S. A. Kivelson, Phys. Rev. Lett. **108**, 126406 (2012).
[24] D. W. Jeong, H. C. Choi, C. H. Kim, S. H. Chang, C. H. Sohn, H. J. Park, T. D. Kang, D.-Y. Cho, S. H. Baek, C. B. Eom, J. H. Shim, J. Yu, K. W. Kim, S. J. Moon, and T. W. Noh, Phys. Rev. Lett. **110**, 247202 (2013).
[25] G. Koster, L. Klein, W. Siemons, G. Rijnders, J. S. Dodge, C.-B. Eom, D. H. A. Blank, and M. R. Beasley, Rev. Mod. Phys. **84**, 253 (2012).
[26] G. Cao, S. McCall, M. Shepard, J. E. Crow, and R. P. Guertin, Phys. Rev. B **56**, 321 (1997).