Suppression of effective spin-orbit coupling by thermal fluctuations in spin-orbit coupled antiferromagnets

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We apply the finite-temperature variational cluster approach to a strongly correlated and spin-orbit coupled model for four electrons (i.e. two holes) in the $t_{2g}$ subshell. We focus on parameters suitable for antiferromagnetic Mott insulators, in particular Ca$_2$RuO$_4$, and identify a crossover from the low-temperature regime, where spin-orbit coupling is essential, to the high-temperature regime where it leaves few signatures. The crossover is seen in one-particle spectra, where $xz$ and $yz$ spectra are almost one dimensional (as expected for weak spin-orbit coupling) at high temperature. At lower temperature, where spin-orbit coupling mixes all three orbitals, they become more two dimensional. However, stronger effects are seen in two-particle observables like the weight in states with definite onsite angular momentum. We thus identify the enigmatic intermediate-temperature ‘orbital-order phase transition’, which has been reported in various X-ray diffraction and absorption experiments at $T \approx 260\,K$, as the signature of the onset of spin-orbital correlations.

I. INTRODUCTION

Ruthenium oxides have for decades attracted considerable attention, first for their complex phase diagrams that bear some similarities to those high-$T_N$ cuprate superconductors, with superconducting and Mott insulating phases \cite{[1]}. More recently, the interplay of spin-orbit coupling (SOC), electron itineracy, electronic correlations, and lattice degrees of freedom, which are all present, has attracted attention. The exotic behavior emerging on this stage includes a potential spin liquid in $\alpha$-RuCl$_3$ \cite{[2, 3]}, enigmatic superconductivity in Sr$_2$RuO$_4$ \cite{[4] and a non-equilibrium Weyl semi metal in Ca$_2$RuO$_4$ \cite{[5].

This last compound, Ca$_2$RuO$_4$, had already been discussed as a Mott insulating end member of the family of compounds including enigmatic superconductors. Its high-temperature metal-insulator transition has been well described by a combination of density-functional theory and dynamical mean-field theory \cite{[6]. The emerging picture is that of a lattice-supported Mott transition, where the $xy$ orbital is lowered in energy and becomes nearly doubly occupied, while a Mott gap opens in the approximately half filled $xz$ and $yz$ orbitals. SOC, which had alternatively been argued to drive the metal-insulator transition\cite{[7], was later shown to have only a weak impact on the gap \cite{[8].

However, this changes decisively when it comes to the magnetic properties of the antiferromagnetic state observed at even lower temperatures. For weak SOC and dominant crystal field (CF), one would expect the half-filled $xz$ and $yz$ orbitals to form a spin one, while the doubly occupied $xy$ orbital would be magnetically inert. However, magnetic excitations turn out to show pronounced X-Y-symmetry as well as Higgs modes \cite{[9, 10], which can more naturally be explained in terms of excitonic antiferromagnetism, which fundamentally relies on SOC.

Orbital angular momentum of two $t_{2g}$ holes can be modeled as an effective $L = 1$. In the idealized picture of an undistorted Ru-O octahedron (i.e. with equivalent $xy$, $yz$, and $xz$ orbitals) SOC would couple total spin $S = 1$ with $L = 1$ into a singlet ground state with total angular momentum $J = 0$ \cite{[10]. When superexchange connects ions, however, higher-energy triplets gain kinetic energy and may condense into a magnetically ordered state. In one dimension, the resulting ground-state phase diagram has been established by use of the density-matrix renormalization group and includes a parameter regime supporting excitonic magnetism \cite{[11, 12]. Recent numerical work using a combination of density-functional theory and variational cluster approach (VCA) has further indicated that the excitonic scenario with SOC as a decisive player indeed applies to the antiferromagnetic low-temperature state of Ca$_2$RuO$_4$ \cite{[13].

It would thus be highly desirable to investigate temperatures between the ground state with a large role for SOC and the high-temperature state, where it only yields small corrections, also with a view towards other ruthenates with similar energy scales. While the metal-insulator transition and the interplay of lattice and correlations is accessible to dynamical mean-field theory with a Monte-Carlo impurity solver, the fermionic minus-sign problem is present at lower temperatures \cite{[14]. Adjusted one-particle states based on total angular momentum can reduce the minus-sign problem \cite{[15}, however, such an optimal basis cannot easily be identified in realistic models, where CFs or anisotropic hoppings compete with SOC.

In order to address low temperature scales of spin-orbit coupled $t_{2g}$ orbitals, described by the three-orbital Hubbard model of Sec. \cite{[11} we thus implement a finite-temperature variant of the VCA, see Sec. \cite{[11}. Exact diagonalization is used to solve a small cluster and to extract its self energy, which is then used to evaluate the Green’s function of the thermodynamic limit. This al-
ows us to treat the antiferromagnetic order, and since we focus here on the Mott insulating regime, bath sites are less necessary.

Based on the results presented in Sec. III we identify a temperature range above the Néel temperature, but in the Mott insulating regime, where the spin-orbital character strongly changes. While the onsite-singlet and triplet states describe the ionic state at low temperatures very well, as expected for the excitonic scenario, they become less useful at higher temperatures. Here, the original orbitals provide a clearer picture, especially in the presence of a CF, as will be seen in the one-particle spectra discussed in Sec. III B.

In $t_{2g}$ models with SOC of a magnitude suitable for excitonic magnetism, there is thus a third temperature scale intermediate between the metal-insulator transition related to charge fluctuations and lattice distortions and the Néel temperature related to magnetic degrees of freedom. We discuss in Sec. IV how this ties in with the enigmatic ‘orbital ordering’ transition that has been debated at intermediate temperatures in Ca$_2$RuO$_4$ [16,17].

II. MODEL AND METHODS

We study here a three-orbital Hubbard model for $t_{2g}$ electrons on a square lattice, where we focus on nearest-neighbor (NN) hopping and tetragonal symmetry. The kinetic energy is then diagonal in orbital indices and takes the form

$$H_{\text{kin}} = -t \sum_{\langle i,j \rangle, \sigma} c_{i,x,y,\sigma}^\dagger c_{j,x,y,\sigma}$$

$$- t \sum_{\langle i,j \rangle} c_{i,x,z,\sigma}^\dagger c_{j,x,z,\sigma} - t \sum_{\langle i,j \rangle} c_{i,y,z,\sigma}^\dagger c_{j,y,z,\sigma} + \text{H.c.}$$

where $c_{i,\alpha,\sigma}$ ($(c_{i,\alpha,\sigma})^\dagger$) annihilates (creates) an electron with spin $\sigma = \uparrow, \downarrow$ in orbital $\alpha = xy, xz, yz$ at site $i$. Nearest-neighbor bonds $(i,j)$ along the two directions $x$ and $y$ are considered and we use $t$ as our unit of energy.

Tetragonal CF splitting

$$H_{\text{CF}} = -\Delta \sum_{i,\sigma} n_{i,x,y,\sigma}$$

with $n_{i,x,y,\sigma} = c_{i,x,y,\sigma}^\dagger c_{i,x,y,\sigma}$ and $\Delta > 0$ is motivated by the shortened octahedra of the low-temperature phase of Ca$_2$RuO$_4$. For a filling of four electrons (two holes), it favors a doubly occupied $xy$ orbital with half filled $xz$ and $yz$ orbitals. We will tune it to interpolate between an orbitally polarized spin-one system at large $\Delta$ and a more equal interplay of degenerate orbitals at small $\Delta$.

In the present paper, the impact of SOC is particularly important, which takes the form

$$H_{\text{SOC}} = \lambda \sum_i \mathbf{l}_i \cdot \mathbf{s}_i = \frac{i\lambda}{2} \sum_{\alpha,\beta,\gamma, \sigma, \sigma'} \varepsilon_{\alpha\beta\gamma} \tau^\alpha_{\sigma\sigma'} c_{i,\beta,\sigma}^\dagger c_{i,\gamma,\sigma'}$$

for $t_{2g}$ orbitals, with the totally antisymmetric Levi-Civita tensor $\varepsilon_{\alpha\beta\gamma}$ and Pauli matrices $\tau^\alpha_{\sigma\sigma'}$, $\alpha = x, y, z$ [13–19]. We focus here on intermediate SOC that is not strong enough to suppress magnetic ordering.

Finally, there are effective onsite Coulomb interactions [20]

$$H_{\text{int}} = U \sum_{i,\sigma} n_{i,\sigma} n_{i,\bar{\sigma}} + \frac{U'}{2} \sum_{i,\sigma \neq \bar{\sigma}} n_{i,\sigma} n_{i,\bar{\sigma}}$$

$$- \frac{1}{2} (U' - J_H) \sum_{i,\sigma \neq \bar{\sigma}} n_{i,\sigma} n_{i,\bar{\sigma}}$$

$$- J_H \sum_{i,\alpha \neq \beta} (c_{i,\alpha}^\dagger c_{i,\beta}^\dagger c_{i,\beta} c_{i,\alpha} - c_{i,\beta}^\dagger c_{i,\alpha}^\dagger c_{i,\alpha} c_{i,\beta})$$

with Coulomb interaction $U$, $U'$ and Hund’s coupling $J_H$ connected via $U' = U - 2J_H$. We are here less interested in varying interactions and rather focus on the Mott insulating regime with a Hund’s-rule coupling larger than SOC, so that $\mathbf{L} \cdot \mathbf{S}$ coupling provides a clearer description than $j$-$j$ coupling. We thus choose $U = 12.5t$ and $J_H = 2.5t$, which is consistent with their order of magnitude in Ca$_2$RuO$_4$ [21].

A. Finite-temperature variational cluster approach

We use the finite-temperature [22] VCA [23,24], where the grand potential $\Omega$ of the system is approximated in terms of a ‘reference system’ that consists of small disconnected clusters, but has the same electron-electron interactions as the Hamiltonian of interest [25].

$$\Omega(\Sigma_{cl}) = \Omega_{cl} + \text{Tr} \ln(-G_{cl}^{-1}) - \text{Tr} \ln(-G_{\text{CPT}}^{-1})$$

with the grand potential $\Omega_{cl}$ and Green’s function $G_{cl}$ obtained from the cluster. The CPT-Green’s Function $G_{\text{CPT}} = (G^{-1}_{cl} - G^{-1}_0 + G^{-1}_0)^{-1}$ replaces the non-interacting cluster Green’s function $G_{cl,0}$ by the non-interacting Green’s function $G_0$ of the full system. The approximation thus consists in replacing the self energy of the physical system by that of the small cluster. In order to improve the approximation, the self-energy–functional approach [24] allows us to optimize the cluster self energy $\Sigma_{cl}$ by varying one-particle parameters of the reference Hamiltonian. The best approximation to the system’s grand potential is a stationary point of $\Omega$ w.r.t. the variational one-particle parameters. Note that this variation affects only the small cluster, the non-interacting Green’s function given by the one-particle part of the physical Hamiltonian remains fixed.

We numerically evaluate the cluster grand potential

$$\Omega_{cl} = -\beta^{-1} \ln(\Xi) = -\beta^{-1} \ln \sum_m e^{-\beta \varepsilon_m}$$

with partition function $\Xi$ and cluster-energies $\varepsilon_m$, as well
as the cluster Green’s function, whose electron part reads

\[ [G_{\text{cl}}^{(\tau)}]_{\alpha\beta}(z) = \sum_{m,n} e^{-\beta E_{nm}^*} \sum_k \frac{\langle \Psi_m | c_{\alpha} | \Psi_n^* \rangle \langle \Psi_n^* | c^\dagger_{\beta} | \Psi_m \rangle}{z - E_{nm}^* + i0^+} \]  

(7)

with energy difference \( E_{nm}^* = \varepsilon_m^* - \varepsilon_m \). We largely follow Seki et al. [22] and obtain the spectrum and eigenstates with band Lanczos to resolve (approximately) degenerate eigenenergies. We use eight starting vectors and converge 120 eigenvectors, which are used to evaluate the Green’s functions in a second Lanczos run. In this second step, band Lanczos did not turn out to be advantageous, and we thus use the conventional algorithm.

Assembling the cluster Green’s function is accelerated with the help of a high-frequency expansion for frequency arguments with absolute values larger than that of the largest pole [22]. [Other frequencies are obtained via [7].] Following Seki et al. [23], the high-energy Green’s function is expanded to 15-th order as

\[ G_{\alpha\beta}(z) = \sum_{k=0}^{\infty} M_{\alpha\beta}^{(k)} z^{-k+1} \]  

(8)

with moments \( M_{\alpha\beta}^{(0)}(z) = \delta_{\alpha\beta} z \) and

\[ M_{\alpha\beta}^{(k>0)} = \sum_{m,n} (E_{nm}^{*})^k \frac{e^{-\beta \varepsilon_m}}{z} \left( \langle \Psi_m | c_{\alpha} | \Psi_n^* \rangle \langle \Psi_n^* | c^\dagger_{\beta} | \Psi_m \rangle + \langle \Psi_n^* | c^\dagger_{\beta} | \Psi_m^* \rangle \langle \Psi_m^* | c_{\alpha} | \Psi_n \rangle \right). \]  

(9)

In order to fix the density to \( N = 4 \) electrons (i.e. two holes), the grand potential is transformed to the free energy \( F(N,V,T) = \Omega(\mu,V,T) + \mu N \) by means of a Legendre transform [26]. There are thus at least two variational parameters, the chemical potential \( \mu \) to fix the density and the cluster chemical potential \( \mu' \) to ensure thermodynamic consistency [27,28]. Additionally, we consider antiferromagnetic order parameters with ordered moment within the a-b-plane or along the c-axis. Previous work for \( T = 0 \) has shown that the z in-plane components lead to quite different grand potentials (as expected for finite SOC), but that the grand potential is very similar for operators like spin, magnetization, or total angular momentum [13]. For this reason and in order to easily compare to the spin-one antiferromagnet, we use here the spin as the order parameter. It has also been shown that a sizable CF as well as SOC both favor ‘checkerboard’ magnetic patterns with ordering vector \( Q = (\pi, \pi) \), so that we use the fictitious Weiss field

\[ H_{\text{Weiss}} = \hbar \sum_i e^{i Q r_i} S^z_i \]  

(10)

with \( i \) labeling the site at \( r_i \) and \( S^z_i = \sum_{\alpha,\beta} c^\dagger_{\alpha \beta} c_{\alpha,\beta} + \text{H.c. and } S^z = \sum_{\alpha,\beta} \langle c^\dagger_{\alpha \beta} c_{\alpha,\beta} \rangle \) are the x and z component of the total electron, respectively. For variational parameters \( \mu, \mu' \) and \( h \) giving stationary grand potentials, we evaluate one-body expectation values (like magnetization or orbital densities) from the Green’s function, as is done for \( T = 0 \). The entropy \( S \) is determined as the derivative of the grand potential via contour integrals:

\[ S = S_{\text{cl}} + \mathcal{A}_{\text{CPT}} + \mathcal{A}_{\text{cl}} - (\Omega_{\text{CPT}} - \Omega_{\text{cl}})/T \]  

(11)

with the contributions

\[ S_{\text{cl}} = -\langle \Omega_{\text{cl}} - \langle H \rangle_{\text{cl}} \rangle/T, \]  

(12)

\[ \mathcal{A}_{\text{CPT}} = \oint dz f(z) \text{tr}[(G_{\text{cl}} G_{\text{CPT}}^{-1} G_{\text{mod}}^{-1})]/T^2, \]  

(13)

\[ \mathcal{A}_{\text{cl}} = \oint dz f(z) \text{tr}[(G_{\text{cl}}^{-1} G_{\text{mod}}^{-1})]/T^2 \]  

(14)

and the abbreviations

\[ (H)_{\text{cl}} = \Xi^{-1} \sum_{m} \varepsilon_m \exp(-\beta \varepsilon_m), \]  

(15)

\[ G_{\text{mod}} = \sum_{m,n} \Xi^{-1} \varepsilon_m \exp(-\beta \varepsilon_m) [G_{\text{cl}}^* + G_{\text{cl}}] \]  

(16)

The specific heat \( C(T) \) is then obtained as the numerical derivative \( C(T) = T \Delta S/\Delta T \) of the entropy.

Since the ordered moment in an excitonic magnet arises through a superposition of the \( J = 0 \) state preferred by onsite SOC and \( J = 1 \) states [10], it is helpful to consider weights \( \langle J \rangle \) found in eigenstates of total onsite angular momentum \( J \). Unfortunately, this is a two-particle quantity and thus not readily available from the VCA. We approximate it as the exact-diagonalization expectation value obtained for the \( N_{\text{sites}} \) sites of the reference cluster with optimized parameters:

\[ \langle J \rangle := \frac{1}{N_{\text{sites}} \Xi} \sum_{i,J} \sum_m e^{-\beta \varepsilon_m} (m|J_i, J^z_i)(J_i, J^z_i|m). \]  

(17)

Eigenstates \( |J_i, J^z_i \rangle \) denote here the state at site \( i \) defined by angular momentum \( J = L + S \).

III. RESULTS

A. Temperature dependence of the onsite angular momentum

At \( T = 0 \), VCA for the \( t_{2g} \) model with four electrons and CF \( \Delta = 0 \) has revealed two different ordering patterns depending on \( \lambda \) [13]: at small \( \lambda \leq 0.4 t \), orbitals and spins order in a strip pattern with orthogonal ordering momenta \((0, \pi)\) for spins and \((\pi, 0)\) for orbitals. For larger \( \lambda \), excitonic antiferromagnetic (AFM) order with momentum \((\pi, \pi)\) takes over, where the out-of-plane \( z \) component is favored over in-plane directions. We are here interested in the latter regime and thus focus on \( \lambda > 0.4 t \).
FIG. 1. Thermodynamics of the excitonic antiferromagnet without CF splitting. (a) gives the magnetic order parameter, i.e. staggered out-of-plane spin and (b) the specific heat for SOC $\lambda = 0.6t, 0.8t, t$. (We do not intend to discuss the complicated spin and orbital stripe pattern found at $\lambda = \Delta = 0$ [13], and accordingly leave out the regime of small $\lambda \leq 0.4t$.)

FIG. 2. Temperature evolution of the average weight found in eigenstates of the total angular momentum, see Eq. (17). (a) $\lambda = 0.6t$, (b) $\lambda = 0.8t$, and (c) for $\lambda = t$.

Figure 1(a) shows the ordered spin moment depending on temperature. As expected, the value at $T = 0$ is reduced when larger $\lambda$ increases the energy gap between the ionic singlet and triplet states, which in turn reduces the triplet admixture into the ground state. Somewhat surprisingly, the Néel temperature is not monotonic. While we can certainly not exclude strong finite-size effects due to the 2x1-site cluster, an alternative explanation may be that system at smallest $\lambda = 0.6 t$ is affected by its closeness to the competing stripy phase. The corresponding specific heat is given in Fig. 1(b) and has a second broad hump at higher temperature $T > T_N$ in addition to the expected peak at the magnetic ordering transition. This feature exists for all three values of $\lambda$ and shifts to slightly higher temperatures for $\lambda = t$.

Figure 2 shows the average weight Eq. (17) found in eigenstates with $J = 0, 1, 2$ of the total onsite angular momentum. Weights are constant in the regime of constant magnetization, and the $J = 0$ ($J = 1$) state looses (gains) weight when magnetic order is lost. This is in clear contrast to a (somewhat artificial) transition to a paramagnet at constant temperature: reducing the ordering field $h$ at $T = 0$ pushes weight from the $J = 1$ states into the $J = 0$ state [13]. At $T_N$, the curves get abruptly steeper and weights in $J = 0$ and $J = 1$ states change substantially at higher temperatures $T > T_N$. Weight in the $J = 2$ states is completely negligible below $T_N$, but similarly begins to grow at $T > T_N$. We are going to argue that this spin-orbital rearrangement is the origin of the second hump in the specific heat.

For comparison, Fig. 3(a) gives the magnetization and specific heat for CF $\Delta = 5t$ that is large enough to enforce complete orbital polarization with a doubly occupied $xy$ orbital at all temperatures shown. The two holes are then found in $xz$ and $yz$ orbitals and form a conventional spin one, with an ordered moment close to two in the AFM state. The specific heat shown in Fig. 3(b) has here only the peak at the Néel temperature and no further features. The expected weights in eigenstates with total onsite angular momentum $J = 0, 1, 2$ are given in Fig. 3(c) and (d) and present a quite different picture from the excitonic case discussed in Fig. 2, while the weights in $J = 0$ and $J = 1$ states change appreciably below $T_N$, only little variation is seen above $T_N$.

Finally, Figs. 4 and 5 discuss intermediate $\Delta = 1.5t$, an order of magnitude appropriate to describe Ca$_2$RuO$_4$. Ground-state VCA calculations have here shown in-plane magnetic moments to be favored over out-of-plane moments [13], in agreement with the AFM state of Ca$_2$RuO$_4$. Again, the Néel temperature is not very sensitive to SOC $\lambda$ while the ordered moment is substantially reduced by it. The system without SOC has the largest ordered moment close to two, see Fig. 4(a). As will be discussed below, its $xy$ orbital is completely filled below the Néel temperature, see Fig. 5(a), so that it comes close to a spin-one scenario. Larger $\lambda \geq 0.6 t$ reduce the ordered moment, which indicates that orbital polarization is not strong enough to quench SOC. The specific heat shown...
in Fig. 4(b) looks qualitatively much more similar to the results for \( \Delta = 0 \) than to those for \( \Delta = 5t \), as a second hump at \( T > T_N \) is clearly seen.

While the transition from spin-one to excitonic antiferromagnetism is a gradual crossover, it was estimated to occur at \( \lambda = 0.7t \) in the ground state [13]. The \( J \)-weights qualitatively agree, with Fig. 5(e) for \( \lambda = 0 \) being similar to the spin-one scenario of Fig. 3(c,d), while Figs. 5(g,h) for \( \lambda = 0.8t \) and \( \lambda = 1 \) resemble more the excitonic case of Fig. 2 Figure 5(f) for \( \lambda = 0.6t \) lies somewhere in between, again in line with the previous estimate.

The second hump in the specific heat for \( \lambda = 0 \) can be understood by noting that the orbital densities in Fig. 3(a) do not remain constant above \( T_N \). Since the CF is here just strong enough to fill the \( xy \) orbital at \( T = 0 \), finite temperature can induce \( xy \)-holes and these orbital fluctuations are reflected in the specific heat. The weights found in states \( J = 0, 1, 2 \), in contrast do here not change above \( T_N \), see Fig. 5(e), when there is no SOC.

In the opposite limit \( \lambda = t \), the orbital densities are nearly constant, a small difference between \( xz \) and \( yz \) below \( T_N \) being due to magnetic symmetry breaking. Weights in \( J = 0 \) and \( J = 1 \) states depend here strongly on temperature at \( T > T_N \), see Fig. 5(h). While low \( T < T_N \) strongly suppressed any weight in \( J = 2 \) states for \( \Delta = 0 \), see Fig. 2, it is here nearly constant, because it is connected to the clear orbital polarization \( n_{xy} > n_{xz/yz} \) [13].

**B. Signatures of SOC in one-particle spectra**

Figure 6 shows the VCA one-particle spectral density for \( \Delta = 0 \) and temperatures \( T = 0, T > T_N \) and \( T > T_N \). At all temperatures, the occupied states are split into three subbands at energies \( \omega \leq 5t, \omega \approx 10t \), and \( \omega \geq 15t \) (with the last having lower weight), which can be related to Hund’s-rule coupling [21]. Below \( T_N \), some signatures of the doubling of the unit cell are visible in the form of shadow bands around \((0,0)\) and \((\pi, \pi)\). Apart from this feature, the predominant effect of temperature is making the spectra less coherent. Overall, temperature effects are here rather subtle.

Temperature-driven orbital reconstruction reveals itself slightly more when CF and SOC compete, see the one-particle spectra shown in Fig. 6 for \( \Delta = 1.5t \) and \( \lambda = t \). The ground-state spectrum Fig. 6(a-c) shows again a slight shadow band due to the doubling of the unit cell and both the empty band (of predominantly \( xz/yz \) character) and the highest occupied band (of predominantly \( xy \) character) have a two-dimensional dispersion, similar to Fig. 6(a-c). In the spectra taken around \( T_N \), see Fig. 6(d-f), the shadow band has vanished. The occupied \( xz/yz \) states have become more coherent than in the ground state. This rather unconventional behavior may be related to the ladder-like features that were re-
cently found in a strong-coupling t-J-like model without SOC, where they arise in the AFM state due to the anisotropic hoppings of these orbitals [29]: when magnetic order is lost, the ladder features become weaker and the underlying dispersion is seen more easily. It is rather one-dimensional, as expected for $xz$ and $yz$ orbitals without SOC. Such a weak impact of SOC at higher binding energies is somewhat reminiscent of the correlation-induced energy dependence of SOC previously reported for metallic Sr$_2$RuO$_4$ [30].

In the unoccupied $xz$ and $yz$ states, on the other hand, incoherent features have gained weight in addition to the coherent band dominating the $T=0$ spectrum. They do not follow the two-dimensional dispersion of the coherent band, but are more one dimensional. At high temperature $T=0.35t$, finally, the unoccupied bands show mostly the one-dimensional dispersion characteristic of $xz/yz$ orbitals in the absence of SOC, see Fig. 7(h-i). In the presence of a CF, SOC thus only couples the three orbitals into a 2D dispersion at lower temperatures and lower excitation energies, while spectra at higher temperatures and energies look similar to the case without SOC.

IV. DISCUSSION AND CONCLUSIONS

We have shown that temperature strongly affects the spin-orbital onsite state in the PM Mott insulating state of spin-orbit coupled $t_{2g}$ systems. We have investigated parameter sets supporting excitonic AFM order at low temperatures, with and without a crystal field. As long as the CF is not strong enough to completely quench the orbital degree of freedom, we consistently find a second broad hump in the specific heat, in addition to the peak at $T_N$. In the same temperature range, onsite total angular momentum changes substantially.

In one-particle spectra, low-energy excitations stemming from $xz$ and $yz$ orbitals are two-dimensional in the ground state, but become more one-dimensional at higher temperatures. This can also be interpreted as SOC being most effective at low temperatures. Overall, signatures of SOC and of the temperature-driven spin-orbital rearrangement are rather subtle in one-particle spectra. Even at low temperatures, where SOC is essential do reproduce the dispersion of magnetic excitations [8, 9], one-particle spectra have thus been reasonably well described already without taking SOC into account [21, 29].

However, we argue that X-ray diffraction and absorption experiments performed on Ca$_2$RuO$_4$ show signatures of the spin-orbital rearrangement found here. Parameters for this compound correspond roughly to those of Figs. 7(c,d) and 7(f), i.e. $\Delta \approx 1.5t$ and $\lambda \approx 0.8 - 1t$ [15]. At temperatures of $\approx 260$ K, i.e. between the metal-insulator transition (which goes together with a structural phase transition) and the Néel transition, signatures of another phase transition were reported early on and interpreted in terms of orbital order [16, 17].

Since this additional transition does not break any spatial symmetries, one can rule out orbital stripe [31] or checkerboard [32] patterns theoretically predicted for absent (or weak) SOC. More recent work established that the transition cannot be related to a change in orbital densities, leaving only the phase in a complex orbital superposition as a possibility [33]. This would fit with our findings of an SOC-driven spin-orbital rearrangement. When SOC prefers the $J=0$ state at low temperatures, this implies for each spin projection a specific phase relation between the orbitals. In contrast, no definite phases are expected at higher temperatures where SOC is less active.

We have thus identified the enigmatic orbital-order transition in Ca$_2$RuO$_4$ as a transition to a spin-orbit coupled onsite wave function. This implies, e.g., that a spin up (down) prefers the complex $|F = \pm 1\rangle$ orbital over the opposite state. This is somewhat reminiscent of ferro-orbital order into complex orbitals, which was early on proposed as a scenario for Ca$_2$RuO$_4$ [17]. More generally, complex-orbital order has been suggested to play a role in doped manganites [34] and the Verwey transition of magnetite [35]. Spontaneous complex-orbital order is, however, rare, because lattice distortions like the Jahn-Teller effect favor real orbital states. The present work not only reconciles this picture with the observation of (nearly) constant density on $x_{z\pm}$ orbitals in Ca$_2$RuO$_4$, but moreover shows the transition to arise naturally in a three-orbital model with SOC.
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