Odd-parity triplet superconductivity in multi-orbital materials with strong spin-orbit coupling: applications to doped Sr$_2$IrO$_4$

Zi Yang Meng,$^1$ Yong Baek Kim,$^{1,2,3}$ and Hae-Young Kee$^{1,2}$

$^1$Department of Physics, University of Toronto, Toronto, Ontario M5S 1A7, Canada
$^2$Canadian Institute for Advanced Research/Quantum Materials Program, Toronto, Ontario MSG 1Z8, Canada
$^3$School of Physics, Korea Institute for Advanced Study, Seoul 130-722, Korea

(Dated: October 24, 2014)

We explore possible superconducting states in $t_{2g}$ multi-orbital correlated electron systems with strong spin-orbit coupling (SOC). In order to study such systems in a controlled manner, we employ large-scale dynamical mean-field theory (DMFT) simulations with the hybridization expansion continuous-time Quantum Monte Carlo (CTQMC) impurity solver. To determine the pairing symmetry, we go beyond the local DMFT formalism using parquet equations to introduce the momentum dependence in the two-particle vertex and correlation functions. In the strong SOC limit, a singlet, $d$-wave pairing state in the electron-doped side of the phase diagram is observed at weak Hund’s coupling, which is triggered by antiferromagnetic fluctuations. When the Hund’s coupling is comparable to SOC, a two-fold degenerate, triplet $p$-wave pairing state with relatively high transition temperature emerges in the hole-doped side of the phase diagram, which is associated with enhanced charge fluctuations. Experimental implications to doped Sr$_2$IrO$_4$ are discussed.

PACS numbers: 74.20.-z, 74.20.Rp, 74.70.-b, 71.10.Fd

Introduction. - The investigation of novel electronic states in correlated electron systems with spin-orbit coupling has been a recent subject of intensive research [1]. Early experiments that prompted such activities are the studies of the iridium perovskite oxide Sr$_2$IrO$_4$ [2–10]. Due to strong SOC, the $t_{2g}$ orbitals of Ir$^{4+}$ ions split into $J_{	ext{eff}} = 1/2$ doublet and $J_{	ext{eff}} = 3/2$ quadruplet, leading to a spin-orbit-induced Mott insulator, with a moderate Hubbard interaction $U$. Given the similarity in lattice structure and Mott physics between Sr$_2$IrO$_4$ and La$_2$CuO$_4$, it was proposed that a spin singlet $d$-wave high temperature (high $T_c$) superconductivity emerges in doped iridates [6–11]. If this turns out to be true, it would be a significant progress in decades-long efforts to find high $T_c$ superconductivity in other oxides materials besides cuprates. On the other hand, doped iridates are inherently multi-orbital systems and the analogy to the cuprates may be justified only in the extremely strong SOC limit. The determination of the ground states in such multi-orbital systems is a highly challenging theoretical work when the SOC and some of the multi-orbital interactions such as Hund’s coupling become comparable to each other, which could easily be the case in 4$d$ or 5$d$ electron systems.

In this letter, we provide a theoretical study of possible superconductivity in $t_{2g}$ multi-orbital systems with SOC using the combination of the DMFT with CTQMC impurity solver [12–15] and self-consistent relations between two-particle correlation/vertex functions in parquet equations [16–20]. The DMFT with CTQMC can capture the local correlation effects, but cannot provide the momentum dependence of the vertex functions or two-particle correlation functions, which is necessary for the determination of the dominant pairing channel and other instabilities. A standard way to introduce the momentum dependence is to generalize the single-site effective impurity problem to a finite cluster. While the cluster DMFT has been successful for one-band Hubbard models [21–24], it would be computationally too costly if one applies it to the multi-orbital models with...
agram of Fig. 1, where different interaction channels between vertex and two-particle correlation functions in dependence of necessary vertex functions via the relations as described below, we use the results of the DMFT and an alternative method via the two-particle diagrammatic intra-, inter-orbital interactions and SOC. Here we use numerical method and implications of our results to doped iridates.

Microscopic Model.- The $t_{2g}$ three-orbital Hubbard model on the square lattice is given by, $H = H_{\text{kin}} + H_{\text{SOC}} + H_I$, where $H_{\text{kin}} = \sum_{\mathbf{k}\alpha\sigma} \epsilon_{\alpha}(\mathbf{k}) c_{\mathbf{k}\alpha\sigma}^\dagger c_{\mathbf{k}\alpha\sigma}$ is the electron operator with momentum $\mathbf{k}$, spin $\sigma = \uparrow, \downarrow$ and orbital $\alpha = (d_\alpha, d_\alpha^{\prime}, d_{\alpha z})$. The SOC term is given by $H_{\text{SOC}} = \lambda \sum_{\mathbf{k}\alpha\sigma,\alpha'\sigma'} \langle \alpha|L_{\alpha}|\alpha'\rangle \langle \sigma |S_{\sigma}| \sigma' \rangle c_{\mathbf{k}\alpha\sigma}^\dagger c_{\mathbf{k}\alpha'\sigma'}$, and $L_{\alpha}(S_{\sigma})$ is the orbital (spin) angular momentum operator. The interaction term can be written as

$$H_I = U \sum_{\mathbf{i}\alpha} n_{\mathbf{i}\alpha\uparrow} n_{\mathbf{i}\alpha\downarrow} + \frac{U'}{2} \sum_{\mathbf{i}\alpha\neq\alpha'} n_{\mathbf{i}\alpha\uparrow} n_{\mathbf{i}\alpha'\downarrow}$$

$$+ \frac{J}{2} \sum_{\mathbf{i}\alpha\neq\alpha', \sigma} c_{\mathbf{i}\alpha\sigma}^\dagger c_{\mathbf{i}\alpha'\sigma}^\dagger c_{\mathbf{i}\alpha'\sigma'} c_{\mathbf{i}\alpha\sigma'}$$

$$+ \frac{J'}{2} \sum_{\mathbf{i}\alpha\neq\alpha'} c_{\mathbf{i}\alpha\sigma}^\dagger c_{\mathbf{i}\alpha'\sigma}^\dagger c_{\mathbf{i}\alpha'\sigma'} c_{\mathbf{i}\alpha\sigma'}$$

where $n_{\mathbf{i}\alpha\sigma} = c_{\mathbf{i}\alpha\sigma}^\dagger c_{\mathbf{i}\alpha\sigma}$ and $n_{\mathbf{i}\alpha} = \sum_\sigma n_{\mathbf{i}\alpha\sigma}$. $U'$ and $J'$ denote inter-orbital Hubbard interaction and pairing hopping, respectively. In the atomic limit, these four Kanamori parameters satisfy the relation, $U = U' + J + J'$ and $J = J'$, which is assumed in the following discussions. Thus we explore the phase diagram in terms of $U$ and the Hund’s coupling $J$.

The SOC mixes electron spin and orbital quantum numbers, hence it is useful to first diagonalize the non-interacting Hamiltonian $H_{\text{kin}} + H_{\text{SOC}} = \sum_{\mathbf{k}\alpha\sigma} E_{\mathbf{k}}(\mathbf{k}) a_{\mathbf{k}\alpha\sigma}^\dagger a_{\mathbf{k}\alpha\sigma}$, where $a_{\mathbf{k}\alpha\sigma}^\dagger$ represent the spin-orbit entangled eigenstates characterized by the band index $m = (1, 2, 3)$ and pseudospin $s$ (a Kramers-doublet) with the dispersion $E_{\mathbf{k}}(\mathbf{k})$. We adopt the tight-banding parameters of $\epsilon_{\alpha}(\mathbf{k})$ used in Refs. [26, 27, 32, 33], the nearest-neighbor hopping between $d_{\alpha\alpha'}$ orbitals as the energy unit $t$, and the spin-orbit coupling $\lambda = 2t$. The energy dispersions $E_{\mathbf{k}}(\mathbf{k})$ and the Fermi surface (FS) at filling $n = 5$ are shown in Fig. 2. The $m = 1$ band, mostly made of $J_{\text{eff}} = 1/2$ state, is separated from the other two bands. Near $n = 5$ band filling, only $m = 1$ band crosses the Fermi level so that there is a single electron-like FS, as shown by the red contour line and its projection to the bottom of the Brillouin zone (BZ) in Fig. 2.

Numerical method.- To solve the interacting electron problem, we employ the DMFT with CTQMC impurity solver [21, 22]. This method maps the original, strongly correlated, lattice system into a quantum impurity problem embedded in a self-consistently-determined bath. In this study, we use the hybridization expansion CTQMC impurity solver [12, 14]. It diagonalizes the atomic limit of the interacting problem, and diagrammatically expands the impurity partition function in powers of the hybridization function between the impurity and the bath. Since this algorithm treats the local interactions exactly, it is particularly efficient at moderate and strong interactions. We use about $10^9$ Monte Carlo samples per simulation to obtain converged single-particle results, and another $10^9$ QMC samples to obtain two-particle quantities. The interaction strength is chosen to be close to the bare bandwidth, $U = 12t$ [34], and we can achieve temperatures as low as $T = 0.05t$ ($\beta T = 20$) before a serious minus-sign problem renders the data untrustable.

In order to obtain information about pairing instabilities, one needs to know the momentum and frequency
To introduce the momentum dependence in the vertex functions starting from $G(P)$, $\chi_{ph}^{d/m}(\omega, \omega', \nu)$, and $\Gamma_{ph}^{d/m}(\omega, \omega'', \nu)$, let us turn to the lattice Bethe-Salpeter equation in Fig. 3 (a); $\chi_{ph}^{d/m}(P, P', Q) = \chi_{0}^{ph}(P, Q) + \chi_{0}^{ph}(P, Q) \sum_{P''} \Gamma_{ph}^{d/m}(P, P', Q) \chi_{ph}^{d/m}(P'', Q')$, where $\chi_{0}^{ph}(P, Q)$ can be constructed from single-particle Green’s functions $\chi_{0}^{ph}(P, Q) = -N \Delta G(P) G(P + Q)$ with $N$, the lattice size. We then use $\Gamma_{ph}^{d/m}(\omega, \omega'', \nu)$ (obtained in the DMFT) as an input for $\Gamma_{ph}^{d/m}(P, P', Q)$, and later find the momentum dependence of this and other quantities using an iteration method. Once $\Gamma_{ph}^{d/m}(\omega, \omega'', \nu)$ is used and the sums over $k, k'$ are applied to both sides of the equation, the Bethe-Salpeter equation is reduced to

dependence of the pairing vertex functions. However, in the DMFT simulation, the two-particle correlation functions in the particle-particle (pp) and particle-hole (ph) channels $\chi_{ph/pp}(\omega, \omega', \nu)$ can only be measured at the impurity site, hence only have frequency-dependence. Here we use the parquet equations to introduce momentum-dependence in two-particle quantities as described below. The parquet equations relate the irreducible vertex function in one interaction channel to those in other channels [16]. In our case, we consider four interaction channels: the particle-hole density/magnetic (ph-m), particle-hole single-particle singlet (pp-s) and particle-particle triplet (pp-t) channels [17] [20] [23] [24]. A detailed description of the parquet formalism is given in the Supplemental Material [31] and here we only outline the main idea.

For example, in order to explore the singlet/triplet pair instabilities, we need to find the momentum and frequency dependence of the irreducible vertex functions in pp-s/t channels, $\Gamma_{pp}^{s/t}(P, P', Q)$, with $P \equiv (k, \omega)$, $P' \equiv (k', \omega')$, $Q \equiv (q, \nu)$. In the DMFT-CTQMC, one obtains the lattice single-particle Green’s function $G(P)$ and the ph-d/m two-particle correlation functions $\chi_{ph}^{d/m}(\omega, \omega', \nu)$ measured on the impurity. We first consider the local version of the Bethe-Salpeter equation, $\chi_{ph}^{d/m}(\omega, \omega', \nu) = \chi_{0}^{ph}(\omega, \nu) + \chi_{0}^{ph}(\omega', \nu) \sum_{\omega''} \Gamma_{ph}^{d/m}(\omega, \omega'', \nu) \chi_{ph}^{d/m}(\omega'', \omega', \nu)$. Using $\chi_{ph}^{d/m}(\omega, \omega', \nu)$ obtained in the DMFT, one can extract the local irreducible vertex functions, $\Gamma_{ph}^{d/m}(\omega, \omega', \nu)$.

![FIG. 3. (color online) (a) Bethe-Salpeter equation in the particle-hole density/magnetic channels. $\chi_{ph}^{d/m}$ and $\Gamma_{ph}^{d/m}$ are two-particle correlation and vertex functions, and $\chi_{0}^{ph}$ is the bare two-particle correlation function. (b) Parquet equation for the particle-particle singlet vertex, $\Gamma_{pp}^{s}(P, P', Q)$. It is decomposed into fully irreducible vertex function $\Lambda_{pp}$ and cross-channel contributions from particle-hole density/magnetic vertex ladders $\Phi_{ph}^{s}(P, \omega, Q)$ and the leading interaction in one interaction channel to those in other channels.](attachment:figure3.png)
The leading eigenvector \( \phi(P) \) need to be analyzed. As temperature approaches the transition temperature \( T_c \), \( \lambda \to 1 \), and the corresponding \( \phi(P) \) shows the momentum-dependence of the gap function \([24, 55] \). Similar analysis can be performed in the ph-d/m channels.

**Results and Discussions.**—We compute the LEVs of \( \Gamma^{(1)} \) and \( \Gamma^{(2)} \) for the corresponding vertex functions as a function of temperature \( T \) for singlet/triplet superconductivity, ferromagnetic and antiferromagnetic instabilities across the phase diagram and the leading eigenvector is used to determine the ground state. Fig. 4 shows the results for the parameter set \( U = 12t, U' = 11.6t, J = 0.2t, n = 5.2 \). This is an electron-doped case with a very small Hund’s coupling \( J/U \sim 0.017 \). The main panel shows the LEVs obtained from Eq. 3 using \( \Gamma^{(1)} \) (the results of \( \Gamma^{(2)} \) show the same trend). As temperature decreases, the (pseudospin-)singlet pairing LEV in the \( m = 1 \) band dominates over other channels and the antiferromagnetic channel is the next leading instability. Moreover, the leading eigenvector of the singlet pairing clearly has the \( d_{x^2-y^2} \) momentum-dependence, as shown in the upper inset (the lower inset shows the leading eigenvector of \( \Gamma^{(2)} \) which has the same \( d \)-wave symmetry). In the electron-doped side, both Hund’s coupling and SOC prefer to have \( J_{\text{eff}} = 3/2 \) bands completely filled, and extra electron goes to the initially half-filled \( J_{\text{eff}} = 1/2 \) band. Thus the \( d \)-wave singlet pairing mainly comes from the \( J_{\text{eff}} = 1/2 \) band. Moreover, the corresponding FS is very similar to that of the hole-doped, one-band Hubbard model on square lattice. As shown in the cluster DMFT computations of one-band Hubbard model, the vertex function for the \( d \)-wave superconducting instability is dominated by antiferromagnetic fluctuations \([24] \). Our analysis of the parquet equation shows that the magnetic vertex ladder \( \Phi_{pp'}^n \) at \( q = (\pi, \pi) \) is indeed the dominant contribution to \( \Gamma_{pp'}^{(1)} \).

In turn, the main panel of Fig. 5 shows the LEVs of \( \Gamma^{(1)} \) at a large Hund’s coupling \( J/U \sim 0.17 \) in a hole-doped case, with the parameter set \( U = 12t, U' = 8t, J = 2t, n = 4.9 \). As the Hund’s coupling increases, the (pseudospin-)triplet pairing in the \( m = 1 \) band becomes the leading instability in the hole-doped side while the \( d \)-wave singlet pairing in the electron-doped side is suppressed. The triplet pairing instability found here has two-fold degenerate LEVs and the corresponding leading eigenvectors have \( p'_{x} = -p_{x} - p_{y} \) and \( p'_{y} = -p_{x} + p_{y} \) symmetries. The upper(lower) inset of Fig. 5 shows the leading eigenvector obtained from \( \Gamma^{(1)}(\Gamma^{(2)}) \) with \( p'_{x} \) symmetry. These results imply that the triplet superconductivity is the dominant instability in the hole-doped side. This \( p \)-wave triplet superconductivity emerges from a delicate balance between SOC and Hund’s coupling \([90] \). When holes are introduced, the Hund’s coupling prefers to have holes in \( J_{\text{eff}} = 3/2 \) bands as well as \( J_{\text{eff}} = 1/2 \) band, while the spin-orbit coupling likes to have \( J_{\text{eff}} = 3/2 \) completely filled and to put all extra holes in the \( J_{\text{eff}} = 1/2 \) band. Thus two interactions are not compatible to each other. Only when the SOC and Hund’s coupling are balanced, ferromagnetic fluctuation induced by Hund’s coupling generates the triplet pairing state. If the Hund’s coupling becomes even larger, as shown in the phase diagram (Fig. 1), in the hole-doped side, the system becomes a ferromagnetic metal. Thus we need a significant Hund’s coupling to induce the triplet pairing via ferromagnetic fluctuations, but not-too-large Hund’s coupling which eventually favors a ferromagnetic metal.
The odd-parity triplet pairing is doubly degenerate with components $p'_x$ and $p'_y$, any linear combination of both $p$-wave components is possible below $T_c$. Given that both Ginzburg-Landau theory and BCS-type mean-field approaches favor a fully-gapped superconducting phase that breaks time-reversal symmetry [30], the $p_x + ip_y$ triplet pairing state could be selected. Therefore, our findings may support the chiral $p_x + ip_y$ topological superconducting phase in the hole-doped side of the phase diagram.

It is clear from Fig. 4 and Fig. 5 that the triplet pairing transition temperature in the hole-doped side is higher than the singlet pairing one in the electron-doped side. That is, the triplet LEVs approaches 1 when $T = 0.05t$ whereas the singlet LEV is still below 1 at $T = 0.05t$. The same behaviors also hold for the LEVs obtained from $\Gamma^{(2)}$ analyses. This implies that the $p$-wave superconductivity in the hole-doped side could have relatively higher $T_c$ than the $d$-wave superconductivity in the electron-doped side. Although superconductivity has not been observed in electron-doped Sr$_2$IrO$_4$ [32], our results could stimulate more experimental efforts in the hole-doped side, which may be achieved by substituting Na, K for Sr.

Acknowledgements.- We would like to acknowledge H. Li, Y. L. Wang for discussions on the DMFT+CTQMC simulation code – the iQIST package [35], K. S. Chen, S. X. Yang for their help on the parquet formalism, and Q.-H. Wang for discussions on possible pairing symmetries and their fRG work [27]. This work is supported by the NSERC, CIFAR, and Centre for Quantum Materials at the University of Toronto. Computations were performed on the GPC supercomputer at the SciNet HPC Consortium. SciNet is funded by: the Canada Foundation for Innovation under the auspices of Compute Canada; the Government of Ontario; Ontario Research Fund - Research Excellence; and the University of Toronto.

\[\text{hykee@physics.utoronto.ca}\]

[1] W. Witczak-Krempa, G. Chen, Y. B. Kim, and L. Balents, Annu. Rev. Condens. Matter Phys. 5, 57 (2014).
[2] G. Cao, J. Bolivar, S. McCall, J. E. Crow, and R. P. Guertin, Phys. Rev. B 57, R11039 (1998).
[3] 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, 076402 (2008).
[4] B. J. Kim, H. Ohsumi, T. Komesu, S. Sakai, T. Morita, H. Takagi, and T. Arima, Science 323, 1329 (2009).
[5] S. Fujiyama, H. Ohsumi, T. Komesu, J. Matsuno, B. J. Kim, M. Takata, T. Arima, and H. Takagi, Phys. Rev. Lett. 108, 247212 (2012).
[6] J. Kim, D. Casa, M. H. Upton, T. Gog, Y.-J. Kim, J. F. Mitchell, M. van Veenendaal, M. Daghofer, J. van den Brink, G. Khaliullin, and B. J. Kim, Phys. Rev. Lett. 108, 177003 (2012).
[7] J.-M. Carter, Y. V. Shankar, M. A. Zeb, and H.-Y. Kee, Phys. Rev. B 85, 115105 (2012).
[8] J.-M. Carter, Y. Shankar V., and H.-Y. Kee, Phys. Rev. B 88, 035111 (2013).
[9] P. Ye, S. Chi, S. Chakounamakos, J. A. Fernandez-Baca, T. Qi, and G. Cao, Phys. Rev. B 87, 140406 (2013).
[10] Q. Li, G. Cao, S. Okamoto, J. Yi, W. Lin, B. C. Sales, J. Yan, R. Arita, J. Kunes, A. V. Kochevnikov, A. G. Eguiluz, M. Imada, Z. Gai, M. Pan, and D. G. Mandler, Sci. Rep. 3, 3073 (2013).
[11] F. Wang and T. Senthil, Phys. Rev. Lett. 106, 136402 (2011).
[12] P. Werner and A. J. Millis, Phys. Rev. B 74, 155107 (2006).
[13] K. Haule, Phys. Rev. B 75, 155113 (2007).
[14] L. Huang and X. Dai, arXiv:1205.1708 (2012).
[15] L. Du, L. Huang, and X. Dai, Eur. Phys. J. B 86, 94 (2013).
[16] C. d. Dominicis and P. C. Martin, J. Math. Phys. 5, 14 (1964); 5, 31 (1964).
[17] N. E. Bickers and S. R. White, Phys. Rev. B 43, 8044 (1991).
[18] S. X. Yang, H. Fotso, J. Liu, T. A. Maier, K. Tomko, E. F. D’Azevedo, R. T. Scalettar, T. Pruschke, and M. Jarrell, Phys. Rev. E 80, 046706 (2009).
[19] G. Rohringer, A. Valli, and A. Toschi, Phys. Rev. B 86, 125114 (2012).
[20] K.-M. Tam, H. Fotso, S.-X. Yang, T.-W. Lee, J. Moreno, J. Ramamujam, and M. Jarrell, Phys. Rev. E 87, 013311 (2013).
[21] T. Maier, M. Jarrell, T. Pruschke, and M. H. Hettler, Rev. Mod. Phys. 77, 1027 (2005).
[22] E. Gull, A. J.Millis, A. I. Lichtenstein, A. N. Rubtsov, M. Troyer, and P. Werner, Rev. Mod. Phys. 83, 349 (2011).
[23] K. S. Chen, Z. Y. Meng, U. Yu, S. Yang, M. Jarrell, and J. Moreno, Phys. Rev. B 88, 041103 (2013).
[24] K.-S. Chen, Z. Y. Meng, S.-X. Yang, T. Pruschke, J. Moreno, and M. Jarrell, Phys. Rev. B 88, 245110 (2013).
[25] R. Arita, J. Kunes, A. V. Kochevnikov, A. G. Eguiluz, and M. Imada, Phys. Rev. Lett. 108, 086403 (2012).
[26] H. Watanabe, T. Shirakawa, and S. Yunoki, Phys. Rev. Lett. 110, 027002 (2013).
[27] Y. Yang, W.-S. Wang, J.-G. Liu, H. Chen, J.-H. Dai, and Q.-H. Wang, Phys. Rev. B 89, 094518 (2014).
[28] J. E. Han, Phys. Rev. B 70, 054513 (2004).
[29] S. Sakai, R. Arita, and H. Aoki, Phys. Rev. B 70, 172504 (2004).
[30] C. M. Puetter and H.-Y. Kee, EPL 98, 27010 (2012).
[31] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.177003, which includes Refs. 2–8, 17–20, 26, 27, 32, and 33 for details about the $t_2g$ three-orbital model, the CTQMC/DMFT+Parquet formalism and its numerical implementation and performance..
[32] H. Watanabe, T. Shirakawa, and S. Yunoki, Phys. Rev. Lett. 105, 216410 (2010).
[33] H. Watanabe, T. Shirakawa, and S. Yunoki, Phys. Rev. B 89, 165115 (2014).
[34] Similar interaction parameters are used in Ref. 25, 26, but phases at finite $J$ in the hole-doped side are not stud-
[35] J. R. Schrieffer, *Theory of Superconductivity* (ABC, Westview Press, 1983).

[36] M. Sigrist and K. Ueda, *Rev. Mod. Phys.* **63**, 239 (1991).

[37] O. B. Korneta, T. Qi, S. Chikara, S. Parkin, L. E. De Long, P. Schlottmann, and G. Cao, *Phys. Rev. B* **82**, 115117 (2010).

[38] L. Huang, Y. Wang, Z. Y. Meng, L. Du, P. Werner, and X. Dai, arXiv:1409.7573 (2014).