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Possible superconductivity with Bogoliubov Fermi surface in lightly doped Kagome U(1) spin liquid

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Whether the doped t-J model on the Kagome lattice supports exotic superconductivity has not been decisively answered. In this paper, we propose a new class of variational states for this model and perform large-scale variational Monte Carlo simulation on it. The proposed variational states are parameterized by the SU(2)-gauge-rotation angles, as the SU(2)-gauge structure hidden in the Gutzwiller-projected mean-field ansatz for the undoped model is broken upon doping. These variational doped states smoothly connect to the previously studied U(1) π-flux or 0-flux states, and energy minimization among them yields a chiral noncentrosymmetric nematic superconducting state with 2×2-enlarged unit cell. Moreover, this pair density wave state possesses a finite Fermi surface for the Bogoliubov quasi particles. We further study experimentally relevant properties of this intriguing pairing state.

Introduction: Quantum spin liquids (QSL) have attracted increasing interest in condensed matter physics in the past decades [1–6]. They represent an exotic class of insulating states which cannot be adiabatically connected into a trivial band insulator. Moreover, a QSL state can support fractionalized excitations with fractional braiding statistics. One of the most intriguing aspects of QSL lies in that doping a QSL might naturally lead to high temperature superconductivity[7–16] or a topologically ordered Fermi liquid state (FL*)[17–19].

One promising model exhibiting a QSL ground state is the spin-1/2 Heisenberg model on the Kagome lattice, which is probably realized by the spin-liquid candidate material Herbertsmithite[3]. Numerous efforts have been devoted to study properties of this model for several decades. Except for a few early results pointing toward the valence bond solid (VBS) state[20–22], dominating numerical results suggest a QSL ground state for this model[23–35]. Particularly, while a number of density-matrix renormalization group (DMRG) simulations on wide cylinders have exhibited evidences of a Z_2 QSL with exponentially decaying spin-spin correlation[23–28], recent iDMRG simulation on infinite cylinders[29], tensor-network simulation on infinite system[30], and variational Monte Carlo (VMC) studies[31–33] suggest that the ground state is a gapless U(1) Dirac QSL with algebraic correlation. While further studies are still needed to reveal the precise nature of the ground state at half filling, it is also desired to study what quantum state would be obtained when mobile charge carriers are introduced into it by doping. Especially, can exotic superconductivity emerge upon doping the Kagome QSL state?

The nature of the lightly doped Kagome system described by the t-J model is not decisively known so far. Nonetheless, recent DMRG study on the model with moderate doping on the 4-leg cylinder provided convincing evidences of an insulating holon Wigner crystal[36]. On the wider system, previous VMC investigation of this model on up to $8^2 \times 9$ lattice in certain doping range suggests that the π-flux Dirac U(1) spin liquid[31] is unstable against a 0-flux state with a VBC ordering[37, 38]. As the π-flux state has lower energy than the 0-flux state at half filling, it is obvious that the 0-flux state obtained by VMC at certain doping range cannot be continuously connected to the undoped π-flux QSL state[31]. It is natural to ask what is the ground state for the lightly doped t-J model on the Kagome lattice assuming that the ground state of the undoped system is a U(1) Dirac QSL.

In this paper, we study the t-J model on the Kagome lattice in the very low doping regime which is expected to smoothly connect with U(1) spin liquid at half-filling[31] by performing VMC simulations. Our study is inspired by a crucial SU(2)-gauge structure[39–41] hidden in the projective construction at half-filling: two different mean-field (MF) ansatzs related by an arbitrary local SU(2)-gauge rotation actually correspond to the same physical spin state after the Gutzwiller-projection. Such gauge-redundancy leads to a many-to-one labeling between the mean-field ansatzs and the projected wave function at half-filling[42]. At finite doping, the breaking of this gauge structure differentiates the many states related by the gauge-rotation, which form our variational groups. We choose the doped 0-flux or π-flux states as our un-rotated starting points. Energy minimizations within both groups of variational states yield chiral noncentrosymmetric nematic superconducting states with 2×2-enlarged unit cell in the very low doping regime, with the gauge-rotated π-flux state smoothly connecting to the undoped π-flux QSL[31]. Remarkably, as the SU(2)-gauge rotation maintains the quasi-particle spectrum, the obtained superconducting states possess finite Fermi surface (FS) for the Bogoliubov quasi-particles. The physical properties of these pairing states are intriguing: although they are superconducting states, they resemble those of the normal FL in many aspects.

Variational states: We study the standard t-J model on the Kagome lattice illustrated in Fig. 1(a):

$$H = -t \sum_{\langle ij \rangle \sigma} P_G (c^\dagger_{i\sigma} c_{j\sigma} + h.c.) P_G + J \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j), \tag{1}$$

where $c_{i\sigma}$ annihilates an electron on site i with spin σ, $\mathbf{S}_i = \frac{1}{2} c^\dagger_{i\alpha} \sigma_{\alpha\beta} c_{i\beta}$ denotes the spin operator and $n_i = \sum_\sigma c^\dagger_{i\sigma} c_{i\sigma}$ is the density operator. $P_G = \prod_i (1 - n_{i\uparrow} n_{i\downarrow})$ is the Gutzwiller-projection operator enforcing no-double-occupancy constraint. $\langle ij \rangle$ represents nearest-neighbor (NN) bonding. Here we set $J = 1$ as the energy scale. The parameter t and the doping concentration δ are set as tuning parameters spanning the phase diagram.

To smoothly connect with the previously studied π-flux state at half-filling[31] and to compare energy with the zero-flux state at finite doping[37, 38], we investigate the Gutzwiller-projected MF states generated by the following MF Hamiltonian,

$$H^0_{MF} = \sum_{\langle ij \rangle \sigma} \chi_{ij} c^\dagger_{i\sigma} c_{j\sigma} + h.c., \tag{2}$$

where $\chi_{ij} = \pm 1$. These states can be characterized by the fluxes $e^{i\phi} = \prod_{\text{plaquette}} \text{sgn}(\chi_{ij})$ through triangle and hexagon plaquettes of the Kagome lattice. In this work, we primarily focus on two types of fluxes: (1) the 0-flux states having zero flux through all the triangles and hexagons shown in Fig. 1(b); (2) the π-flux state having π flux through the hexagons and zero flux through the triangles as shown in Fig. 1(c). At half filling, both flux states after the projection are QSL. While the former has a large spinon FS, the latter is a U(1) Dirac QSL. Previous VMC studies[31] showed that the π-flux state has the lowest energy among all studied states.

The key point lying behind the present work is the following SU(2)-gauge structure hidden in the projective construction at half-filling[40, 41]. Let's perform the following local SU(2)-gauge transformation W_i on the two component spinor $\psi_i = (c_{i\uparrow}, c^\dagger_{i\downarrow})^T$,

$$\begin{bmatrix} c_{i\uparrow} \\ c^\dagger_{i\downarrow} \end{bmatrix} \to W_i \begin{bmatrix} c_{i\uparrow} \\ c^\dagger_{i\downarrow} \end{bmatrix}. \tag{3}$$

At half-filling, any two MF ansatzs connected by this local SU(2)-gauge rotation label the same physical spin state after projected into the single-occupance subspace,

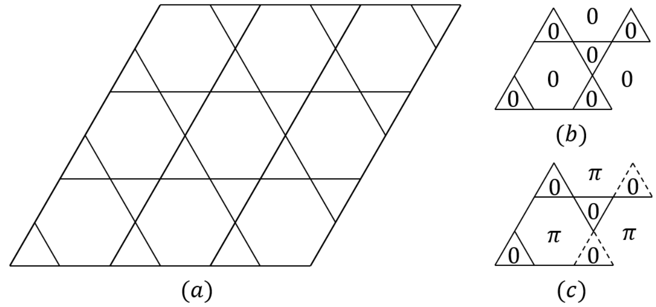

FIG. 1. (a) A schematic representation of the Kagome lattice. (b) The 0-flux state with $\chi_{ij} = 1$ on each bond. (c) The π-flux state with zero flux through triangles and π-flux through hexagonals. Dashed lines indicate the $\chi = -1$ bonds.

as the spin operator $\mathbf{S}_i$ keeps invariant under this gauge transformation[40, 41]. However, this many-to-one labeling is absent once the system is doped away from half filling. Consequently, the many states related by the gauge rotation before projection can represent physical states with distinct physical properties at finite doping. One may naturally raise the following question: what is the lowest-energy state among all those gauge-rotated π- or 0-flux states for the system with very low doping? To answer this question, we choose the local SU(2)-gauge rotation angles as variational parameters, from which we construct MF Hamiltonian to generate the variational physical states by projection, for energy minimization in both flux sectors.

Our trial wave functions are generated by the following local SU(2)-gauge-rotated Bogoliubov-de Genes (BdG) MF Hamiltonian,

$$H_{MF} = \sum_{ij} \begin{bmatrix} c^\dagger_{i\uparrow} & c_{i\downarrow} \end{bmatrix} W_i \begin{bmatrix} \chi_{ij} & 0 \\ 0 & -\chi_{ji} \end{bmatrix} W^\dagger_j \begin{bmatrix} c_{j\uparrow} \\ c^\dagger_{j\downarrow} \end{bmatrix}. \tag{4}$$

Here the unrotated MF parameter χ_{ij} on the NN-bond $\langle ij \rangle$ for the π- and 0-flux states have been introduced above. We set the on-site term χ_{ii} to a uniform value $\chi_{ii} = \chi_0$ as the chemical potential term. The local SU(2) rotation matrix W_i can be parameterized by the following three rotation angles α_i, β_i and γ_i as

$$W_i = \begin{bmatrix} e^{i\beta_i} \cos\alpha_i & e^{i\gamma_i} \sin\alpha_i \\ -e^{-i\gamma_i} \sin\alpha_i & e^{-i\beta_i} \cos\alpha_i \end{bmatrix}. \tag{5}$$

Our trial wave function $P_G |\Psi_{\text{MF}}\{\chi_0, \alpha, \beta, \gamma\}\rangle$ now depends on the set of variational parameters $\{\alpha_i, \beta_i, \gamma_i\}_{i=1,\cdots,N}$ and χ_0. Here $|\Psi_{\text{MF}}\{\chi_0, \alpha, \beta, \gamma\}\rangle$ is the MF ground state of Eq. (4).

VMC results: We adopt standard Monte Carlo approach to simulate the variational states $P_G |\Psi_{\text{MF}}\{\chi_0, \alpha, \beta, \gamma\}\rangle$ on the Kagome lattice with size $3 \times L \times L$ and periodic boundary condition, where the two adopted lattice sizes $L = 8$ and $L = 12$ lead to consistent results. The numerical complexity arising from

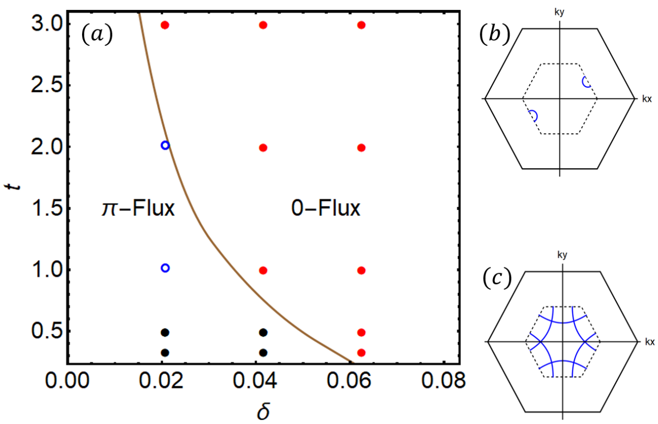

FIG. 2. (a) phase diagram of the slightly doped t-J model on a $8\times8\times3$ lattice. The black circles in the π-flux sector represent metallic phase without pairing. (b) Nearly doubly degenerate small FSs of the slightly doped π-flux state located around the two folded Dirac points of undoped state. (c) Folded FSs of the doped 0-flux state.

		$\delta=0.92\%$	$\delta=1.85\%$	$\delta=2.78\%$
t=2	0-flux	-0.96037(2)	-1.00873(3)	-1.05669(3)
	π-flux	-0.97105(5)	-1.01197(2)	-1.05238(3)
	Z_2 QSL	-0.97106(2)	-1.01196(4)	-1.05231(4)
	VBC-D	-0.96066(3)	-1.00921(2)	-1.05710(3)
	CDW	-0.9112(3)	/	/
t=1	0-flux	-0.92894(3)	-0.94680(1)	-0.96408(2)
	π-flux	-0.94347(2)	-0.95691(2)	-0.97010(4)
	Z_2 QSL	-0.94348(3)	-0.95686(4)	-0.97010(3)
	VBC-D	-0.92933(2)	-0.94698(2)	-0.96442(2)
	CDW	-0.9104(4)	/	/
t=0.5	0-flux	-0.91336(4)	-0.91565(3)	-0.91772(3)
	π-flux	-0.92967(3)	-0.92939(2)	-0.92828(2)
	Z_2 QSL	-0.92965(2)	-0.92936(3)	-0.92827(3)
	VBC-D	-0.91367(2)	-0.91588(3)	-0.91808(2)
	CDW	-0.9154(2)	/	/

TABLE I. Optimized energy of part of the candidates on the model with $t=0.5\sim2$ and $\delta=0.92\%\sim2.78\%$ on a $3\times12\times12$ lattice. A complete table with more candidate ansatz can be found in SM.

optimizing a large number of variational parameters is overcome by the stochastic reconfiguration (SR) method [43]. We further reduce the number of SU(2) rotation angles by restricting the parameters in the super-cell with size $3\times2\times2$. We have checked that increasing the size of the super-cell does not lead to a lower optimized energy (See Supplemental material (SM) for detail).

Our main results are summarized in the phase diagram shown in Fig. 2(a), where we consider several t ranging from 1/3 to 3 and several doping levels below 7% on the Kagome lattice with $L=8$. Starting from the undoped π-flux state, the lowest-energy state stays in the π-flux sector until beat by the optimized states in 0-flux sector at a finite doping concentration δ_c depending on t. For small $t\sim1/3$, the gauge-rotated π-flux state is stable until the doping concentration reaches $\delta_c\sim5\%$. While for large t, a smaller doping is enough to drive the system away from the π-flux sector, consistent with previous VMC studies at $J=0.4t$ [37, 38]. To explore the possible finite size effect, we also studied the models on $L=12$ lattice with 4 to 12 doped holes and find that the gauge-rotated π-flux state is still the lowest-energy state for most of the cases at small doping region.

The physical properties of the gauge-rotated π-flux phase are mainly determined by the optimized SU(2) rotation angles, which are provided in the SM[44]. Except for the two parameter points in the small J and δ region of the π-flux sector (black circles in Fig. 2), we find that the optimized angle α_i for both flux sectors are neither 0 nor π. Consequently, the non-zero off-diagonal terms in the gauge-rotation matrices W_i defined in Eq. (5) bring about a singlet pairing term $H_\Delta=-\sum_{ij}c^\dagger_{i\uparrow}c^\dagger_{j\downarrow}\left[\chi_{ij}e^{i(\beta_i+\gamma_j)}\cos\alpha_i\sin\alpha_j+(i\rightleftarrows j)\right]+h.c.$ in H_{MF}. Note that the gauge rotation (3) as a unitary transformation does not change the quasi-particle spectra [40, 41], but it only leads to enlargement of the unit cell. As a result, the superconducting states generated here will have quasi-particle FSs simply folded from those of the doped 0- or π-flux states before the gauge rotation, as shown in Fig. 2(b) and (c). Therefore, we have obtained here singlet pairing states with finite Bogoliubov FS. Such SC states breaking translational symmetry with finite FS were pair-density-wave states[45–58].

The optimized gauge-rotation angles in the π-flux sector are complicated because all the $\{\alpha_i,\beta_i,\gamma_i\}$ within the super cell are non-zero and non-uniform, breaking the TRS, the lattice-rotation, the inversion and the translational symmetries. The pairing and hopping terms generated by the gauge rotations are generally complex and are of the same order of magnitude, which suggests a typical inter-band pairing state. More details of the optimized gauge-rotation angles and the resulting gauge-rotated MF Hamiltonian are provided in the SM. In spite of the complicated pairing and hopping terms, the resulting MF Hamiltonian exhibits finite Bogoliubov FS shown in Fig. 2(b), which comprises two nearly doubly degenerate small pockets folded from those of the un-rotated π-flux state.

At infinitesimal doping, the gauge-rotated π-flux state is reasonably the lowest-energy VMC state due to the finite energy difference between this state and other states presented in the previous VMC study of the undoped case. When the doping concentration becomes larger, besides the gauge-rotated π- or 0- flux states, other competitive states such as the holon Wigner crystal[36], the doped Z_2 QSL[34], various types of VBC states[37, 38], and the uniform-pairing states[7, 59, 60] should also be considered in the VMC calculations. In Table I, we list the optimized energies for part of the lowest-energy states we obtained on the $L=12$ lattice, which suggests that

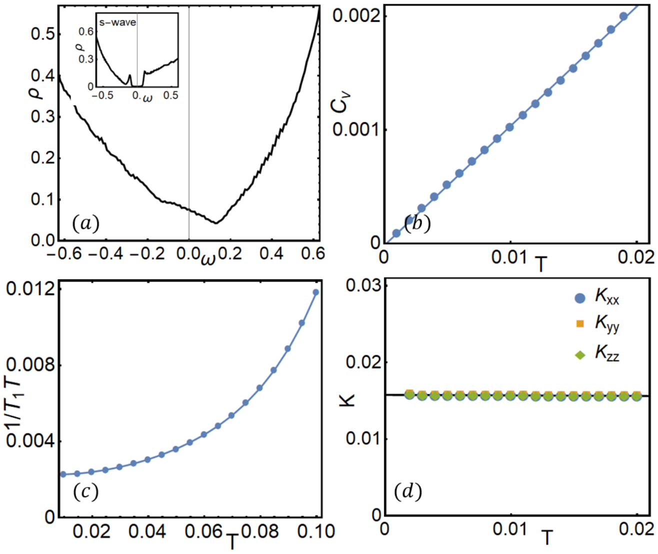

FIG. 3. Experiment-relevant quantities for the optimized gauge-rotated π-flux state. (a) $dI/dV \sim V$ curve for the STM. The inset is the dI/dV curve for the model with uniform on-site s-wave (b) the specific heat $C_v \sim T$. (c) the NMR relaxation rate $1/T_1T$. (d) the NMR Knight-shift K as function of T, three colors stand for K_{xx}, K_{yy} and K_{zz} respectively. The optimal gauge-rotation angles are obtained from parameter setting $t = 0.5$ and $\delta = 2.08\%$.

in the small doping region the gauge-rotated π-flux state has lower energy than the other VMC candidates. We can see the doped Z_2 QSL [34] provides similar energy as the gauge-rotated π-flux state because after optimization such state actually flows back to the U(1) Dirac spin liquid (π-flux state) for all the cases we studied. In the 0-flux sector, we find that the D-type VBC state has slightly lower energy than the gauge-rotated 0-flux state. Another important candidate, the holon Wigner crystal, is mimicked by the CDW ansatz in the VMC calculation. Restricted by the finite lattice size, we only consider the four-hole doped $L = 12$ system with $3 \times 6 \times 6$ supercell. Though we observe the similar density distribution as the Wigner crystal, the VMC energy of this CDW state is higher than the gauge-rotated π-flux state. We also consider the uniform-pairing states with both the extended s-wave and d-wave pairing parameters lived on the nearest and second nearest neighbor bonds, which also provide higher energies in the small doping region. A more complete comparison of all the competing states we considered on the $L = 12$ lattice and the detailed VMC realization of them are presented in the SM[44].

Singlet pairing with finite FS: The singlet pairing with Bogoliubov FS obtained here is distinct from conventional superconductors. To reveal the physical properties of this intriguing pairing state relevant to experiments, we shall perform MF studies below toward the zero- and finite-temperature behaviors of the system represented by the optimized H_{MF}. Consequently, this pairing state is found to be very exotic.

On one hand, the breaking of U(1)-gauge symmetry leads to finite superfluid density as expected (see SM[44] for details), which will result in detectable Meissner effect. On the other hand, the presence of the full FS causes finite density of state (DOS) which, in combination with the singlet-pairing signature, makes this pairing state look like a normal FL in the aspects of low lying quasi-particle and spin excitations, as shown in Fig. 3 for the gauge-rotated π-flux state. In the zero-temperature dI/dV curve for the STM spectrum shown in Fig. 3(a), a finite zero-bias conductance appears caused by the finite DOS, in comparison with the U-shaped curve for the s-wave SC shown in the inset. Fig. 3(b) shows that the specific-heat $C_v \propto T$ at $T \to 0$, resembling the normal FL. Fig. 3(c) illustrates that the relaxation rate $1/T_1T$ of the nuclear magnetic resonance (NMR) saturates to a finite value at $T \to 0$, obeying a Korringa-law-like behavior for the FL, different from the $1/T_1T \to 0$ behavior for conventional fully-gapped ($\propto e^{-\Delta/T}$) or nodal ($\propto T^3$) SC. Fig. 3(d) exhibits that the NMR Knight-shift K saturates to a finite value for $T \to 0$, independent of the orientation of the exerted magnetic field, similarly to the Pauli-susceptibility behavior for standard FL. This behavior is distinct from the $K \to 0$ behavior of conventional singlet SC with full or nodal gap or the obvious magnetic-field-orientation-dependence of K for the triplet SC. Although both the gauge-rotated π- or 0- flux states exhibit BFS, the different doping dependences of the area enclosed by their FSs can be distinguished by the ARPES, which can also lead to different behaviors such as the doping dependence of C_v/T. Details of these MF studies are provided in the SM[44].

Discussion and Conclusion: Note that, starting with a U(1) QSL at half-filling, we have only considered the gauge-rotation angles as variational parameters and neglect the amplitude fluctuation of χ_{ij} before the gauge rotation. Such a treatment is reasonable only at zero-doping limit. For higher dopings, lower variational energy is generally expected if we include the variation of the amplitude of χ_{ij}. The band structure of such improved state can be strongly modified, i.e. Hastings-type VBC order can gap out the Dirac points[61]. We have briefly investigated the fate of Hastings-type VBC in the unrotated π flux state, and found that it becomes visible when the doping concentration is larger than $\delta_c \sim 4\%$. Therefore, close to the zero-doping limit, the Bogoliubov FS is more likely to survive.

Previous studies[63–67] have shown the survival of the FS under the Gutzwiller projection, although some other MF properties might be modified[68], such as the quasiparticle weight. Similar phenomenon, namely the survival of the FS under Gutzwiller projection, is also directly observed for our projected gauge-rotated states by numerically detecting the FS-jump in the occupation-number distribution of Bogoliubov quasiparticles in the momentum space (see the SM for details[44]). Another concern about the stability of the Bogoliubov FS obtained here under possible remnant interactions among the Bogoliubov quasi-particles neglected in the VMC treatment. Indeed the FSs shown in Fig. 2(b) and (c) satisfy the relation $\varepsilon_{\mathbf{k}} = \varepsilon_{-\mathbf{k}}$ as the unitary SU(2)-gauge rotation adopted here maintains the quasi-particle energy, which will suffer from the Cooper instability under remnant interactions. However, note that the two superconducting states obtained here break both the TRS and the inversion symmetry[44]. Without the protection of these two symmetries [62], the relation $\varepsilon_{\mathbf{k}} = \varepsilon_{-\mathbf{k}}$ cannot survive such perturbations as the further variations of $\{\chi_{ij}, \Delta_{ij}\}$ after the gauge rotation, which can always exist for finite doping. Consequently, the Bogoliubov FSs obtained here should be stable against weak remnant interactions among the quasi-particles.

Evidences of SC with Bogoliubov FS can also appear in other contexts such as the FFLO state induced in the magnetic field[69, 70], the cubic system with $j = 3/2$ total-angular-momentum degree of freedom[71] and some iron-based superconductors with spin-orbit coupling and interband pairing[72]. The recently synthesized YPtBi multi-band superconductor with strong spin-orbit-coupling[73, 74] might also exhibits Bogoliubov FS if it breaks the TRS[75]. While these systems host similar normal FL-like quasi-particle excitations as here, their spin excitations have different properties from those of the singlet pairing state obtained here. In summary, we propose a new way to obtain the Bogoliubov FS: doping a U(1) QSL. The key point lies in that the local SU(2)-gauge rotation, which brings about SC to the doped QSL, will not alert the quasi-particle energy, which is different from doping a QSL with spinon FS [76]. Such mechanism not only applies to the doped Kagome U(1) QSL, but also applies to other doped U(1) QSL, which could be a promising way to obtain the new type of unconventional gapless SC in strongly-correlated electronic systems.

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