

## Triplet FFLO superconductivity in the doped Kitaev-Heisenberg honeycomb model

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We provide analytical and numerical evidence of spin-triplet Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconductivity in the itinerant Kitaev-Heisenberg model (antiferromagnetic Kitaev coupling and ferromagnetic Heisenberg coupling) on the honeycomb lattice around quarter filling. The strong spin-orbit coupling in our model leads to the emergence of six inversion symmetry centers for the Fermi surface at nonzero momenta in the first Brillouin zone. We show how the Cooper pairs condense into these nontrivial momenta, causing spatial modulation of the superconducting order parameter. Applying a Ginzburg-Landau expansion analysis, we find that the superconductivity has three separated degenerate ground states with three different spin-triplet pairings. Exact diagonalizations on finite clusters support this picture while ruling out a spin (charge) density wave.

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*Introduction.* Mott insulators and high- $T_c$  superconductors are closely related since the latter can be obtained from doping the half-filled Mott insulator [1–5]. One key element in superconductivity is the emergence of off-diagonal long-range order, which results in a Bardeen-Cooper-Schrieffer ground state where Cooper pairs have a zero net momentum. The  $\eta$  pairing, proposed by Yang [6], binds electrons with momenta  $\mathbf{k}$  and  $\pi - \mathbf{k}$ , and therefore involves a superconductivity with a nonzero Cooper pair momentum. This superconductivity is referred to as Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconductivity [7,8]. FFLO superconductivity, which supports spatial modulation for the electron pairing due to the nontrivial Cooper pair momentum, was first proposed in the 1960's in a system with a significant Zeeman interaction, which shifts the Fermi surfaces for the up and down spins. Experimental realizations of FFLO superconductivity have been proposed, for example, in heavy fermions [9], ultracold atom systems [10–16], BEC analogs [17], and in magnetic analog materials [18–27]. However, this exotic phase of matter has been observed only in a small number of systems so far [9,16]. Indeed, the large magnetic field has a strong pair-breaking effect and limits the stability region of the FFLO phase. Models without explicit time-reversal symmetry breaking have been considered in the context of superfluid  $^3\text{He}$  [28] and unconventional superconducting [29] films. Here, we propose a theoretical model where the time-reversal symmetry is not explicitly broken and is purely two dimensional (as opposed to Refs. [28,29]). Thus our approach is suitable for the realization of FFLO superconductivity in the context of two-dimensional “iridate” materials.

Lately, the studies of iridates, a family of materials with significant spin-orbit coupling, have aroused great interest [30–32] partly because of the emergence of topological Mott physics [33] and its connection to the Kitaev anyon model [34,35]. It has been shown both theoretically and experimentally that the existence of zigzag-magnetic order

results from a Kitaev-Heisenberg magnetic coupling in the two-dimensional sodium iridate family [36–40]. An additional symmetric-off-diagonal exchange term can also be added in the analysis [41]. Doping these spin-orbit Mott insulators has been addressed theoretically [42–44] and has started to attract some experimental attention [45]. Here, we address superconductivity in the presence of a large Hubbard interaction and adopt a localized magnetism point of view where the Kitaev-Heisenberg spin Hamiltonian originates from superexchange processes [46]. Such a magnetic system with spin-orbit coupling and Kitaev-Heisenberg physics can also be realized in cold atom systems [47–51]. Using both analytical and numerical methods, we provide convincing evidence of a spin-triplet FFLO superconductor owing to spin-orbit coupling close to quarter filling without breaking the time-reversal symmetry. This provides an exotic scenario to reach a spin-triplet FFLO superconductor without breaking time-reversal symmetry with applications in quantum materials and ultracold atoms.

Before showing detailed derivations, we summarize the main points. Kitaev-Heisenberg coupling entails spin-triplet pairing that engenders spinor condensates [52–54] in momentum space. One important ingredient here is the appearance of six inversion symmetry centers for the Fermi surface at nonzero momenta in the first Brillouin zone. This will allow the Cooper pairs with triplet pairing to condense at nontrivial momenta. In Fig. 1, we show the band structure of the spin-orbit coupling model and the symmetry centers of the Fermi surface. Electron pairs around these symmetry centers with nontrivial momenta  $\mathbf{q}_\alpha$  form spin-triplet pairs with Cooper pair momenta  $\mathbf{Q}_\alpha = 2\mathbf{q}_\alpha$ . We shall study the superconductivity by calculating the Cooper pairs' response in the Ginzburg-Landau theory for both spin-triplet and spin-singlet pairing. We provide compelling evidence of a triplet FFLO superconductor through a Ginzburg-Landau expansion and an exact diagonalization analysis.

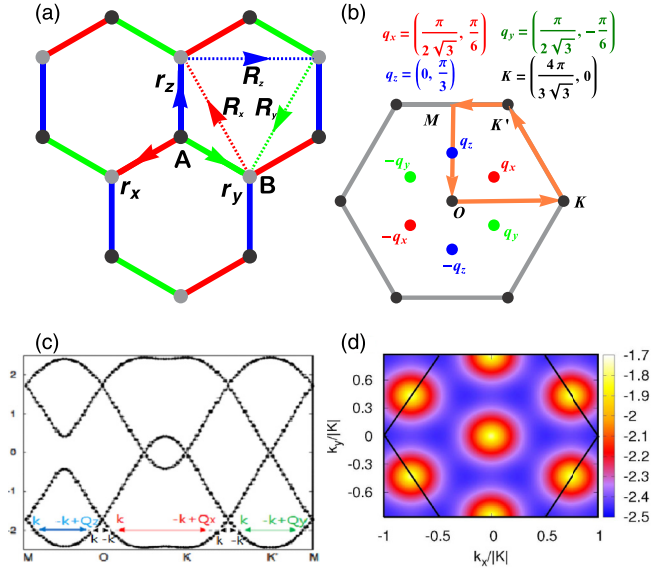


FIG. 1. (a) The Kitaev-Heisenberg model on the honeycomb lattice: In Eq. (1),  $\alpha$  denotes respectively  $x$  on the red links,  $y$  on the green links, and  $z$  on the blue links, each of them corresponding to  $\mathbf{r}_x = (-\frac{\sqrt{3}}{2}, -\frac{1}{2})$ ,  $\mathbf{r}_y = (\frac{\sqrt{3}}{2}, -\frac{1}{2})$ , and  $\mathbf{r}_z = (0, 1)$ ; the lattice vectors are  $\mathbf{R}_x = (-\frac{\sqrt{3}}{2}, \frac{3}{2})$ ,  $\mathbf{R}_y = (-\frac{\sqrt{3}}{2}, -\frac{3}{2})$ , and  $\mathbf{R}_z = (\sqrt{3}, 0)$ . We have taken the lattice spacing to be 1. (b) The first Brillouin zone, in which, apart from the center of the FBZ, there are six additional centers of inversion symmetry for the Fermi surface of the tight-binding part. (c) The band structure of the spin-orbit model ( $t = 0$ ,  $t' = 1$ ).  $M$ ,  $O$ ,  $K$ ,  $K'$  are denoted in (b). When the chemical potential is fixed, electrons on the Fermi surface form triplet Cooper pairs with nontrivial momentum  $\mathbf{Q}_x$ ,  $\mathbf{Q}_y$ , and  $\mathbf{Q}_z$ .  $\mathbf{Q}_\alpha = 2\mathbf{q}_\alpha$ . (d) Energy color plots for the lowest band in units of  $t' = 1$ .

*Model Hamiltonian.* For the doped Kitaev-Heisenberg model, we consider the following Hamiltonian on the honeycomb lattice,

$$\begin{aligned}
 H &= H_0 + H_J, \\
 H_0 &= - \sum_{(i,j)} P_i [t c_{i\sigma}^\dagger d_{j\sigma} + t' c_{i\sigma}^\dagger d_{j\sigma'} \tau_{\sigma\sigma'}^\alpha + \text{H.c.}] P_j, \\
 H_J &= J_1 \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{(i,j)} [S_i^\alpha S_j^\alpha - S_i^\beta S_j^\beta - S_i^\gamma S_j^\gamma], \quad (1)
 \end{aligned}$$

where  $i$  and  $j$  refer to the site index, and  $c_{i\sigma}$  and  $d_{j\sigma}$  to electron operators on the lattices A and B in Fig. 1(a).  $\sigma$  and  $\sigma'$  are the spins of the electrons and  $\tau$  the Pauli matrix with  $\alpha = x, y, z$ , respectively, for red, green, and blue links ( $\mathbf{r}_i - \mathbf{r}_j = \mathbf{r}_\alpha$ ) and  $\beta, \gamma$  take components other than  $\alpha$  [see Fig. 1(a)]. We note the Gutzwiller projectors as  $P_i = (1 - \sum_\sigma c_{i\sigma}^\dagger c_{i\sigma})$  or  $P_j = (1 - \sum_\sigma d_{j\sigma}^\dagger d_{j\sigma})$  according to the sublattice [55–57]. The filling factor  $n$  and the doping level  $\delta$  are connected by the relation  $n = \frac{1}{2} - \delta$ . In contrast to previous analyses [42,43], we include a spin-orbit term of the (doped) model [46], such that the antiferromagnetic Kitaev and ferromagnetic Heisenberg couplings at half filling are microscopically obtained from second-order superexchange processes:  $J_1 = \frac{4t^2}{U}$ ,  $J_2 = \frac{4t'^2}{U}$  with  $U$  the Hubbard interaction. Due to the sign conventions in Eq. (1), positive  $J_2$  values favor ferromagnetic correlations.

The singlet component would rather involve small- $Q$  wave vectors. Setting  $J = J_1 - J_2$  and  $K = J_2$ , we recover the model used in Ref. [36] describing the half-filled system. One shall assume that  $t'$  is real to avoid an induced Dzyaloshinskii-Moriya interaction. However, an imaginary  $t'$  does not change the physics in the limit of  $t = 0$ . With a purely imaginary  $t'$ , the time-reversal symmetry (TRS) is restored and we will show the presence of FFLO superconductivity with TRS in this limit.

*Band structure around quarter filling.* Around quarter filling, which is sufficiently away from half filling, one can assume that the effect of the Gutzwiller weights on the values of  $t'$  is weak and neglect the renormalization of  $t'$ . We can then diagonalize  $H_0$ ,

$$\begin{aligned}
 H_0 &= \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger \mathcal{H}_0(\mathbf{k}) \Psi_{\mathbf{k}}, \quad \Psi_{\mathbf{k}}^\dagger = (c_{\mathbf{k}\uparrow}^\dagger, c_{\mathbf{k}\downarrow}^\dagger, d_{\mathbf{k}\uparrow}^\dagger, d_{\mathbf{k}\downarrow}^\dagger), \\
 \mathcal{H}_0(\mathbf{k}) &= \begin{pmatrix} 0 & M^\dagger(\mathbf{k}) \\ M(\mathbf{k}) & 0 \end{pmatrix}, \\
 M(\mathbf{k}) &= t g(\mathbf{k}) \tau^0 + \sum_{\alpha=x,y,z} t' g_\alpha(\mathbf{k}) \tau^\alpha, \\
 h_\alpha(\mathbf{k}) &= 2t'^2 \sin \mathbf{k} \cdot \mathbf{R}_\alpha + 2tt'[1 + \cos \mathbf{k} \cdot (\mathbf{r}_\alpha - \mathbf{r}_\beta) \\
 &\quad + \cos \mathbf{k} \cdot (\mathbf{r}_\alpha - \mathbf{r}_\gamma)], \quad (2)
 \end{aligned}$$

in which  $\alpha \neq \beta, \gamma$  and  $g(\mathbf{k}) = \sum_\alpha e^{i\mathbf{k} \cdot \mathbf{r}_\alpha}$ , and  $g_\alpha(\mathbf{k}) = e^{i\mathbf{k} \cdot \mathbf{r}_\alpha}$  ( $\alpha = x, y, z$ ). We see that in the spin-orbit coupling limit ( $t = 0$ ) the Fermi surface has six additional inversion symmetry centers, apart from the inversion symmetry center  $O$  with trivial momentum  $\mathbf{Q}_0 = \mathbf{0}$ , in the first Brillouin zone (FBZ)  $\mathbf{k} \leftrightarrow 2\mathbf{q}_\alpha - \mathbf{k}$  ( $\alpha = x, y, z$ ), as indicated in Fig. 1(b). This derives from the sine function remaining invariant under the change of  $\mathbf{k} \cdot \mathbf{R}_\alpha \leftrightarrow \pi - \mathbf{k} \cdot \mathbf{R}_\alpha$ . In Fig. 1(c), we show the band structure at the spin-orbit coupling limit  $t = 0$ ,  $t' \neq 0$ : The four bands have a conic structure for the Fermi surface at half and quarter filling.

*Superconducting instability.* The doped itinerant Kitaev-Heisenberg model in the spin-orbit limit ( $t = 0$ ) has seven symmetry centers around quarter filling with momenta:  $\pm \mathbf{q}_\alpha$  ( $\alpha = x, y, z$ ) and  $\mathbf{q}_0 = \mathbf{0}$ . There are four kinds of Cooper pairs around these symmetry centers [59,60]:

$$\hat{\Delta}_{\alpha\mathbf{Q}_\alpha}^\dagger(\mathbf{k}) = i \tau_{\sigma\sigma'}^y \tau_{\sigma''\sigma'}^\alpha c_{\mathbf{k}\sigma}^\dagger d_{-\mathbf{k}+\mathbf{Q}_\alpha\sigma'}^\dagger \quad (\alpha = 0, x, y, z). \quad (3)$$

In direct space, the three types of spin-triplet pairing and the spin-singlet pairing in competition are

$$\begin{aligned}
 \hat{\Delta}_{ij}^x &= c_{i\uparrow} d_{j\uparrow} - c_{i\downarrow} d_{j\downarrow}, & \hat{\Delta}_{ij}^y &= i(c_{i\uparrow} d_{j\uparrow} + c_{i\downarrow} d_{j\downarrow}), \\
 \hat{\Delta}_{ij}^z &= c_{i\uparrow} d_{j\downarrow} + c_{i\downarrow} d_{j\uparrow}, & \hat{\Delta}_{ij}^0 &= c_{i\uparrow} d_{j\downarrow} - c_{i\downarrow} d_{j\uparrow}. \quad (4)
 \end{aligned}$$

Kitaev-Heisenberg coupling involves the density channel  $\hat{\chi}_\alpha = c_{i\sigma}^\dagger d_{j\sigma'} \tau_{\sigma\sigma'}^\alpha + \text{H.c.}$  besides the superconductivity pairing. We have checked that around quarter filling the density channel renormalizes the spin-orbit coupling term  $t'$  and such renormalization is negligible [60]. Then we can decompose the

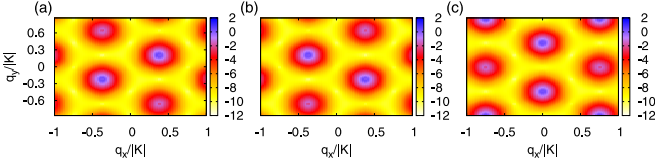


FIG. 2. The inverse of the vertex function  $\Gamma_{\alpha}^{-1}(\mathbf{Q}, T)$  at quarter filling  $\alpha = x, y, z$  as a function of  $\mathbf{q} = \frac{\mathbf{Q}}{2} \in \text{FBZ}$  (first Brillouin zone) at quarter filling for the spin-triplet pairing (a)  $\Delta_x\mathbf{Q}$ , (b)  $\Delta_y\mathbf{Q}$ , and (c)  $\Delta_z\mathbf{Q}$  at temperature  $k_B T = 0.01t'$  and  $t' = 1$ .

Kitaev-Heisenberg coupling at the mean-field level as

$$\begin{aligned} J_2 \sum_{(i,j)} [S_i^{\alpha} S_j^{\alpha} - S_i^{\beta} S_j^{\beta} - S_i^{\gamma} S_j^{\gamma}] \\ = \frac{3J_2 N_s}{4} \sum_{\alpha, \mathbf{Q}} |\Delta_{\alpha\mathbf{Q}}|^2 - J_2 \sum_{\alpha, \mathbf{k}, \mathbf{Q}} [g_{\alpha}(\mathbf{k}) \Delta_{\alpha\mathbf{Q}} \hat{\Delta}_{\alpha\mathbf{Q}}^{\dagger}(\mathbf{k}) \\ - g_{\alpha}(\mathbf{k}) \Delta_{0\mathbf{Q}} \hat{\Delta}_{0\mathbf{Q}}^{\dagger}(\mathbf{k}) + \text{H.c.}], \end{aligned} \quad (5)$$

in which  $\Delta_{\alpha\mathbf{Q}} = \frac{1}{N_s} \sum_{(i,j)} e^{i\mathbf{Q}\cdot\mathbf{r}_j} \langle \hat{\Delta}_{ij}^{\alpha} \rangle$  is the Fourier transform of the order parameter  $\langle \hat{\Delta}_{ij}^{\alpha} \rangle$  in Eq. (4) with spatial phase modulation  $e^{i\mathbf{Q}\cdot\mathbf{r}_j}$ .  $N_s$  denotes here the number of unit cells.

We constitute the Nambu spinor for the four Cooper pairs  $\Phi_{\mathbf{k}\mathbf{Q}} = (\Psi_{\mathbf{k}}, \Psi_{-\mathbf{k}}^{\dagger})$  [ $\Psi_{\mathbf{k}}$  is defined in Eq. (2)] and write down their Gor'kov-Green function  $G_{\alpha}^{-1}(\omega, \mathbf{k}, \mathbf{Q})$  ( $\alpha = 0, x, y, z$ ,  $\mathbf{Q}/2 \in \text{FBZ}$ ). We then pursue the Landau expansion [61]. In the spin-orbit coupling limit ( $t = 0$ ,  $t' \neq 0$ ), we have the second-order Landau expansion (here we fix  $U = 6$  following Ref. [42]),

$$F_{\text{BCS}} \approx - \sum_{\alpha, \beta=0,x,y,z} \sum_{\mathbf{Q}} N_s \Gamma_{\alpha\beta}^{-1}(\mathbf{Q}, T) \Delta_{\alpha\mathbf{Q}} \Delta_{\beta\mathbf{Q}}^*, \quad (6)$$

in which  $F_{\text{BCS}}$  is the free energy and to the lowest (second) order is proportional to the inverse of the Cooper pair vertex function  $\Gamma_{\alpha\beta}^{-1}(\mathbf{Q}, T)$  [61]. When  $\alpha \neq \beta$ , we have checked that  $\Gamma_{\alpha\beta}^{-1}(\mathbf{Q}, T)$  is negligible because of frustration in the momentum space; therefore we focus our attention on the diagonal part of the inverse of the Cooper pair vertex function that we denote as  $\Gamma_{\alpha}^{-1}(\mathbf{Q}, T) \equiv \Gamma_{\alpha\alpha}^{-1}(\mathbf{Q}, T)$ . When  $\Gamma_{\alpha}^{-1}(\mathbf{Q}, T) > 0$ , the triplet superconductor pairing  $\Delta_{\alpha\mathbf{Q}}$  is stable [62]. In Fig. 2, we show  $\Gamma_{\alpha}^{-1}(\mathbf{Q}, T)$  as a function of  $\mathbf{q} = \mathbf{Q}/2 \in \text{FBZ}$  at temperature  $k_B T = 0.01t'$ , in which we remark the condensation of spin-triplet Cooper pairs  $\Delta_{\alpha\mathbf{Q}}$  into the peaks at wave vector  $\mathbf{q}_{\alpha} = \frac{\mathbf{Q}_{\alpha}}{2}$ . We have three spin-triplet condensates at different momenta, as shown in Figs. 3(a)–3(c).

We also study the peak of the static Cooper pair response  $\Gamma_{\alpha}^{-1}(\mathbf{Q}_{\alpha}, T)$  as a function of temperature at different doping levels  $\delta$ : The peak remains finite at quarter filling, while it has logarithmic divergence at zero temperature when the doping diverts from quarter filling [Fig. 4(a)]. Here,  $\Gamma_{\alpha}^{-1}(\mathbf{Q}_{\alpha}, T)$  is proportional to the density of states at the Fermi level, which vanishes linearly as  $\delta \rightarrow 1/4$ , which means that at quarter filling, superconductivity disappears and we have a free electron system, assuming  $J_2$  is not too large compared to  $t'$ . Indeed, we have checked that at quarter filling, the critical value of  $J_2$  to reach a superconducting instability is  $J_{2c} \simeq 0.6t'$ , as shown in Fig. 5(b). At low temperature, the peak of the condensate

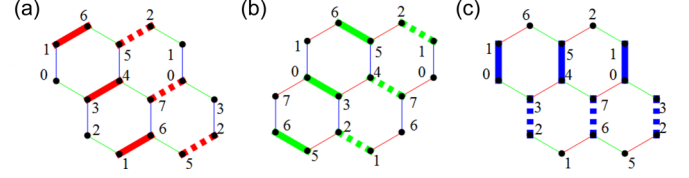


FIG. 3. Graphical representation of the three times degenerate ground state wave function of the FFLO superconductivity around quarter filling. The bold line signifies a spin-triplet pairing on the link  $\Delta_{ij}^{\alpha}$  with the spin-triplet type  $\alpha$  [Eq. (4)] in correspondence with the type of link [(a)  $x$  red, (b)  $y$  green, and (c)  $z$  blue]. The dashed line represents the same pairing but with a  $\pi$  phase (opposite sign in the wave function). Here, we only show the nearest-neighbor electron pairing. Long-range electron pairing exists and depends on the correlation length of the superconductor [58].

profile  $\Gamma_x^{-1}(\mathbf{Q}_x, T) = \Gamma_y^{-1}(\mathbf{Q}_y, T) = \Gamma_z^{-1}(\mathbf{Q}_z, T)$  stays positive while the peak of the spin-singlet condensate profile  $\Gamma_0^{-1}(0, T)$  remains negative at all temperatures [Fig. 4(b)]. This indicates that in the spin-orbit coupling limit, the doped itinerant Kitaev-Heisenberg model hosts only the three spin-triplet ground states. Since the phase related to  $\mathbf{Q}_{\alpha}$  is  $\pi$ , the analysis for  $-\mathbf{Q}_{\alpha}$  remains the same.

The three spin-triplet condensates may interact with each other, and we have calculated the box diagram to study this effect by extending the Landau expansion to the fourth order. We note

$$\begin{aligned} b_{xq}^{\dagger} &= \frac{1}{N_s} \sum_{\mathbf{k}} (c_{\mathbf{k}\uparrow}^{\dagger} d_{-\mathbf{k}+\mathbf{q}\uparrow}^{\dagger} - c_{\mathbf{k}\downarrow}^{\dagger} d_{-\mathbf{k}+\mathbf{q}\downarrow}^{\dagger}), \\ b_{yq}^{\dagger} &= -i \frac{1}{N_s} \sum_{\mathbf{k}} (c_{\mathbf{k}\uparrow}^{\dagger} d_{-\mathbf{k}+\mathbf{q}\uparrow}^{\dagger} + c_{\mathbf{k}\downarrow}^{\dagger} d_{-\mathbf{k}+\mathbf{q}\downarrow}^{\dagger}), \\ b_{zq}^{\dagger} &= -\frac{1}{N_s} \sum_{\mathbf{k}} (c_{\mathbf{k}\uparrow}^{\dagger} d_{-\mathbf{k}+\mathbf{q}\downarrow}^{\dagger} + c_{\mathbf{k}\downarrow}^{\dagger} d_{-\mathbf{k}+\mathbf{q}\uparrow}^{\dagger}), \end{aligned} \quad (7)$$

the creation operators for the three Cooper pairs. Since the three Cooper pairs condense at different momenta  $\mathbf{Q}_{\alpha}$ , the box diagram is actually the only one respecting momentum conservation. To the fourth order, we obtain the free energy of

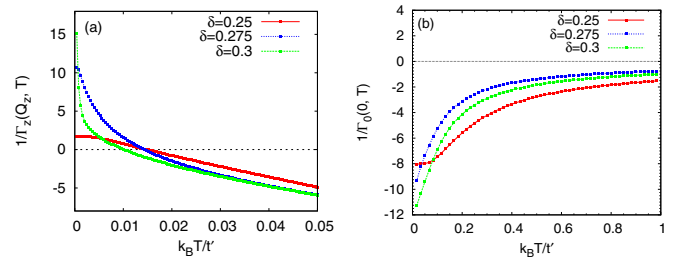


FIG. 4. In the limit  $t = J_1 = 0$ : (a) The peak of Cooper pair vertex function  $\Gamma_{\alpha}^{-1}(\mathbf{Q}_{\alpha}, T)$  as a function of temperature at different doping levels ( $\delta = 0.25$  is the quarter filling). (b) The vertex function of the singlet Cooper pair  $\Gamma_0^{-1}(0, T)$  as a function of temperature at different doping levels.

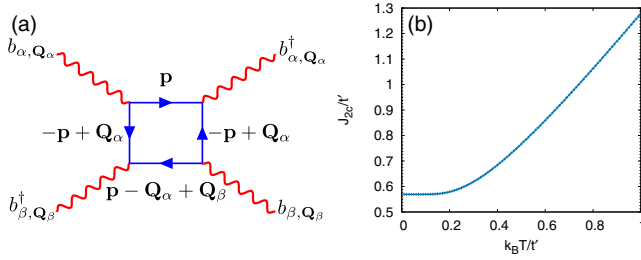


FIG. 5. Left panel: The box diagram of the fourth-order Landau expansion describing the interaction between the triplet pairing. Right panel:  $J_{2C}/t'$  as a function of temperature at quarter filling  $\delta = 0.25$ . The critical value of  $J_{2C}$  below which superconductivity instability is induced in the limit  $t = J_1 = 0$ .

the three condensates,

$$F_{\text{BCS}} = N_s \sum_{\alpha=x,y,z} \left\{ -\Gamma_\alpha^{-1}(\mathbf{Q}_\alpha, T) |\Delta_{\alpha\mathbf{Q}_\alpha}|^2 + C_1 |\Delta_{\alpha\mathbf{Q}_\alpha}|^4 \right\} + N_s C_2 \sum_{\alpha \neq \beta} |\Delta_{\alpha\mathbf{Q}_\alpha}|^2 |\Delta_{\beta\mathbf{Q}_\beta}|^2, \quad (8)$$

in which  $C_1$  and  $C_2$  are positive numbers obtained from the calculation of the box diagram in the left panel of Fig. 5(a). We have checked that  $C_2 > C_1 > 0$  and thus we deduce that mixing of the three superconducting condensates is not energetically favorable, and there is phase separation among the three types of fermionic pairs. Consequentially, the ground state wave function at zero temperature is three times degenerate (see Fig. 3): The modulated  $\Delta_{ij}^\alpha$  [Eq. (4)] are represented by bold and dashed lines [(a) red for  $x$ , (b) green for  $y$ , and (c) blue for  $z$ ].

When  $t$  and  $J_1$  are small compared to  $t'$  and  $J_2$ , the three FFLO states are still stable when the temperature is low enough [ $\Gamma_x^{-1}(\mathbf{Q}_x, T) = \Gamma_y^{-1}(\mathbf{Q}_y, T) = \Gamma_z^{-1}(\mathbf{Q}_z, T) > 0$ ]. The FFLO phase remains stable as long as the energy related to the critical temperature is bigger than the gap of the free electron system around quarter filling opened by the  $t$  term, i.e.,  $k_B T_c(\delta) > t$  [63]. In Fig. 5(b), we have plotted the critical value of  $J_{2C}$  for the superconductivity instability as a function of temperature  $T$ .

*Exact diagonalization of the Kitaev-Heisenberg model.* We have done an exact diagonalization of the Kitaev-Heisenberg model of Eq. (1) in the spin-orbit coupling limit  $t = 0, t' = 1$ . The exact diagonalization treats the Gutzwiller projectors exactly in Eq. (1). We fix the parametrization  $J_1 = \frac{4t'^2}{U}$ ,  $J_2 = \frac{4t'^2}{U}$  (here we choose  $U = 6$ , as suggested by Ref. [42]). The system has  $N_s = N_x \times N_y$  plaquettes with periodic boundary conditions in both directions, and is filled with  $N$  electrons on the  $2N_x \times N_y$  sites.  $N_x$  and  $N_y$  are both even numbers in order to avoid frustration of the FFLO condensates. Due to computational constraints, we reduce our study to three system sizes:  $N_y = 2, N_x = 2, 4, 6$ . For an odd number of Cooper pairs (doped system), the lowest-energy eigenstates appear in momentum sectors  $\mathbf{k}_x = (N_x/2, N_y/2)$ ,  $\mathbf{k}_y = (N_x/2, 0)$ , and  $\mathbf{k}_z = (0, N_y/2)$  [in the bases of  $\mathbf{k}_1 = \frac{1}{N_x}(0, \frac{4\pi}{3})$ ,  $\mathbf{k}_2 = \frac{1}{N_y}(\frac{2\pi}{\sqrt{3}}, -\frac{2\pi}{3})$ ], as shown in Figs. 6(b), 6(d), and 6(g). The degeneracy for the three spin-triplet states is partially lifted

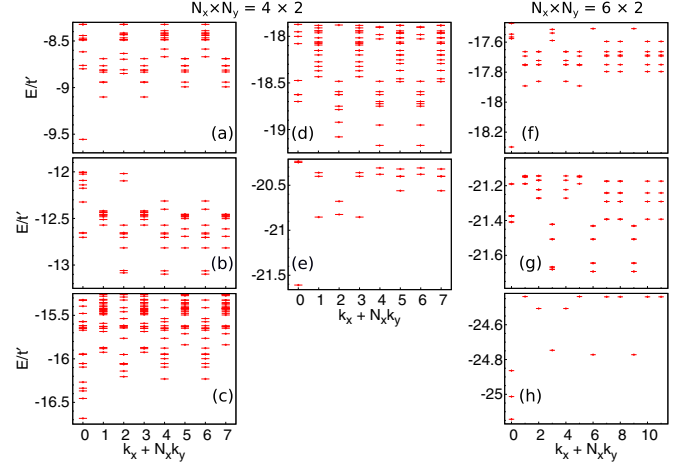


FIG. 6. Energy spectra as a function of the linearized momentum  $k_x + N_x k_y$  of the Hamiltonian in Eq. (1) with periodic boundary conditions and  $t = J_1 = 0, t' = 1, J_2 = 0.667$ . The left column and middle column show a system of  $N_x \times N_y = 4 \times 2$  plaquettes with particle numbers (a)  $N = 4$ , (b)  $N = 6$ , (c)  $N = 8$  or quarter filling (d)  $N = 10$  and (e)  $N = 12$ . The right column only provides the (f)  $N = 8$ , (g)  $N = 10$ , (h)  $N = 12$  spectra on a  $6 \times 2$  system [the largest Hilbert space dimension involved for (h) is  $\simeq 1.7 \times 10^8$ ]. Note that for this system, only the few first energy levels are shown.

when  $N_x \neq N_y$ , which breaks the symmetry of a  $2\pi/3$  rotation, followed by a permutation of spin components. For an even number of Cooper pairs, the ground state appears in momentum sector  $\mathbf{k}_0 = (0, 0)$ , as shown in Figs. 6(a), 6(c), 6(e), 6(f), and 6(h). In agreement with the theory,  $\mathbf{k}_\alpha$  coincides with the three discrete versions of the FFLO Cooper pair momenta  $\mathbf{q}_\alpha$  ( $\alpha = x, y, z$ ) for an odd number of Cooper pairs, while for an even number of Cooper pairs  $\mathbf{k}_0 \equiv 2\mathbf{k}_\alpha = 2\mathbf{q}_\alpha \pmod{(N_x, N_y)}$ . This alternation of the ground state momentum sector as a function of particle numbers distinguishes the FFLO superconductivity here from other modulated orders such as spin or charge density waves [64]. The quasidegeneracy in Fig. 6(b) is yet to be understood and might just be a finite size effect.

*Conclusion.* We have provided both analytical and numerical evidence of a pure spin-triplet FFLO superconductor in the doped itinerant Kitaev-Heisenberg model in the spin-orbit coupling limit ( $t, J_1 \rightarrow 0$ ). When  $t'$  is purely imaginary, the time-reversal symmetry (TRS) is restored, which might overcome the difficulties of the experimental realization of the FFLO phase. The key ingredient of the FFLO superconductivity here is the symmetry centers of the Fermi surface at nontrivial momenta instead of a Zeeman field. The ground state is three times degenerate with respectively the three spin-triplet pairing  $\Delta_{ij}^\alpha$  in the  $p$ -wave state with nontrivial Cooper pair momentum  $\mathbf{Q}_\alpha = 2\mathbf{q}_\alpha$  and spatial modulation of  $\pi$  phase in the direction of lattice vector  $\mathbf{R}_\alpha$  for the order parameter. These results may have relevance for doped iridate honeycomb materials or in ultracold atom systems. This FFLO state could be detected by possible Josephson effect measurements, by coupling such an FFLO material with a usual superconductor as proposed in several works, such as Ref. [65]. This FFLO state could also reveal interesting (short-range) magnetic fluctuations in

connection with the zigzag phase at half filling, which is beyond the scope of the present work.

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- [1] P. Anderson, *Science* **235**, 1196 (1987).  
 [2] F. C. Zhang and T. M. Rice, *Phys. Rev. B* **37**, 3759 (1988).  
 [3] M. Sigrist and K. Ueda, *Rev. Mod. Phys.* **63**, 239 (1991).  
 [4] P. W. Anderson, P. A. Lee, M. Randeria, T. M. Rice, N. Trivedi, and F. C. Zhang, *J. Phys.: Condens. Matter* **16**, R755 (2004).  
 [5] K. L. Hur and T. Maurice Rice, *Ann. Phys.* **324**, 1452 (2009).  
 [6] C. N. Yang, *Phys. Rev. Lett.* **63**, 2144 (1989).  
 [7] P. Fulde and R. A. Ferrell, *Phys. Rev.* **135**, A550 (1964).  
 [8] A. Larkin and Y. Ovchinnikov, *Sov. Phys. JETP* **20**, 762 (1965).  
 [9] A. Bianchi, R. Movshovich, C. Capan, P. G. Pagliuso, and J. L. Sarrao, *Phys. Rev. Lett.* **91**, 187004 (2003).  
 [10] Y.-a. Liao, A. S. C. Rittner, T. Paprotta, W. Li, G. B. Partridge, R. G. Hulet, S. K. Baur, and E. J. Mueller, *Nature (London)* **467**, 567 (2010).  
 [11] M. W. Zwierlein, A. Schirotzek, C. H. Schunck, and W. Ketterle, *Science* **311**, 492 (2006).  
 [12] Z. Cai, Y. Wang, and C. Wu, *Phys. Rev. A* **83**, 063621 (2011).  
 [13] C. Mora and R. Combescot, *Phys. Rev. B* **71**, 214504 (2005).  
 [14] F. Chevy and C. Mora, *Rep. Prog. Phys.* **73**, 112401 (2010).  
 [15] T. K. Koponen, T. Paananen, J.-P. Martikainen, and P. Törmä, *Phys. Rev. Lett.* **99**, 120403 (2007).  
 [16] H. Mayaffre, S. Krämer, M. Horvatić, C. Berthier, K. Miyagawa, K. Kanoda, and V. F. Mitrović, *Nat. Phys.* **10**, 928 (2014).  
 [17] I. Vasić, A. Petrescu, K. Le Hur, and W. Hofstetter, *Phys. Rev. B* **91**, 094502 (2015).  
 [18] A. M. Berridge, A. G. Green, S. A. Grigera, and B. D. Simons, *Phys. Rev. Lett.* **102**, 136404 (2009).  
 [19] Y. V. Fominov, Y. Tanaka, Y. Asano, and M. Eschrig, *Phys. Rev. B* **91**, 144514 (2015).  
 [20] S. Mironov, A. Mel'nikov, and A. Buzdin, *Phys. Rev. Lett.* **109**, 237002 (2012).  
 [21] F. Konschelle, J. Cayssol, and A. I. Buzdin, *Europhys. Lett.* **79**, 67001 (2007).  
 [22] T. Hotta, *J. Phys. Soc. Jpn.* **78**, 123710 (2009).  
 [23] P. Coleman, A. M. Tsvelik, N. Andrei, and H. Y. Kee, *Phys. Rev. B* **60**, 3608 (1999).  
 [24] E. Abrahams, A. Balatsky, J. R. Schrieffer, and P. B. Allen, *Phys. Rev. B* **47**, 513 (1993).  
 [25] E. Edel'shtein, *Sov. Phys. JETP* **68**, 1244 (1989).  
 [26] L. P. Gor'kov and E. I. Rashba, *Phys. Rev. Lett.* **87**, 037004 (2001).  
 [27] V. Mineev and K. Samokhin, *Sov. Phys. JETP* **78**, 401 (1994).  
 [28] A. B. Vorontsov and J. A. Sauls, *Phys. Rev. Lett.* **98**, 045301 (2007).  
 [29] A. B. Vorontsov, *Phys. Rev. Lett.* **102**, 177001 (2009).  
 [30] W. Witczak-Krempa and Y. B. Kim, *Phys. Rev. B* **85**, 045124 (2012).  
 [31] A. Shitade, H. Katsura, J. Kuneš, X.-L. Qi, S.-C. Zhang, and N. Nagaosa, *Phys. Rev. Lett.* **102**, 256403 (2009).  
 [32] J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, *Annu. Rev. Condens. Matter Phys.* **7**, 195 (2016).  
 [33] D. Pesin and L. Balents, *Nat. Phys.* **6**, 376 (2010).  
 [34] A. Kitaev, *Ann. Phys.* **321**, 2 (2006).  
 [35] G. B. Halász, J. T. Chalker, and R. Moessner, *Phys. Rev. B* **90**, 035145 (2014).  
 [36] J. Chaloupka, G. Jackeli, and G. Khaliullin, *Phys. Rev. Lett.* **105**, 027204 (2010).  
 [37] G. Jackeli and G. Khaliullin, *Phys. Rev. Lett.* **102**, 017205 (2009).  
 [38] Y. Singh and P. Gegenwart, *Phys. Rev. B* **82**, 064412 (2010).  
 [39] Y. Singh, S. Manni, J. Reuther, T. Berlijn, R. Thomale, W. Ku, S. Trebst, and P. Gegenwart, *Phys. Rev. Lett.* **108**, 127203 (2012).  
 [40] J. Reuther, R. Thomale, and S. Trebst, *Phys. Rev. B* **84**, 100406 (2011).  
 [41] S. H. Chun, J.-W. Kim, J. Kim, H. Zheng, C. C. Stoumpos, C. D. Malliakas, J. F. Mitchell, K. Mehlawat, Y. Singh, Y. Choi, T. Gog, A. Al-Zein, M. Moretti Sala, M. Krisch, J. Chaloupka, G. Jackeli, G. Khaliullin, and B. J. Kim, *Nature Physics* **11**, 462 (2015).  
 [42] T. Hyart, A. R. Wright, G. Khaliullin, and B. Rosenow, *Phys. Rev. B* **85**, 140510 (2012).  
 [43] Y.-Z. You, I. Kimchi, and A. Vishwanath, *Phys. Rev. B* **86**, 085145 (2012).  
 [44] D. D. Scherer, M. M. Scherer, G. Khaliullin, C. Honerkamp, and B. Rosenow, *Phys. Rev. B* **90**, 045135 (2014).  
 [45] G. Cao, T. F. Qi, L. Li, J. Terzic, V. S. Cao, S. J. Yuan, M. Tovar, G. Murthy, and R. K. Kaul, *Phys. Rev. B* **88**, 220414 (2013).  
 [46] S. R. Hassan, S. Goyal, R. Shankar, and D. Sénéchal, *Phys. Rev. B* **88**, 045301 (2013).  
 [47] J. Simon, W. S. Bakr, R. Ma, M. E. Tai, P. M. Preiss, and M. Greiner, *Nature (London)* **472**, 307 (2011).  
 [48] C. J. Kennedy, G. A. Siviloglou, H. Miyake, W. C. Burton, and W. Ketterle, *Phys. Rev. Lett.* **111**, 225301 (2013).  
 [49] J. Struck, J. Simonet, and K. Sengstock, *Phys. Rev. A* **90**, 031601 (2014).  
 [50] V. Galitski and I. B. Spielman, *Nature (London)* **494**, 49 (2013).  
 [51] L.-M. Duan, E. Demler, and M. D. Lukin, *Phys. Rev. Lett.* **91**, 090402 (2003).  
 [52] A. J. Leggett, *Rev. Mod. Phys.* **47**, 331 (1975).  
 [53] T. Zibold, V. Corre, C. Frapolli, A. Invernizzi, J. Dalibard, and F. Gerbier, *Phys. Rev. A* **93**, 023614 (2016).  
 [54] V. Corre, T. Zibold, C. Frapolli, L. Shao, J. Dalibard, and F. Gerbier, *Europhys. Lett.* **110**, 26001 (2015).  
 [55] B. Edegger, V. N. Muthukumar, and C. Gros, *Adv. Phys.* **56**, 927 (2007).  
 [56] T. M. Rice and K. Ueda, *Phys. Rev. Lett.* **55**, 995 (1985).

- [57] T. M. Rice and K. Ueda, *Phys. Rev. Lett.* **55**, 2093 (1985).
- [58] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.94.180506> for the section entitled “Coherence length.”
- [59] The superconducting instability is most significant around the symmetry centers of the Fermi surface with momentum  $\mathbf{q}$ , since, for any  $\mathbf{k}$ , electron pairs with respectively momentum  $\mathbf{k}$  and  $2\mathbf{q} - \mathbf{k}$  are both located on the Fermi surface, therefore the population of electrons forming Cooper pairs is the largest.
- [60] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.94.180506> for the section entitled “Spin-triplet pairings.”
- [61] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.94.180506> for the section entitled “Gor’kov-Green function.”
- [62] A. Atland and B. Simons, *Condensed Matter Field Theory* (Cambridge University Press, Cambridge, U.K., 2006).
- [63] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.94.180506> for the section entitled “Stability of the FFLO phase when  $0 < t, J_1 \ll t', J_2$ .”
- [64] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.94.180506> for the section entitled “Bloch theorem analysis for FFLO ground state wave function.”
- [65] K. Yang and D. F. Agterberg, *Phys. Rev. Lett.* **84**, 4970 (2000).