Interplay between Pair Density Wave and a Nested Fermi Surface

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We show that spontaneous time-reversal-symmetry (TRS) breaking can naturally arise from the interplay between pair density wave (PDW) ordering at multiple momenta and nesting of Fermi surfaces (FS). Concretely, we consider the PDW superconductivity on a hexagonal lattice with nested FS at 3/4 electron filling, which is related to a recently discovered superconductor CsV$_3$Sb$_5$. Because of nesting of the FS, each momentum $k$ on the FS has at least two counterparts $\pm k \pm Q_\alpha (\alpha = 1, 2, 3)$ on the FS to form finite momentum ($\pm Q_\alpha$) Cooper pairs, resulting in a TRS and inversion broken PDW state with stable Bogoliubov Fermi pockets. Various spectra, including (local) density of states, electron spectral function and the effect of quasi-particle interference, have been investigated. The partial melting of the PDW will give rise to 4 × 4 and $\frac{1}{3} \times \frac{1}{3}$ charge density wave (CDW) orders, in addition to the 2 × 2 CDW. Possible implications to real materials such as CsV$_3$Sb$_5$ and future experiments have been further discussed.

Introduction. — A pair density wave (PDW) is a superconducting (SC) state in which Cooper pairs carry finite momentum and its SC order parameter is spatially modulated without external magnetic field [1–19] (see, e.g. a recent review [20]). Such kind of state is similar to the one proposed earlier by Fulde-Ferrell (FF) [21] and Larkin-Ovchinnikov (LO) [22] (together known as FFLO) in magnetic field above the Pauli limit. As a mother state for various descendant orders, e.g., charge density wave (CDW), loop current, and charge-4e superconductivity, PDW has been receiving increasing attentions from diverse fields in physics [20, 23]. In particular, PDW was recently proposed as a promising candidate for explaining various interesting phenomena in cuprates and other strongly correlated systems [20, 24–27].

In previous studies of PDW, only electrons near hot spots on Fermi surface (FS) can come into being finite momentum Cooper pairs and are gapped, while other parts of FS remain gapless. In contrast, as will be revealed in this work, the FS nesting feature admits full PDW pairing around the FS, that will gain more condensation energy than the partial pairing usually, while in-gap quasi-particle excitations are still allowed. Thus, it would be of great interest to examine the interplay between these two, PDW and FS nesting. (Note that the role of FS nesting was considered mostly to CDW or spin-density-wave, e.g. see Ref. [28–33]).

In this letter, we study PDW ordering in the presence of a nested FS on hexagonal lattices. We found that a time reversal symmetry (TRS) breaking PDW state is energetically favored. Bogoliubov quasi-particle excitations, density of states (DOS), local DOS (LDOS), and electron spectral function will be investigated as well as the quasi-particle interference (QPI) in scanning tunnelling microscopy (STM). The implications to recently discovered Kagome SC AV$_3$Sb$_5$ (A=K,Rb,Cs) will be discussed.

Model. — We start with a single band model on a hexagonal lattice, on which the FS is nested as illustrated in Fig. 1. The Hamiltonian takes the form:

\[
H = \sum_{k,\sigma} \xi_k \epsilon_{k,\sigma}^{\dagger} \epsilon_{k,\sigma} + \sum_{k,\alpha} \left[ \Delta_{Q_\alpha}(k) \epsilon_{k,\uparrow}^{\dagger} \epsilon_{k+Q_\alpha,\downarrow} \right. \\
\left. + \Delta_{\alpha Q_\alpha}(k) \epsilon_{k,\downarrow}^{\dagger} \epsilon_{k-Q_\alpha,\uparrow} \right] + h.c.,
\]

(1)

where $\epsilon_{k,\sigma}^{\dagger}$ ($\epsilon_{k,\sigma}$) is electron creation (annihilation) operator with momentum $k$ and spin $\sigma = \uparrow, \downarrow$, $\xi_k = k - \mu$ is the energy measured from the chemical potential $\mu$. $\Lambda_{Q_\alpha}(k) = \Delta_{Q_\alpha} \exp \left[ \frac{-(\xi_k + 2\xi_{Q_\alpha})}{(2\Lambda)} \right] (\alpha = 1, 2, 3)$ indicates the Cooper pairing with total momentum $\pm Q_\alpha$. Here $\Lambda$ is an energy cutoff. Setting lattice constant $a = 1$, we consider $Q_1, Q_2, Q_3 = \left(0, \frac{\pi}{\sqrt{3}} \right), \left(-\frac{\pi}{2}, -\frac{\pi}{2\sqrt{3}} \right), \left(\frac{\pi}{2}, -\frac{\pi}{2\sqrt{3}} \right)$, since they are most relevant to CsV$_3$Sb$_5$ for which experimental evidences of period 4 PDW was recently reported.

FIG. 1. First Brillouin zone (BZ) of a hexagonal lattice and nesting of Fermi surfaces (FS) at 3/4 electron filling. (a) BZ and the nested FS at 3/4 filling. The boundary of BZ is in blue and red lines represent the FS. (b) Each $k$ on a FS segment along the $Q_\alpha$ direction has two counterparts $\pm k \pm Q_\alpha$ on the FS to form finite momentum ($\pm Q_\alpha$) Cooper pairs.
As shown in Fig. 1(a), a hexagonal and nested FS can be realized by choosing $\mu$ or the electron filling properly. As a simple example, we focus on a triangular lattice, set the nearest neighboring (NN) hopping integral $t = 1$, and choose $\mu = 2$ (or $3/4$ filling), such that $\xi_{k} = -2 \left[ \cos(k_{x}) + \cos\left(\frac{1}{2}k_{x} + \frac{\sqrt{3}}{2}k_{y}\right) + \cos\left(\frac{1}{2}k_{x} - \frac{\sqrt{3}}{2}k_{y}\right) + 1 \right]$. Note that our main results will also be applicable to more generic situations, including honeycomb and Kagome lattices.

From Fig. 1(b), one sees that each $k$ on a FS segment along the $Q_{a}$ direction has at least two counterparts $-k \pm Q_{a}$ to form finite momentum Cooper pairs. Moreover, $M$ and $X$ points have four momenta for pairing. These mean that the nesting feature allows full pairing in the region near the FS, which is in contrast with generic FS without nesting.

**Time reversal symmetry.** — The TRS of the Hamiltonian is respected if and only if $\xi_{-k} = \xi_{k}$ and $\Delta_{+\alpha}^{*}(k + Q_{a}) = \Delta_{-\alpha}(k)$. For the aforementioned form of $\Delta_{\alpha}(k)$, sufficient and necessary conditions for TRS reduce to $\Delta_{+\alpha} = \Delta_{-\alpha}$. The finite momentum pairing leads to a spatially varying pairing function in real space, resulting in a PDW. To be simple, we set $\Delta_{\alpha} = \Delta e^{\text{i} \theta_{\alpha} / 2}$, where $\Delta$ is real and positive, $\theta_{\alpha} \in (-\pi, \pi]$ and $\phi_{\alpha} \in (-\pi, \pi]$. Thus the pairing function $\Delta(r)$ reads

$$\Delta(r) = 2\Delta \sum_{\alpha} e^{\text{i} \theta_{\alpha}} \cos \left( Q_{a} \cdot r + \phi_{\alpha} / 2 \right).$$

**Commensurate PDW and descendant CDW order.** — When the PDW melts partially, i.e., the $U(1)$ gauge symmetry is restored but not the translational symmetry, a descendant CDW order will arise with wave vectors $q = \pm Q_{a} \pm Q_{p} \neq 0$. These wave vectors can be classified into three sets: (B) $q = \pm Q_{a}$ associated with a $4 \times 4$ CDW; (C) $q = \pm Q_{b}$ associated with a $2 \times 2$ CDW; (D) $q = \pm (Q_{b} - Q_{a})$ associated with a $4 \times \sqrt{3}$ CDW [see Fig. 3(c)]. Note that the FS is nested by $q = \pm 2Q_{a}$ in C but not those in B or D. So that the descendant CDW order can be of $4 \times 4$ and $\sqrt{3} \times \sqrt{3}$, in addition to the $2 \times 2$ CDW originating from the FS nesting.

**Quasi-particles.** — To study quasi-particle excitations in such a PDW associated with a $4 \times 4$ folded BZ, we introduce

$$\vec{C}_{k,\sigma}^{\dagger} = \left( c_{k+Q_{b},\sigma}^{\dagger}, c_{k-Q_{b},\sigma}^{\dagger}, c_{k+Q_{a},\sigma}^{\dagger}, c_{k-Q_{a},\sigma}^{\dagger}, c_{k+Q_{b}+Q_{a},\sigma}^{\dagger}, c_{k-Q_{b}+Q_{a},\sigma}^{\dagger}, c_{k+Q_{b}-Q_{a},\sigma}^{\dagger}, c_{k-Q_{b}-Q_{a},\sigma}^{\dagger}, c_{k+Q_{b}+Q_{a}-Q_{b},\sigma}^{\dagger}, c_{k-Q_{b}+Q_{a}-Q_{b},\sigma}^{\dagger}, c_{k+Q_{b}-Q_{a}+Q_{b},\sigma}^{\dagger}, c_{k-Q_{b}-Q_{a}+Q_{b},\sigma}^{\dagger}, c_{k+Q_{b}+Q_{b},\sigma}^{\dagger}, c_{k-Q_{b}+Q_{b},\sigma}^{\dagger}, c_{k+Q_{b}-Q_{b},\sigma}^{\dagger}, c_{k-Q_{b}-Q_{b},\sigma}^{\dagger}, c_{k+Q_{b}+Q_{b}-Q_{b},\sigma}^{\dagger}, c_{k-Q_{b}+Q_{b}-Q_{b},\sigma}^{\dagger}, c_{k+Q_{b}-Q_{b}+Q_{b},\sigma}^{\dagger}, c_{k-Q_{b}-Q_{b}+Q_{b},\sigma}^{\dagger} \right),$$

and rewrite Eq. (1) in a matrix form:

$$H = \frac{1}{16} \sum_{k} H_{k} + \sum_{k} \xi_{k},$$

$$= \frac{1}{16} \sum_{k} \left( \vec{C}_{k,\uparrow}^{\dagger}, \vec{C}_{-k,\downarrow}^{\dagger} \right) \hat{H}_{k} \left( \vec{C}_{k,\uparrow}^{\dagger}, \vec{C}_{-k,\downarrow}^{\dagger} \right) + \sum_{k} \xi_{k},$$

$$\hat{H}_{k} = \left( \hat{D}(k) \quad \hat{\Lambda}(k) \right) \left( \hat{\Lambda}^{\dagger}(k) \quad -\hat{D}(-k) \right).$$
numerical calculation finds that $E$ (d) Energy dispersion and Bogoliubov Fermi pockets for the lowest function of $E$ spontaneously. Owing to the approximate $\phi$ maxima at $\theta = \frac{\pi}{2}$, $\theta_1 = 0$, $\theta_2 = 2\pi/3$ and $\theta_3 = -2\pi/3$. (b) $E(k)^2$ around $X$ point that are plotted along $\Gamma - X - K$. (c) Bogoliubov Fermi pockets at $M$ and $X$ points and their periodic replica due to the PDW. (d) Quasi-particle (hole) pocket around $X$ point.

does not work any more. We shall diagonalize $\mathcal{H}_k$ numerically, and study the ground state and low energy excitations. Hereafter we set $\Lambda = 0.1$ and $\Delta = 0.02$, unless otherwise specified.

Condensation energy. — The condensation energy $E_c \equiv E_\nu - E_s$, that defined by the energy difference between a SC ground state and corresponding normal state [36], has been found as $E_c = \frac{1}{\pi} \sum_k \left( \frac{1}{2} \left| E(k) \right|^2 - E(k) \right)$ [see Eq. (2)]. The numerical calculation finds that $E_c(\theta_1, \theta_2, \theta_3, \phi)$ reaches local maxima at $\phi = \pm \pi/2$. This agrees with the above analysis of approximate $E(k)$ [see Eq. (6)] well. Moreover, as shown in Fig. 2 (a), the PDW state acquires maximum $E_c$ at $\phi = \pm \pi/2$ and $\theta_1 = 0$, $\theta_2 = \pm 2\pi/3$, breaking the TRS spontaneously. Owing to the $\mathbb{Z}_2$ symmetry theorem, $(\theta_2 \leftrightarrow \theta_3)$, the period is $\pi$ instead of $2\pi$ here.

Ginzburg-Landau free energy.— The TRS breaking and the $\mathbb{Z}_2$ symmetry can be verified in Ginzburg-Landau (GL) theory. Up to quartic order in $\Delta_{Q_s}$, the GL free energy can be written as [35],

$$ F[\Delta_{Q_s}] = F^{(0)} + F^{(2)}[\Delta_{Q_s}] + F^{(4)}[\Delta_{Q_s}], $$

where $F^{(0)}$ is $\Delta_{Q_s}$-independent, $F^{(2)} = g^{(2)} \sum_{\alpha = 1}^{3} \left( |\Delta_{Q_s}|^2 + |\Delta_{Q_s}|^2 \right)$ with $g^{(2)} < 0$, and $F^{(4)} = F^{(4)}_0 + F^{(4)}_\phi + F^{(4)}_\theta$. Here $F^{(4)}_0$ depends on $|\Delta_{Q_s}|$ only. $F^{(4)}_\phi$ and $F^{(4)}_\theta$ read

$$ F^{(4)}_\phi = g^{(4)}_\phi \left( |\Delta_{Q_s}|^2 Q_{s}^2 + |\Delta_{Q_s}|^2 Q_{s}^2 \right) + c.c. $$

and

$$ F^{(4)}_\theta = g^{(4)}_\theta \left( |\Delta_{Q_s}|^2 Q_{s}^2 + |\Delta_{Q_s}|^2 Q_{s}^2 \right) + c.c. $$

respectively, where both $g^{(4)}_\phi$ and $g^{(4)}_\theta$ are positive and $\Delta_{Q_s}$-independent [35]. Putting $\Delta_{Q_s} = \Delta e^{i\phi} e^{i \frac{\pi}{6}}$ into the above leads to $F^{(4)} = 2g^{(4)}_\phi \sum_{\alpha} \cos(2\alpha\theta_\alpha) + 2g^{(4)}_\theta \sum_{\alpha} \cos(2\alpha\theta_\alpha)$. Thus, the lowest free energy is achieved at $\phi = \pm \pi/2$ and $\theta_1 = 0$, $\theta_2 = \pm 2\pi/3$, and $\phi = \pi/2$, and study various electronic spectra.

Bogoliubov Fermi pockets. — As shown in Fig. 2 (b), around $M$ and $X$ points, $E(k)$ sinks down while $E(k)$ rises up, such that both of them go across zero energy. This means that quasi-particles (holes) possess FS indeed, namely, Bogoliubov Fermi pockets come into being. These Fermi pockets are located at $M$ and $X$ points and their periodic replica by the PDW (shifted by $q = \pm Q_\alpha \pm Q_\beta \neq 0$), as indicated in Fig. 2 (c). It is displayed in Fig. 2 (d) that these Fermi pockets exhibit $D_3$ symmetry.

Density of States. — The differential conductance $dI/dV$ measured by STM [37] is proportional to the DOS that reads $\rho(\omega) = -\frac{1}{2\pi} \sum_{k} \sum_{\nu, j=1}^{16} \left| u(k)_j \right|^2 \frac{\partial \delta E(k)^j}{\partial \omega} +$...
Define the integrated intensity of these CDW orders in addition to \( C \). As demonstrated in Fig. 3(d), \(|\tilde{\rho}(\mathbf{k}, \omega + i\delta)|^2\) is Green’s function in the absence of scatterings \( \tilde{T}(\omega) = \left[ (V, \tau_3)^{-1} - \frac{1}{N} \sum_{\mathbf{k}} \delta\mathbf{g}(\mathbf{k}, \omega + i\delta) \right]^{-1} \) is the scattering matrix. Here \( V_4 \) is the nonmagnetic scattering impurity strength, and \( \tau_3 \) is the Pauli matrix spanning Nambu space.

The modulation \(|\tilde{\rho}(\mathbf{q}, \omega)|\) with \( V_4 = 0.1 \) at \( \omega = 0.01(\Delta = 0.02) \) is plotted in Fig. 4(a). For comparison, we also study QPI of a uniform s-wave superconductor, as shown in Fig. 4(b). In both figures the intensity at \( \mathbf{q} = \mathbf{0} \) has been subtracted.

Electron spectral function. — The LDOS modulation due to scatterings can be analyzed by electron spectral function \( A(\mathbf{k}, \omega) = -\frac{1}{N} \text{Im} \left( \tilde{\mathcal{G}}(\mathbf{k}, \omega + i\delta) \hat{T}(\omega) \tilde{\mathcal{G}}(\mathbf{k}, \omega + i\delta) \right) \) in the absence of scatterings. As is pointed out in Ref. [39], the summation in Eq. (9) is dominated by terms in which both \( \mathbf{k} \) and \( \mathbf{k} + \mathbf{q} \) are poles of \( \tilde{\mathcal{G}} \). Thus the vectors \( \mathbf{q} \) associated with the scattering processes connecting two points with large \( A(\mathbf{k}, \omega) \) will show significant \(|\tilde{\rho}(\mathbf{q}, \omega)|\). This feature of \( \mathbf{q} \) is displayed in Fig. 4(c). An essential difference between the PDW state and a uniform s-wave state is that in-gap state is absent in the latter and the corresponding \( A(\mathbf{k}, \omega) \) will show significant \(|\tilde{\rho}(\mathbf{q}, \omega)|\) at \( \omega < \Delta \), as shown in Fig. 4(b) and (d). This also provides an experiment scheme to probe PDW states.

Discussions and conclusions. — (i) Recently discovered Kagome SC AV\(_3\)Sb\(_5\) (A=K,Rb,Cs) with a nearly \( 3/4 \) filled electron band is a natural platform towards the realization of the interplay between PDW and FS nesting [40–45]. TRS breaking signatures have been extensively discussed both experimentally and theoretically in AV\(_3\)Sb\(_5\) [46–51]. For the SC properties, the AV\(_3\)Sb\(_5\) is shown to be a spin-singlet SC hosting s-wave features [52–54]. However, a residual thermal transport at \( T = 0 \) and “multi-gap” V-shape DOS with residual zero-energy contributions were observed in SC states [34, 54–56], which conflicts with the conventional s-wave nature. This contradiction can be resolved within the TRS breaking PDW scenario proposed in the present work. More importantly, a PDW state ordering \( \mathbf{Q}_0 \) has been observed in recent STM measurements [34]. Therefore, our theory may provide new insight into the PDW states and TRS breaking in AV\(_3\)Sb\(_5\). Indeed, both \( 2 \times 2 \) and \( 4 \times 4 \) CDW have been observed in STM. Our theory suggests that the \( \Delta = 0.02 \) CDW should appear as well, as long as the frequency \( \omega \) is chosen properly.

(ii) Indeed, such a TRS breaking SC state breaks the spatial inversion symmetry as well [see Eq. (2)], resulting in a
chiral state with stable residual gapless quasi-particle excitations. The ground state is a flux state with spontaneous loop current [57, 58], as calculated in the Supplementary Material [35]. And the Bogoliubov Fermi pockets yield the linear $T$-dependent specific heat at low temperature.

(iii) One of remaining issues is what microscopic theory may give rise to the finite-momentum Cooper pairing instability on a nested FS. In the weak interaction limit, pairing at zero momentum is usually favored. Nonetheless, strong correlation might favor PDW instability against uniform pairing (see, e.g. Ref. [15, 59]). By establishing the microscopic model, the comparison with relevant models [60–63] based on the conventional CDW instabilities with nesting vector $2Q_a$ is one of the essential topics.

In summary, we have found that the FS nesting allows a full PDW pairing and in-gap states simultaneously. Such a PDW ansatz will give rise to a TRS breaking ground state. Subsequently, descendant CDW orders and various electronic spectra have been studied, and the relevance to newly discovered Kagome SC has been revealed.

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Supplementary Material for "Interplay between Pair Density Wave and a Nested Fermi Surface"

This supplementary material provides more details on our hexagonal lattice model, including the proof of $\mathbb{Z}_2$ symmetry theorem, the calculation of $E(k)^+$ along the FS segment, more numerical study on the condensation energy, ground state degeneracy and the derivation of Ginzburg-Landau free energy and local density of states.

I. THE PROOF OF $\mathbb{Z}_2$ Symmetry Theorem

Here we provide more details for the proof of the $\mathbb{Z}_2$ theorem presented in the main text.

**Theorem:** For each $\alpha$, the transformation $\Delta_\alpha \eta \mapsto -\Delta_\alpha \eta$ does not change the energy spectra of the system.

**Proof:** Without loss of generality, we consider $\alpha = 1$ and a centrosymmetric bipartition (A and B) of first BZ as illustrated in Fig. S1. Define a function $\eta(k) := +(-)1$ for $k \in A(B)$, we have $\eta(Q_1 - k) = \eta(Q_3 - k) = -\eta(Q_1 - k) = \eta(k)$. Thus, the unitary transformation: $c_{k,\uparrow} \mapsto c_{k,\uparrow}$ and $c_{k,\downarrow} \mapsto \eta(k)c_{k,\downarrow}$ gives rise to $\Delta_{\alpha \eta} \mapsto -\Delta_{\alpha \eta}$ and $\Delta_{\alpha \eta^2} \mapsto \Delta_{\alpha \eta^2}$, and will not change energy spectra. QED.

II. $E(k)^+$ Along the Hexagonal Fermi Surface

We now study the approximate $E(k)^+$ along the FS. By symmetry, we consider the FS segment $M-X$ [see Figs. 1(a) and 1(b) in the main text] only. For $k$ satisfies $k_x = \pi$ and $k_y \in \left(0, \frac{\pi}{\sqrt{3}}\right)$, with sufficiently small energy cutoff $\Lambda$, the decomposition of $\hat{H}_k$ in Eq. (3b) is of the following form,

$$H_k = \left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{k,\downarrow}\right) \hat{H}_k^p \left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{k,\downarrow}\right) + \left(\hat{c}_{\bar{k},\uparrow}^\dagger, \hat{\bar{c}}_{\bar{k},\downarrow}\right) \hat{H}_k^f \left(\hat{c}_{\bar{k},\uparrow}^\dagger, \hat{\bar{c}}_{\bar{k},\downarrow}\right).$$

(S1a)

where the pairing part $\hat{H}_k^p$ can be further decomposed as

$$\left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{\bar{k},\downarrow}\right) \hat{H}_k^p \left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{\bar{k},\downarrow}\right) = \left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{k,\uparrow}^\dagger, \hat{c}_{\bar{k},\downarrow}, \hat{c}_{\bar{k},\downarrow}\right) \hat{H}_k^p \left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{\bar{k},\downarrow}, \hat{c}_{\bar{k},\downarrow}, \hat{c}_{\bar{k},\downarrow}\right) + \left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{k,\uparrow}^\dagger, \hat{c}_{\bar{k},\downarrow}, \hat{c}_{\bar{k},\downarrow}\right) \hat{H}_k^p \left(\hat{c}_{k,\uparrow}^\dagger, \hat{c}_{\bar{k},\downarrow}, \hat{c}_{\bar{k},\downarrow}, \hat{c}_{\bar{k},\downarrow}\right).$$

FIG. S1. (a) A centrosymmetric bipartition for the 4 × 4 folded BZ. The folded zone is divided into two parts A and B, such that A is the inversion of B. (b) A $Q_1$ stripy tiling of the reciprocal plane with folded zone and its inversion. They are aligned alternatively along $Q_2$ and $Q_3$ directions, while keep $Q_1$ translational invariant. This tiling gives rise to a centrosymmetric bipartition for the whole BZ, such that $k$ and $Q_1 - k$ belong to opposite parts (A and B), while $Q_2 - k$ and $Q_1 - k$ belong to the same part. The red hexagon encloses first (unfolded) BZ.
Here $\hat{H}_k^{\hat{\phi}}$ reads

$$
\hat{H}_k^{\hat{\phi}} = \begin{pmatrix}
0 & 0 & \Delta_{Q_1} & \Delta_{Q_2} & \Delta_{Q_3} \\
0 & 0 & \Delta_{Q_1} & -\Delta_{Q_2} & -\Delta_{Q_3} \\
\Delta_{Q_1} & \Delta_{Q_1} & 0 & 0 & 0 \\
-\Delta_{Q_2} & -\Delta_{Q_2} & 0 & 0 & 0 \\
\Delta_{Q_3} & \Delta_{Q_3} & 0 & 0 & 0
\end{pmatrix} = \Lambda \begin{pmatrix}
0 & 0 & e^{i\phi_1 + \phi_2} & e^{-i\phi_1 + \phi_2} & e^{i\phi_1 - \phi_2} & e^{-i\phi_1 - \phi_2} \\
0 & 0 & e^{-i\phi_1 + \phi_2} & e^{i\phi_1 + \phi_2} & 0 & 0 \\
e^{i\phi_1 + \phi_2} & e^{-i\phi_1 + \phi_2} & 0 & 0 & 0 \\
e^{-i\phi_1 - \phi_2} & e^{i\phi_1 - \phi_2} & 0 & 0 & 0 \\
e^{-i\phi_1 - \phi_2} & e^{-i\phi_1 + \phi_2} & 0 & 0 & 0
\end{pmatrix} \begin{pmatrix}
0 & 0 & \Delta_{Q_1} & \Delta_{Q_2} & \Delta_{Q_3} \\
0 & 0 & \Delta_{Q_1} & -\Delta_{Q_2} & -\Delta_{Q_3} \\
\Delta_{Q_1} & \Delta_{Q_1} & 0 & 0 & 0 \\
-\Delta_{Q_2} & -\Delta_{Q_2} & 0 & 0 & 0 \\
\Delta_{Q_3} & \Delta_{Q_3} & 0 & 0 & 0
\end{pmatrix},
$$

(S1b)

$\hat{C}_k^{\hat{\phi}} = (c_{k+Q_1}^\dagger, c_{k+Q_2}^\dagger, c_{k+Q_3}^\dagger, c_{k+2Q_1}^\dagger, c_{k+2Q_2}^\dagger, c_{k+2Q_3}^\dagger, c_{k+Q_1+Q_2}^\dagger, c_{k+Q_1+Q_3}^\dagger, c_{k+Q_1+Q_2+Q_3}^\dagger, c_{k+Q_2+Q_3}^\dagger, c_{k+Q_3+Q_1}^\dagger, c_{k+Q_2+Q_1}^\dagger)$ and

$$
\hat{H}_k^{\hat{\phi}} = \begin{pmatrix}
\hat{D}'(k) & 0 \\
0 & -\hat{D}'(-k)
\end{pmatrix}.
$$

(S1c)

where $\hat{D}'(k) = \text{diag}(\xi_{k_i})$ is a diagonal matrix and $k_i^\dagger$ is the $i$-th momentum in $\hat{C}_k^{\hat{\phi}}$.

We define $E^p(k)$ and $E^f(k)$ as the lowest non-negative eigenvalues of $\hat{H}_k^{\hat{\phi}}$ and $\hat{H}_k^{\hat{\phi}}$, respectively. The eigenvalues of Eq. (S1b) are $\pm 2\Delta \sin (\phi_2/2)$ and $\pm 2\Delta \cos (\phi_2/2)$. Thus $E^p(k) = 2\Delta \min \{ |\sin (\phi_2/2)|, |\cos (\phi_2/2)| \}$. From Eq. (S1c), we obtain that $E^f(k) = \min(|\xi_{k_i}|)$. Hence we have

$$
E(k)_i^+ \approx \min \{ E^p(k), E^f(k) \} = \min \left\{ 2\Delta \min \left\{ |\sin (\phi_2/2)|, |\cos (\phi_2/2)| \right\}, \min(|\xi_{k_i}|) \right\}.
$$

(S2)

Notice that the eigenvalues of Eq. (S1b) and Eq. (S1c) are $\theta_\alpha$-independent. To study the effect of TRS breaking, we need to consider pairing along all the three directions of $Q_\alpha$ numerically.

### III. NUMERICAL STUDY ON CONDENSATION ENERGY

To show that the aforementioned approximation of $E(k)_i^+$ is reasonable and $E_c$ acquires local maxima at $\phi_\alpha = \pm \pi/2$, we study $E_c$ at different $\phi_\alpha$ numerically, keeping $\Delta = 0.02$ and $\Lambda = 0.1$. We fix the values of $\theta_\alpha$ randomly and find $\phi_\alpha$ that maximize $E_c$. For various random initial variables $\phi_\alpha$, the results that maximize the condensation energy fit $\phi_\alpha = \pm \pi/2$ well.

Here we present a numerical result of a special case: keeping $\theta_\alpha = 0, 2\pi/3, -2\pi/3$ and setting $\phi_1 = \phi_2 = \phi_3 = \phi$. The result of $E_c(\phi)$ is shown in Fig. S2. We can see that the maximum of $E_c(\phi)$ is found at $\phi = \pm \pi/2$, which is in good agreement with our analysis of approximate $E(k)_i^+$.

To further check our results, we maximize the condensation energy $E_c[\theta_2, \theta_3, \phi]$ numerically, keeping $\theta_1 = 0, \Delta = 0.02$ and $\Lambda = 0.1$. For various random initial variables $[\theta_2, \theta_3, \phi]$, the results that maximize the condensation energy fit $\phi_\alpha = \pm \pi/2$ and $\theta_2 - \theta_1 = \theta_3 - \theta_2 = \mp 2\pi/3$ (mod $\pi$) well.

![Condensation energy $E_c$ as a function of $\phi$. Here $\theta_1 = 0, \theta_2 = 2\pi/3, \theta_3 = -2\pi/3, \phi_1 = \phi_2 = \phi_3 = \phi, \Delta = 0.02$ and $\Lambda = 0.1$ have been chosen.](image)
IV. GROUND STATE DEGENERACY

Since the condensation energy is maximized at $\phi_0 = \pm \pi/2$ and $\theta_2 - \theta_1 = \theta_3 - \theta_2 \equiv \pm 2\pi/3 \pmod{\pi}$, each set of $[\theta, \phi]$ satisfies these conditions will give rise to a ground state of the system. We now discuss the ground state degeneracy of our PDW state. Notice that a gauge transformation reads $\varepsilon_{r,s} \mapsto e^{i\theta/2} \varepsilon_{r,s}$ leads to a transformation of $\theta_\alpha$ as follows,

$$[\theta_1, \theta_2, \theta_3] \mapsto [0, \theta_2 - \theta_1, \theta_3 - \theta_1].$$

Thus, we can always set $\theta_1 = 0$ and focus on different sets of $[\theta_2, \theta_3, \phi_0]$ satisfy $\phi_0 = \pm \pi/2$ and $\theta_2 = \theta_3 - \theta_2 \equiv \pm 2\pi/3 \pmod{\pi}$. There are 8 sets of $[\theta_2, \theta_3]$ satisfy $\theta_2 = \theta_3 - \theta_2 \equiv \pm 2\pi/3 \pmod{\pi}$ (it can be seen from the 8 maxima in Fig. 2(a) in the main text) and 8 sets of $[\phi_0, \phi_2, \phi_3]$ satisfy $\phi_0 = \pm \pi/2$. Consequently, the ground state is of $8 \times 8$-fold degeneracy.

The 64 different ground states can be related through several symmetry operators. It can be seen from the transformation of $\Delta(r)$ under the corresponding symmetry operator. Recall the form of $\Delta(r)$ in Eq. (2),

$$\Delta(r) = 2\Delta \sum_{\alpha} e^{i\theta_\alpha} \cos \left( Q_\alpha \cdot r + \frac{\phi_\alpha}{2} \right).$$

The $\mathbb{Z}_2$ transformation $\Delta_{\pm Q_\alpha} \mapsto -\Delta_{\pm Q_\alpha}$ gives rise to $\theta_\alpha \mapsto \theta_\alpha + \pi$ (for a certain $\alpha$). The time reversal transformation leads to $\Delta(r) \mapsto \Delta'(r)$ as well as $\theta_\alpha \mapsto -\theta_\alpha$ (acting on all the three $\alpha$ simultaneously). Thus the 8 sets of $[\theta_2, \theta_3]$ can be related with each other through these two kinds of transformations. As for the 8 sets of $[\phi_0, \phi_2, \phi_3]$, we can apply the lattice translation operator: $T_i : r \mapsto r + a_i$ ($Q_1 \cdot a_j = \pi/2 \delta_{ij}$, $i, j = 1, 2$), the spatial inversion operator $\mathcal{P} : r \mapsto -r$ and the $\mathbb{Z}_2$ transformation to change the sign of one $\phi_\alpha$ and keep the other two unchanged. This process provides a path links two sets of $[\phi_0, \phi_2, \phi_3]$ and all the 8 sets of $[\phi_0, \phi_2, \phi_3]$ are related with each other through these paths. As a concrete example, we give the specific process of the transformation $[\pi/2, \pi/2, \pi/2] \mapsto [-\pi/2, \pi/2, \pi/2]$ here. Begin with $[\phi_0, \phi_2, \phi_3] = [\pi/2, \pi/2, \pi/2]$, $\Delta(r)$ is of the following form

$$\Delta_0(r) = 2\Delta \left[ \cos \left( Q_1 \cdot r + \frac{\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_2 \cdot r + \frac{\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_3 \cdot r + \frac{\pi}{4} \right) \right].$$

(S3a)

Under $T_2 : r \mapsto r + a_2$, $\Delta(r)$ becomes

$$\Delta_0(r) \mapsto \Delta_1(r) = 2\Delta \left[ \cos \left( Q_1 \cdot r + \frac{\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_2 \cdot r + \frac{3\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_3 \cdot r - \frac{\pi}{4} \right) \right].$$

(S3b)

Then the transformation $\Delta_{\pm Q_1} \mapsto -\Delta_{\pm Q_1}$ ($\theta_1 \mapsto \theta_1 + \pi$) gives rise to

$$\Delta_1(r) \mapsto \Delta_2(r) = 2\Delta \left[ \cos \left( Q_1 \cdot r + \frac{\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_2 \cdot r - \frac{\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_3 \cdot r - \frac{\pi}{4} \right) \right].$$

(S3c)

Finally, we obtain $\Delta(r)$ with $[\phi_0, \phi_2, \phi_3] = [-\pi/2, \pi/2, \pi/2]$ under the operator $\mathcal{P}$,

$$\Delta_2(r) \mapsto \Delta_3(r) = 2\Delta \left[ \cos \left( Q_1 \cdot r - \frac{\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_2 \cdot r + \frac{\pi}{4} \right) + e^{i\theta_0} \cos \left( Q_3 \cdot r + \frac{\pi}{4} \right) \right].$$

(S3d)

V. GINZBURG-LANDAU FREE ENERGY

We begin with the Gorkov Green’s function $G(i\omega_n, k)$ in our PDW state,

$$G^{-1}(i\omega_n, k) \equiv G_0^{-1}(i\omega_n, k) + \Sigma(k),$$

(S4)

where

$$G_0^{-1}(i\omega_n, k) = \begin{pmatrix} G_0^{-1}(i\omega_n, k) & 0 \\ 0 & -G_0^{-1}(i\omega_n, k) \end{pmatrix} \Sigma(k) = \begin{pmatrix} 0 & -\hat{\Delta}(k) \\ -\hat{\Delta}^\dagger(k) & 0 \end{pmatrix}.$$

(S5)

Here $G_0(i\omega_n, k)$ is the normal state Green’s function. In our PDW state, $G_0(i\omega_n, k)$ is a $16 \times 16$ matrix satisfies $G_0(i\omega_n, k)_{ij} = (i\omega_n - \xi_k)^{-1} \delta_{ij}$ ($\xi_k$ is the $i$-th momentum in $C_k^{(2)}$ in the main text). $\hat{\Delta}(k)$ is a $16 \times 16$ pairing matrix defined by $\Delta_{\pm Q_\alpha}(k)$.

The mean-field free energy can be expressed as

$$\mathcal{F}[\Delta_{\pm Q_\alpha}] \equiv -\frac{1}{16\beta} \sum_{n,k} \mathrm{Tr} \ln G^{-1}_{\pm}(i\omega_n, k) = \mathcal{F}^{(0)} - \frac{1}{16\beta} \sum_{n,k} \mathrm{Tr} \ln (1 + G_0(i\omega_n, k) \Sigma(k)) = \mathcal{F}^{(0)} + \sum_{j=1}^{\infty} \mathcal{F}^{(2j)},$$

(S6)
where $F^{(0)}$ is a constant that is independent of $\Delta_Q$. $F^{(2)}$ is the free energy of order $|\Delta_Q|^2$ of the following form,

$$F^{(2)} = \frac{1}{32\beta} \sum_{n,k} \text{Tr} \left[ G_0(i\omega_n, k) \Sigma(k) \right]^2 = \frac{1}{16\beta} \sum_{n,k} \text{Tr} \left[ (-G_0(i\omega_n, k) \Delta(k) G_0(-i\omega_n, -k) \Delta'(k)) \right].$$  

(S7)

Notice that the factor $\frac{1}{16\beta}$ comes from the $4 \times 4$ folding of the system and the summation over $k$ is performed in the first BZ. For $j = 1$,

$$F^{(2)} = -\frac{1}{16\beta} \sum_{n,k} \text{Tr} \left[ G_0(i\omega_n, k) \Delta(k) G_0(-i\omega_n, -k) \Delta'(k) \right] = \frac{1}{16\beta} \sum_{n,k} e^{\frac{|k|}{\beta}} a(i\omega_n, k),$$  

(S8)

where $\Lambda$ is the energy cutoff and

$$a(i\omega_n, k) = \sum_{n=1}^3 \left( e^{-\frac{|k|}{\beta}} |\Delta_Q|^2 + e^{-\frac{|k|}{\beta}} |\Delta_{-Q}|^2 \right).$$  

(S9)

By performing the summation over $i\omega_n$ and $k$, we can obtain the following $F^{(2)}$,

$$F^{(2)} = g^{(2)} \sum_{n=1}^3 (|\Delta_Q|^2 + |\Delta_{-Q}|^2),$$  

(S10)

where $g^{(2)}$ is the corresponding coefficient.

For $j = 2$,

$$F^{(4)} = \frac{1}{32\beta} \sum_{n,k} \text{Tr} \left[ G_0(i\omega_n, k) \Delta(k) G_0(-i\omega_n, -k) \Delta'(k) \right] = \frac{1}{32\beta} \sum_{n,i,j} \sum'_{k} \frac{1}{(i\omega_n - \xi_k)} \frac{1}{(i\omega_n + \xi_{-k})} b(i\omega_n, k, j),$$  

(S11)

where we introduce $\sum'$ to represent the summation over $k$ with the energy cutoff as follows,

$$\sum' = \frac{1}{(i\omega_n - \xi_k)} \frac{1}{(i\omega_n + \xi_{-k})} b(i\omega_n, k, j),$$  

(S12)

Since the system is of $4 \times 4$ folded, we can keep $i = 1$ as well as $k = k$ and calculate the summation over $j, i\omega_n$ and $k$. Then the summation over $i$ will give rise to 16 copies. Thus, we can obtain the following $F^{(4)}$ according to Eq. (S11),

$$F^{(4)} = \frac{1}{16\beta} \sum_{n,i,j} b(i\omega_n, k, j),$$  

(S13)

where $b(i\omega_n, k, j)$ reads

$$(S14a)$$

$$b(i\omega_n, k, k + Q_1) = \left( \frac{\Delta_{Q}^{\ast} \Delta_Q}{(i\omega_n + \xi_{-k+Q_1})} + \frac{\Delta_Q^{\ast} \Delta_{Q_2}}{\text{c.c.}} \right) \left( \frac{\Delta_{Q}^{\ast} \Delta_Q}{(i\omega_n + \xi_{k+Q_1})} + \frac{\Delta_Q^{\ast} \Delta_{Q_2}}{\text{c.c.}} \right),$$

(S14b)

$$b(i\omega_n, k, k - Q_1) = \left( \frac{\Delta_{Q}^{\ast} \Delta_Q}{(i\omega_n + \xi_{-k+Q_1})} + \frac{\Delta_Q^{\ast} \Delta_{Q_2}}{\text{c.c.}} \right) \left( \frac{\Delta_{Q}^{\ast} \Delta_Q}{(i\omega_n + \xi_{k+Q_1})} + \frac{\Delta_Q^{\ast} \Delta_{Q_2}}{\text{c.c.}} \right),$$

(S14c)
\[ b(i\omega_n, \mathbf{k} + \mathbf{Q}) = \left( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - k - \mathbf{Q}_1} + \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - k - \mathbf{Q}_1} \right) \left( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - k + \mathbf{Q}_1} + \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - k + \mathbf{Q}_1} \right) \]

\[ = \left( \frac{\Delta_0^2}{i\omega_n + \xi - k - \mathbf{Q}_1} + \frac{\Delta_0^2}{i\omega_n + \xi - k - \mathbf{Q}_1} \right) \left( \frac{\Delta_0^2}{i\omega_n + \xi - k + \mathbf{Q}_1} + \frac{\Delta_0^2}{i\omega_n + \xi - k + \mathbf{Q}_1} \right) \]

\[ \text{for } \Delta_0^2 = \Delta_0^2 \text{ and } \Delta_0^2 = \Delta_0^2 \text{.} \]

\[ (S14d) \]
\begin{align}
\bar{b}(i\omega_n, \mathbf{k}, \mathbf{k} - \mathbf{Q}_1 + \mathbf{Q}_3) &= \bigg( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} + \frac{\Delta_0 \Delta_0^{-1}}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} \bigg) \bigg( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} + \frac{\Delta_0 \Delta_0^{-1}}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} \bigg) + c.c. \\
\bar{b}(i\omega_n, \mathbf{k}, \mathbf{k} + \mathbf{Q}_2 - \mathbf{Q}_3) &= \bigg( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} + \frac{\Delta_0 \Delta_0^{-1}}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} \bigg) \bigg( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} + \frac{\Delta_0 \Delta_0^{-1}}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} \bigg) + c.c. \\
\bar{b}(i\omega_n, \mathbf{k}, \mathbf{k} - \mathbf{Q}_2 + \mathbf{Q}_3) &= \bigg( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} + \frac{\Delta_0 \Delta_0^{-1}}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} \bigg) \bigg( \frac{\Delta_0 \Delta_0^*}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} + \frac{\Delta_0 \Delta_0^{-1}}{i\omega_n + \xi - \mathbf{k} \cdot \mathbf{Q}} \bigg) + c.c.
\end{align}

By performing the summation over \( i\omega_n \) and \( \mathbf{k} \), \( \mathcal{F}^{(4)} \) is of the following form,

\[
\mathcal{F}^{(4)} = 6 \left( g_1^{(4)} + \frac{g_2^{(4)}}{2} + g_3^{(4)} + g_4^{(4)} \right) \Delta^4 + 2 g_5^{(4)} \Delta^4 \sum_{n=1}^{3} \cos(2\phi_n) + 2 g_6^{(4)} \Delta^4 [\cos(2\theta_2 - 2\theta_1) + \cos(2\theta_3 - 2\theta_1) + \cos(2\theta_1 - 2\theta_3)].
\]

We are interested in the signs of \( g_5^{(4)} \) and \( g_6^{(4)} \) since they determine the values of \( \phi_n \) and \( \theta_n \) that minimize the free energy respectively. According to Eq. (S13) and Eqs. (S14), \( g_5^{(4)} \) and \( g_6^{(4)} \) can be expressed as

\[
\begin{align}
\bar{g}_5^{(4)} &= \frac{1}{\beta} \sum_{n} \sum_{\mathbf{k}} \frac{1}{(i\omega_n - \xi) (i\omega_n - \xi + 2\mathbf{Q}) (i\omega_n + \xi + \mathbf{Q}) (i\omega_n + \xi - \mathbf{Q})}, \\
\bar{g}_6^{(4)} &= \frac{1}{\beta} \sum_{n} \sum_{\mathbf{k}} \frac{1}{(i\omega_n + \xi - \mathbf{Q}) (i\omega_n + \xi + \mathbf{Q}) (i\omega_n + \xi - \mathbf{Q}) + (i\omega_n - \xi) (i\omega_n - \xi + 2\mathbf{Q}) (i\omega_n + \xi + \mathbf{Q}) (i\omega_n + \xi - \mathbf{Q})}.
\end{align}
\]

where we have used \( \xi = \xi - \mathbf{k} \) to combine terms in \( g_\theta^{(4)} \). Now we can use the following formula to achieve the summation over \( i\omega_n \),

\[
\begin{align}
\frac{1}{\beta} \sum_{n} \frac{1}{(i\omega_n - \xi_k) (i\omega_n - \xi_k) (i\omega_n + \xi_k) (i\omega_n + \xi_k)} &= \frac{n_F(\xi_k)}{\xi_k - \xi_k} + \frac{n_F(-\xi_k)}{-\xi_k - \xi_k} + \frac{n_F(\xi_k)}{\xi_k + \xi_k} + \frac{n_F(-\xi_k)}{-\xi_k + \xi_k}.
\end{align}
\]
where \( n_F \) is the Fermi-Dirac distribution function.

By performing the summation over \( i\omega_n \), \( g^{(4)}_\phi \) is of the following form,

\[
g^{(4)}_\phi = \frac{1}{\beta} \sum_k \frac{n_F(\xi_k)}{2} \left[ \left( \xi_k - \xi_{k+2Q1} \right) \left( \xi_k + \xi_{k+Q1} \right) \right] + \frac{n_F(\xi_{k+2Q1})}{2} \left[ \left( \xi_{k+2Q1} - \xi_k \right) \left( \xi_{k+2Q1} + \xi_{k-Q1} \right) \right] \quad (S19)
\]

where we have used \( n_F(x) + n_F(-x) = 1 \). Due to the energy cutoff \( \Lambda \), only the summation near the FS segment defined by \( k_x = \pm \pi \) will give rise to a sizable contribution to the expression above. Notice that near \( k_x = \pm \pi \), the following relation that represents the nesting feature of the hexagonal FS holds,

\[
\xi_{k+2Q1} \approx -\xi_k, \quad \xi_{-k-Q1} \approx -\xi_{-k-Q1}.
\]

In the low-energy limit \( (\beta \xi_k \ll 1) \), the function \( 2n_F(\xi_k) - 1 \) can be expanded as follows

\[
2n_F(\xi_k) - 1 \approx -\frac{\beta \xi_k^2}{2} + \frac{\beta^3 \xi_k^2}{24}.
\]

Then we have

\[
g^{(4)}_\phi \approx \frac{1}{\beta} \sum_k \frac{\beta \xi_k^2}{96} \frac{\beta^3 \xi_k^2}{96} = \frac{1}{\beta} \sum_k \frac{\beta}{4} \left( \xi_k^2 - \xi_{-k-Q1}^2 \right) = \sum_k \frac{\beta^3}{96} > 0.
\]  

We draw the conclusion that \( g^{(4)}_\phi \) is positive.

We now focus on the derivation of \( g^{(4)}_\phi \). Similarly, we perform the summation over \( i\omega_n \) at first,

\[
\frac{1}{\beta} \sum_k \sum_{k'} \frac{1}{i(\omega_n - \xi_k)(\omega_n - \xi_{k-Q1})(\omega_n + \xi_{-k-Q1})(\omega_n + \xi_{-k-Q1})} n_F(\xi_k) + n_F(\xi_{k+2Q1}) \quad (S21a)
\]

By taking the substitution \( k \rightarrow -k + Q_1 \) and adding it to the original expression, we obtain the following expression,

\[
g^{(4)}_\phi \approx \sum_k \frac{\beta^3}{96} \left( \xi_k^2 - \xi_{-k-Q1}^2 \right) = \sum_k \frac{\beta^3}{96} > 0.
\]  

We draw the conclusion that \( g^{(4)}_\phi \) is positive.
Then \( g^{(4)}_\phi \) is of the following form,

\[
g^{(4)}_\phi = g^{(4)}_{\phi,1} + g^{(4)}_{\phi,2},
\]

\[
g^{(4)}_{\phi,1} = 2 \sum' \frac{2n_F(\xi_k) - 1}{(\xi_k - 3q^2_\phi)} (\xi_k + 3q^2_\phi),
\]

\[
g^{(4)}_{\phi,2} = 2 \sum' \frac{2n_F(\xi_k) - 1}{(\xi_k - 3q^2_\phi)} (\xi_k + 3q^2_\phi). \tag{S22c}
\]

For \( g^{(4)}_{\phi,1} \), only \( k \) near \((0, 2\pi/\sqrt{3})\) or \((-\pi, 0)\) gives a sizable contribution to the summation above due to the energy cutoff. For \( k = (0, 2\pi/\sqrt{3}) + q \), we have

\[
\xi_k \approx \frac{1}{2}(q^2_\phi - 3q^2_\phi), \quad \xi_{k+Q} \approx 2q_x, \quad \xi_{-k+Q} \approx -q_x + \sqrt{3}q_y, \quad \xi_{-k-Q} \approx -q_x - \sqrt{3}q_y,
\]

where \( q \) is a small vector. Similarly, for \( k = (-\pi, 0) + q \), we have

\[
\xi_k \approx -2q_x, \quad \xi_{k+Q} \approx -q_x + \sqrt{3}q_y, \quad \xi_{-k+Q} \approx q_x, \quad \xi_{-k-Q} \approx q_x - \sqrt{3}q_y.
\]

Thus we can obtain the following approximation of \( g^{(4)}_{\phi,1} \),

\[
g^{(4)}_{\phi,1} \approx \sum_q \frac{2n_F(\xi_k)}{3(q^2_\phi - 3q^2_\phi)} - \frac{\beta(-2q_x + (\beta(-2q_x))^2)}{12}.
\]

\[
= \sum_q \frac{\beta}{q^2_\phi - 3q^2_\phi} + \frac{\beta^3 q^2_\phi}{3(q^2_\phi - 3q^2_\phi),} \tag{S23}
\]

where the first term in the first line vanishes since it changes sign under \( q \to -q \).

For \( g^{(4)}_{\phi,2} \), only \( k \) near \((\pm\pi/2, \sqrt{3}\pi/2)\) gives a sizable contribution to the summation above. For \( k = (\pi/2, \sqrt{3}\pi/2) + q \), we have

\[
\xi_k \approx q_x + \sqrt{3}q_y, \quad \xi_{k+Q} \approx q_x - \sqrt{3}q_y, \quad \xi_{-k+Q} \approx -2q_x, \quad \xi_{-k-Q} \approx -q_x - \sqrt{3}q_y.
\]

For \( k = (-\pi/2, \sqrt{3}\pi/2) + q \), we have

\[
\xi_k \approx -q_x + \sqrt{3}q_y, \quad \xi_{k+Q} \approx -q_x - \sqrt{3}q_y, \quad \xi_{-k+Q} \approx 2q_x, \quad \xi_{-k-Q} \approx q_x - \sqrt{3}q_y.
\]

Thus we can obtain the following approximation of \( g^{(4)}_{\phi,2} \),

\[
g^{(4)}_{\phi,2} \approx \sum_q \frac{-\beta(q_x + \sqrt{3}q_y) + \beta^3(q_x + \sqrt{3}q_y)^2}{12}.
\]

\[
= \sum_q \frac{\beta}{q^2_\phi - 3q^2_\phi} - \frac{\beta^3 q^2_\phi}{4(q^2_\phi - 3q^2_\phi),} \tag{S24}
\]

Finally, we obtain the following expression,

\[
g^{(4)}_\phi = g^{(4)}_{\phi,1} + g^{(4)}_{\phi,2} \approx \sum_q \frac{\beta^3 q^2_\phi}{12(q^2_\phi - 3q^2_\phi)} = \sum_q \frac{\beta^3}{12} > 0. \tag{S25}
\]

Hence, we find that \( g^{(4)}_\phi \) is also positive.

For positive \( g^{(4)}_\phi \) and \( g^{(4)}_\theta \), according to Eq. (S16), we should minimize the following functions to obtain the minimum free energy up to \( \Delta^4 \),

\[
h_\phi = \cos(2\phi_1) + \cos(2\phi_2) + \cos(2\phi_3), \tag{S26a}
\]

\[
h_\theta = \cos(2\theta_2 - 2\theta_1) + \cos(2\theta_3 - 2\theta_2) + \cos(2\theta_1 - 2\theta_3). \tag{S26b}
\]
It is easy to see that $h_0$ reaches its minimum $-3$ at $\phi_0 = \pm \pi/2$. For $h_0$, let $x = \theta_2 - \theta_1$ and $y = \theta_3 - \theta_2$, we have

$$h_0 = \cos(2x) + \cos(2y) + \cos(2(x + y)).$$

$h_0$ reaches its minimum $-3/2$ at $x = y = \pm 2\pi/3 \pmod{\pi}$. Thus, our PDW state acquires minimum free energy at $\phi = \pm \pi/2$ and $\theta_2 - \theta_1 = \theta_3 - \theta_2 = \pm 2\pi/3 \pmod{\pi}$, leading to a spatial inversion symmetry and TRS breaking state.

VI. LOCAL DENSITY OF STATES

Since there is no spin-flip effect, the charge density of the system is of the following form,

$$\rho(\mathbf{r}) = \frac{1}{N} \langle \hat{c}_{\mathbf{r},\uparrow} \hat{c}_{\mathbf{r},\uparrow} \rangle + \frac{2}{N} \langle \hat{c}_{\mathbf{r},\downarrow} \hat{c}_{\mathbf{r},\downarrow} \rangle = \frac{2}{N} \langle \hat{c}_{\mathbf{r},\uparrow} \hat{c}_{\mathbf{r},\uparrow} \rangle. \quad (S27)$$

where $c_{\mathbf{r},\sigma}^\dagger$ is fermion creation operator at site $\mathbf{r}$ with spin $\sigma$. $\langle \rangle$ denotes the expectation value. By performing the Fourier transformation $c_{\mathbf{r},\sigma} = \frac{1}{\sqrt{N}} \sum_k e^{i \mathbf{k} \cdot \mathbf{r}} c_{\mathbf{k},\sigma}$, we obtain the charge density in $\mathbf{q}$ space,

$$\rho(\mathbf{q}) = \sum_{\mathbf{r}} e^{-i \mathbf{q} \cdot \mathbf{r}} \rho(\mathbf{r}) = \frac{2}{N} \sum_{\mathbf{r} \mathbf{q}} e^{-i \mathbf{q} \cdot \mathbf{r}} \langle \hat{c}_{\mathbf{r},\uparrow} \hat{c}_{\mathbf{r},\uparrow} \rangle = \frac{2}{N} \sum_{\mathbf{q}} \langle \hat{c}_{\mathbf{q},\uparrow} \hat{c}_{\mathbf{q},\uparrow} \rangle = \frac{2}{N} \sum_{\mathbf{q}} \langle \hat{c}_{\mathbf{q},\uparrow} \hat{c}_{\mathbf{q},\uparrow} \rangle. \quad (S28)$$

Using Eqs. (5) in the main text, $\rho(\mathbf{q})$ can be expressed in terms of Bogoliubov quasi-particle $\gamma_{\mathbf{k},\sigma,i}$,

$$\rho(\mathbf{q}) = \frac{2}{N} \sum_{\mathbf{k} \mathbf{j} \mathbf{l}} \sum_{\mathbf{j} \mathbf{l} \mathbf{j} \mathbf{l}} \left( \langle u_{\mathbf{j} \mathbf{l}}(\mathbf{k}) \gamma_{\mathbf{k},\uparrow,j} \rangle \langle u_{\mathbf{l} \mathbf{j}}(\mathbf{k} + \mathbf{q}) \gamma_{\mathbf{k},\uparrow,l} \rangle + \langle v_{\mathbf{j} \mathbf{l}}(\mathbf{k}) \gamma_{\mathbf{k},\downarrow,j} \rangle \langle v_{\mathbf{l} \mathbf{j}}(\mathbf{k} + \mathbf{q}) \gamma_{\mathbf{k},\downarrow,l} \rangle \right) \quad (S29)$$

where we use $\langle \gamma_{\mathbf{k},\uparrow,j} \gamma_{\mathbf{k},\downarrow,j} \rangle = n_F(\mathbf{E}(\mathbf{k})) \delta_{\mathbf{k},\mathbf{k}'}$ and $\langle \gamma_{\mathbf{k},\downarrow,j} \gamma_{\mathbf{k},\uparrow,l} \rangle = n_F(\mathbf{E}(\mathbf{k})) \delta_{\mathbf{k},\mathbf{k}'}$. Here

$$\delta_{\mathbf{k},\mathbf{k}'} = \begin{cases} 1, & \mathbf{k} - \mathbf{k}' = \mathbf{mQ}_1 + \mathbf{nQ}_2, m, n \in \mathbb{Z} \\ 0, & \text{otherwise} \end{cases}$$

Thus, the Fourier transformation of the LDOS, $\rho(\mathbf{q}, \omega)$ reads

$$\rho(\mathbf{q}, \omega) = -\frac{2}{N} \sum_{\mathbf{k} \mathbf{j} \mathbf{l}} \left[ \langle u_{\mathbf{j} \mathbf{l}}(\mathbf{k}) \rangle \langle u_{\mathbf{l} \mathbf{j}}(\mathbf{k} + \mathbf{q}) \rangle \frac{\partial n_F(\omega - \mathbf{E}(\mathbf{k}))}{\partial \omega} + \langle v_{\mathbf{j} \mathbf{l}}(\mathbf{k}) \rangle \langle v_{\mathbf{l} \mathbf{j}}(\mathbf{k} + \mathbf{q}) \rangle \frac{\partial n_F(\omega - \mathbf{E}(\mathbf{k}))}{\partial \omega} \right] \delta_{\mathbf{k},\mathbf{k}+\mathbf{q}}. \quad (S30)$$

Using the folding property, Eq. (S30) can be written into the following form,

$$\rho(\mathbf{q}, \omega) = -\frac{1}{8N} \sum_{\mathbf{k} \mathbf{j} \mathbf{l}} \sum_{i=1}^{16} \left[ \langle u_{\mathbf{j} \mathbf{l}}(\mathbf{k}) \rangle \langle u_{\mathbf{l} \mathbf{j}}(\mathbf{k} + \mathbf{q}) \rangle \frac{\partial n_F(\omega - \mathbf{E}(\mathbf{k}))}{\partial \omega} + \langle v_{\mathbf{j} \mathbf{l}}(\mathbf{k}) \rangle \langle v_{\mathbf{l} \mathbf{j}}(\mathbf{k} + \mathbf{q}) \rangle \frac{\partial n_F(\omega - \mathbf{E}(\mathbf{k}))}{\partial \omega} \right] \delta_{\mathbf{k},\mathbf{k}+\mathbf{q}}. \quad (S31)$$

We can see from Eq. (S31) that $\mathbf{q} = m\mathbf{Q}_1 + n\mathbf{Q}_2, \ (m, n \in \mathbb{Z})$ is necessary for $\rho(\mathbf{q}, \omega)$ being nonzero. Taking into account the symmetry of the system and $\rho(\mathbf{q}, \omega) = \rho^*(\mathbf{q}, \omega)$, there are four possible nonzero $|\rho(\mathbf{q}, \omega)|$ in the BZ,

$$\rho_{A}(\omega) = |\rho(\mathbf{q} = \mathbf{0}, \omega)|, \quad (S32a)$$

$$\rho_{B}(\omega) = |\rho(\mathbf{q} = \pm \mathbf{Q}_1, \omega)| = |\rho(\mathbf{q} = \pm \mathbf{Q}_2, \omega)| = |\rho(\mathbf{q} = \pm \mathbf{Q}_3, \omega)|, \quad (S32b)$$

$$\rho_{C}(\omega) = |\rho(\mathbf{q} = \pm \mathbf{Q}_1, \omega)| = |\rho(\mathbf{q} = \pm \mathbf{Q}_2, \omega)| = |\rho(\mathbf{q} = \pm \mathbf{Q}_3, \omega)|, \quad (S32c)$$

$$\rho_{D}(\omega) = |\rho(\mathbf{q} = \pm (\mathbf{Q}_1 - \mathbf{Q}_2), \omega)| = |\rho(\mathbf{q} = \pm (\mathbf{Q}_2 - \mathbf{Q}_3), \omega)| = |\rho(\mathbf{q} = \pm (\mathbf{Q}_3 - \mathbf{Q}_1), \omega)|. \quad (S32d)$$

$\rho_A(\omega)$ is just the DOS $\rho(\omega)$ we calculated in the main text.
VII. SPONTANEOUS LOOP CURRENT AT GROUND STATE

The following formula of nearest neighbor current \( J_{i,j} \) is adopted to study the loop current at ground state,

\[
J_{i,j} = \sum_{\sigma} \left\langle i \left( c^\dagger_{r,i,\sigma} c_{r,j,\sigma} - c^\dagger_{r,j,\sigma} c_{r,i,\sigma} \right) \right\rangle \\
= \frac{2i}{N} \sum_{k,k'} \left( e^{-ik \cdot r_i} c^\dagger_{k,i,\uparrow} c_{k',j,\uparrow} - e^{-ik' \cdot r_j} c^\dagger_{k,j,\uparrow} c_{k',i,\uparrow} \right) \\
= \frac{2i}{N} \sum_{k,q} e^{iqr} \left( e^{i(k+q) \cdot \delta r} - e^{-i(k-q) \cdot \delta r} \right) \left( c^\dagger_{k,i,\uparrow} c_{k+q,j,\uparrow} \right),
\]

where \( \delta r = r_j - r_i \).

As we do in the calculation of LDOS above, \( J_{i,j} \) can be expressed in terms of Bogoliubov quasi-particle \( \gamma_{k,\sigma,j} \) according to Eqs. (5),

\[
J_{i,j} = \frac{2i}{N} \sum_{k,q} e^{iqr} \left( e^{i(k+q) \cdot \delta r} - e^{-i(k-q) \cdot \delta r} \right) \left[ u_{ij}(k)u^*_{ij}(k+q) n_F \left( E(k)_j \right) + v_{ij}(k)v^*_{ij}(k+q) n_F \left( E(k)_j \right) \right] \delta_{k,k+q}. \tag{S33}
\]

for a given \( k \), there are 16 \( q \) in the first BZ that can give a nonzero contribution to the summation \( \sum_q \), according to the factor \( \delta_{k,k+q} \).

Using Eq. (S33), we calculate the loop current in the 4 \( \times \) 4 enlarged unit cell at \( \frac{k_B T}{\Delta} = \frac{1}{10} \), the result is shown in Fig. S3. We can see from Fig. S3 that the ground state shows spontaneous loop current.

**FIG. S3.** Spontaneous loop current \( |J_{i,j}| \) at ground state at \( \frac{k_B T}{\Delta} = 1/10 \). The red arrows show the directions of currents and the circles at the corresponding links represent the intensity. Here \( \Lambda = 0.1, \Delta = 0.02, \phi_0 = \pi/2, \theta_1 = 0, \theta_2 = 2\pi/3 \) and \( \theta_3 = -2\pi/3 \) have been chosen.