Frustrated superconductivity and sextetting order

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The superconducting state typically favors a uniform spatial distribution akin to ferromagnetism. Nevertheless, the pair-density-wave state exhibits sign changes in the pairing order, leading to potential frustrations in phase coherence. We propose a mechanism to the sextetting order stemming from the frustrations in the phase coherence of a pair-density-wave state, whose spatial modulation manifests a vortex-antivortex honeycomb lattice. The classical ground state configurations are mapped to Baxter’s three-coloring model, revealing a macroscopic degeneracy accompanied by extensive entropy. The phase coherence problem intertwines the U(1) phases and the vorticity variables. While the resultant color and phase fluctuations suppress the pair-density-wave order, they maintain the sextetting order above the superconducting transition temperature ($T_c$). The 1/3-fractional vortex emerges as the fundamental topological defect in the sextetting order. This novel mechanism of frustrated superconductivity provides an alternative explanation for the experimental observed fractional oscillations in CsV$_3$Sb$_5$.

I. INTRODUCTION

The multi-fermion ordering has received considerable attention in a wide range of research fields in modern physics. Inside a heavy nucleus, the alpha-particle-like quartetting instability involving two protons and two neutrons competes with the deuteron-like instability of pairing between one proton and one neutron $^1$. Its many-body version has been investigated in the 1D spin-$\frac{3}{2}$ ultra-cold fermion system via bosonization, revealing the Ising-dual relation between these two instabilities $^2$. In the context of superconductivity (SC) in electron systems, the multi-fermion ordering, quartetting or sextetting, can emerge as a vestigial secondary order of the charge-2e Cooper pairing SC above the superconducting transition temperature $T_c$, often termed as the so-called charge-4e, or, charge-6e SC states $^3,^4$. The quartetting or sextetting states can appear as a consequence of melting the pair-density-wave (PDW) state, i.e., the superconducting pairing orders $\Delta_{Q}$ carrying finite momenta $^6$ -$^8$. A PDW state can exhibit multiple incommensurate wave vectors $Q$, and the incommensurabilities render the relative phases of the translational degree of freedom gapless. There exist topological defects related to the translational phase, and the system undergoes a Kosterlitz-Thouless (KT) type transition $^{17}$ as temperature increases. The PDW superconducting order disappears above a critical temperature $T_c$ due to the strong fluctuations in the relative phase channels, nevertheless, the overall phase remains ordered, leading to the quartetting or sextetting state.

Here we propose an alternative mechanism based on the frustrations of the superconducting phase coherence, which naturally leads to the sextetting order. Usually the frustrated systems are characterized by the macroscopic degeneracy of ground-state configurations at the classic level $^{18,19}$. The three-coloring model on the honeycomb lattice $^{20}$ is a celebrated example, which applies to various frustrated systems including classical antiferromagnets $^{21}$, Josephson junction arrays $^{22,23}$, orbital-active materials $^{24,25}$, and other systems $^{11,27,28}$. Each bond of the honeycomb lattice is painted by one of the three colors $R$ (red), $G$ (green), $B$ (blue) under the so-called “color constraint” that three bonds connected to the same vertex are pained by different colors. The classical ground-state configurations exhibit a macroscopic degeneracy, leading to an extensive entropy of 0.38$k_B$ per hexagon $^{20}$. The strong frustrations kill the order of the single color variable, but a combined three-color order could survive. This frustration mechanism has also been extended to the four-coloring model on the three-dimensional diamond lattice $^{29}$, which exhibits the dipolar correlation characterized by a cubic power decay $1/R^3$.

In this article, we investigate the sextetting order in the superconductivity via frustrations to the PDW order. The corresponding superconducting phase coherence problem is mapped to the three-coloring model, exhibiting macroscopic degeneracy arising from color configurations. The low-energy degrees of freedom encompass the local pairing-phases coupled to the vorticity variables. Monte-Carlo simulations are performed to obtain the phase diagram, showing the competition between the Cooper pairing and sextetting order. The entropy associated with the color configurations dominates, leading to
the sextetting state at a higher temperature before the system enters the completely disordered regime. Moreover, the sextetting order manifests $1/3$-fractional vortex, providing an alternative mechanism to the fractional flux oscillations.

The proposed mechanism of sextetting order through frustrated superconductivity in the PDW state differs from previous scenario \cite{8,31}. Previous analysis primarily focused on the generation of fractional vorticities due to the phase winding among the relative phases, which only involve a few superconducting gap functions. In this article, frustration of phase coherence problem in the vortex/antivortex lattice is related to the macroscopic number of degrees of freedom from the vortices/antivortices. The number of degenerate states at the classic level grows up exponentially as the lattice size increases, which yields extensive entropy.

II. VORTEX-ANTIVORTEX HONEYCOMB LATTICE

Frustrated superconductivity emerges from phase coherence of a vortex-antivortex honeycomb lattice. The vortex lattice model could be realized through Josephson junction arrays \cite{22}, $p$-orbital unconventional superfluidity \cite{30} or PDW state \cite{8,31}. The cores of single vortices/anti-vortices are located at the sites of the honeycomb lattice, with phase winding $\pm 2\pi$ around it. A pair of neighboring single vortices couple with each other through Josephson coupling and inter-vortex coupling. In the ground state, all the vortices and antivortices exhibit phase coherence, leading to unconventional superconductivity with simultaneously time-reversal symmetry breaking \cite{30}. A typical vortex-antivortex pattern with alternating vortices and anti-vortices is illustrated in Fig. 1(a). Additionally, the superconducting state induced by a fixed coherent arrangement of vortices and anti-vortices can exhibit spatial modulation, giving rise to the pair-density-wave order.

The vortex-antivortex model can be mapped to the three-coloring problem on the honeycomb lattice \cite{22,30}. (Its generalization to the four-coloring problem in the diamond lattice was done by Chern and Wu \cite{29}.) The superconducting phase winds $\pm 2\pi$ around a site $j$ of the honeycomb lattice, depending on whether a vortex or antivortex is located at that site. Each site is connected to its three nearest neighbors and the angles between two bond directions are all 120°. Hence, to minimize the phase difference across each bond, without loss of generality, we can assign three colors $R, G, B$ corresponding to the phases $1, e^{i\frac{2\pi}{3}}, e^{i\frac{4\pi}{3}}$, respectively, and paint each bond according to this color constraint. The clockwise or anticlockwise ordering of the colors $R, G, B$ defines the chirality $\tau_j = \pm 1$, or, the vorticity. Under time-reversal transform, the vorticities are flipped. If only considering the color frustration effect, the charge-2e correlation of the phases exhibits power-law decay, $G_2(r) \sim r^{-4/3}$ \cite{21}.

To fully describe the phase coherence of the superconducting state, it is essential to account for U(1) phase fluctuations. For later convenience, the following convention is employed: The honeycomb lattice system is divided into two sublattices $A$ and $B$, as depicted in Fig. 1(b). Considering a site $j$ belonging to sublattice $A$, it emits three bonds denoted as $\hat{e}_j$ whose azimuthal angles are $90°$, $210°$, $330°$ for bond index $\nu = 1, 2, 3$, respectively. The superconducting phase along the $\hat{e}_j$ direction is denoted as $\theta_j$. On the other hand, for a site $k$ belonging to sublattice $B$, its superconducting phase along $-\hat{e}_j$ direction is set as $\theta_k$. Then around any site $j$, the superconducting phase along the bond $\nu$ is represented as $\theta_j + \tau_j \phi_{\nu}$, where $\phi_{\nu} = 0, \frac{2\pi}{3}, \frac{4\pi}{3}$ for $\nu = 1, 2, 3$, respectively. Then the effective Hamiltonian reads \cite{30},

$$H = -J_1 \sum_{j \in A, \nu} \cos \left[ \theta_j - \theta_k + (\tau_j - \tau_k) \phi_{\nu} \right]$$

$$-J_2 \sum_{j \in A, \nu} \cos \left[ \theta_j - \theta_k + (\tau_j - \tau_k) \phi_{\nu} + \frac{\pi}{2} \tau_j + \tau_k \right],$$

where the summation over $j$ is only carried on sublattice $A$, and $k$ belongs to sublattice $B$ along the bond direction of $\hat{e}_\nu$.

The $J_1$-term represents the nearest-neighbor Josephson coupling, whose dependence on the bond geometry can be viewed as a vector potential $A_{jk} = (\tau_j - \tau_k) \phi_{\nu}$. $J_1$-term characterizes the phase coherence along the bond direction, enforcing the "color constraint" within the classical ground state. The $J_2$-term coupling arises from the non-trivial winding numbers, or angular momenta, of the vortices, similar to the phase coherence problem encountered in orbital physics \cite{30}. This term reflects the phase coherence along the bisection of the bond between two nearest-neighbor vortices. Typically, in realistic systems, $J_2$ is much smaller than $J_1$-term. A slight $J_2$-term lifts the degeneracy among color configuration and stabilizes the vortex configuration, fixing it into an antiferromagnetic (AFM)-like staggered vortex-antivortex pattern. In this pattern, adjacent neighbors display oppos-
FIG. 2. A schematic phase diagram of the effective model in Eq. (1). At low temperatures \( T < T_c \), 2e quasi-long-range order (QLRO) dominates \( (\eta_{2e} < \eta_{6e}) \), while at intermediate temperatures \( T_c < T < T_{KT} \), 6e QLRO takes precedence. Above the KT temperature \( T_{KT} \), the system is in the disordered phase.

If the temperature \( T \) is lower than the KT transition temperature \( T_{KT} \) of the U(1) phase \( \theta_i \), but is still high enough such that the inter-vortex interactions \( J_2 \)-term can be neglected, then the entropy of the color configurations dominates. As a result, there is no long-range pairing order, but the cube of the pairing order is long-range ordered since color variables \( 1, \omega, \omega^2 \) are cubic roots of 1 with \( \omega = e^{\frac{2\pi i}{3}} \). This manifests as the sextetting superconductivity. As the temperature is lowered enough, the inter-vortex \( J_2 \) coupling will lift the degeneracy among different vortex configurations. The ordered PDW configuration emerges, resulting in the transition to the charge-2e superconducting state. A schematic phase diagram of the system is summarized as in Fig. 2(a).

III. NUMERICAL SIMULATION

Monte-Carlo simulations are performed to investigate the Hamiltonian of Eq. (1). Importantly, the method of updating configurations depends on the relevant temperature regime. Similar situation appears in many frustrated systems, e.g., in the Ref. [27]: The acceptance ratio of local updates becomes vanishingly small at low temperatures which cannot sample the macroscopic degeneracy of the ground states. The same strategy in Ref. [27], will be followed by separating the temperature into lower and higher regimes. Above the KT temperature of the U(1) phase \( \theta_i \), 1/3-fractional vortices proliferate and the U(1) phase coherence is destroyed, which also disorders the color configurations. The U(1) phase correlation, or charge-2e correlation, \( \langle e^{i \theta_j} e^{-i \theta_k} \rangle \) between two sites \( j, k \) decays exponentially as the relative distance increases. The local update of the chiralities and phase variables works well in the disordered phase. As temperature decreases, the vorticity configuration tends to satisfy the color constraint and the local update breaks down [27]. Alternatively, we apply the loop update in which the color constraint is maintained in the vorticity configuration in each update. The U(1) phase variables are updated simultaneously according to the vorticity configuration. (see App.A for the details).

The simulations to the scaling dimensions for the superconducting phase correlations in the low temperature regime are summarized in Fig. 3(a). It shows two competing phases: the lower temperature charge-2e superconductivity, and the higher temperature charge-6e dominated phase. Correlations of both orders are characterized by the quasi-long-range ordering (QLRO) of the relevant phase correlations defined as

\[
\langle e^{i \theta_j} e^{-i \theta_k} \rangle \sim r^{-\eta_{2e}}, \quad \langle e^{3i \theta_j} e^{-3i \theta_k} \rangle \sim r^{-\eta_{6e}},
\]

respectively, where \( r \) is the distance between the two sites \( j, k \). Here, \( \eta_{2e} \) and \( \eta_{6e} \) are exponents for the charge-2e and 6e correlations, representing the scaling dimensions of the orders. For comparison, the exponent \( \eta_{4e} \) of the 4e-correlation, \( \langle e^{2i \theta_j} e^{-2i \theta_k} \rangle \sim r^{-\eta_{4e}} \), is also deduced from the numerical estimation. To suppress finite size effect and obtain the power-decay exponents more precisely, the numerical estimation. To suppress finite size effect and obtain the power-decay exponents more precisely, the numerical estimation.

The three exponents fit well with the following relations,

\[
\eta_{2e} = \eta_c + \eta_{6e}/9, \quad \eta_{4e} = \eta_c + 4\eta_{6e}/9.
\]

Here, \( \eta \) represents the contribution from color frustration in the vorticity correlation, which does not contribute to the charge-6e correlation. At low enough temperatures, the vortex configuration is fixed with \( \eta_c = 0 \) and the exponents arise from the QLRO of the U(1) phase variables, exhibiting \( \eta_{2e} = \eta_{4e}/4 = \eta_{6e}/9 \), as depicted in Fig. 3(b). It is indicated that charge-4e is never the leading correlation. The most dominant correlation is either charge-2e at low temperatures, or charge-6e at high temperatures.
The transition from charge-2e to charge-6e is driven by the competition between the energy consideration and entropy contribution. At lower temperatures, energetic minimization stemming from the combination of $J_1$ and $J_2$ terms favors the AFM-like staggered ordering of the vortex/anti-vortex configuration. The low-temperature regime is characterized by the dominance of charge-2e behavior ($\eta_{2e} < \eta_{6e}$). As temperature increases, color frustration begins to manifest. Configurations satisfying the three-coloring constraint minimize $J_1$-term ($J_2$ is much smaller than $J_1$). The number of such kind of configurations scales exponentially as increasing the system size, exhibiting an extensive entropy. The scaling dimensions of 2e and 6e correlations intersect at a transition temperature $T_c$. Above $T_c$, but still below the KT temperature, the entropy associated with color configurations takes precedence over the energy cost arising from the $J_2$-term and the charge-2e correlation is suppressed. The charge-6e behavior dominates over the charge-2e correlation, as evidenced by the relationship $\eta_{6e} < \eta_{2e}$. Moreover, the estimated color correlation $\eta_e$ is much smaller than 4/3 for the pure three-color model[21]. This is because the degeneracy among different color configurations is broken by the $J_2$ term.

The behaviors of the scaling dimensions related to 2e, 4e and 6e order under temperature for smaller $J_2$ exhibit qualitatively consistent behavior as in Fig. 3, see simulations for $J_2/J_1 = 0.015, 0.02$ in App. C. As $J_2$-term increases, the energy cost lifting the degeneracy of the color configurations increases, leading to an enhancement of $T_c$. When the $J_2$-term becomes large enough, the energetic consideration could always outweigh the entropy contribution, resulting in the absence of charge-6e QLRO, as depicted in the simulation result for $J_2/J_1 = 0.03$ in App. C. In reality, $J_2$ coupling along the bisection line of the bond is always exists, while its amplitude is typically much smaller than $J_1$ term, aligning with the scenario depicted in Fig. 2.

Above the KT temperature $T_{KT}$ of the U(1) phase variables $\theta_i$, both charge-2e and 6e correlations exhibit exponential decay, leading the system into a disordered phase. The numerical simulation confirms the exponentially decay behavior of the correlations in this regime. However, due to limitations in Monte Carlo simulation, as mentioned earlier, precise determination of the KT temperature is not feasible. As the temperature decreases from high to the critical temperature $T_{KT}$, the effectiveness of local updates gradually diminishes, reflected in rapidly increasing acceptance rates and computation times. The temperature at which the local updates become ineffective is approximately around $T_0 \approx 0.13$, establishing an upper bound for $T_{KT}$. This kind of numerical difficulty is common in frustrated system [27]. More advanced method is necessary for completely simulating the problem, which is left for further works.

The preceding analysis focuses on the inherently two-dimensional system, where charge-2e and charge-6e correlations exhibit only quasi-long-range order. In the real materials, it is natural that the system comprises a layered 2D lattice, thereby forming a quasi-two-dimensional model. The inter-layer couplings between the 2D layers enhance the QLRO, stabilizing a true long-range order of either the charge-2e type (for $\eta_{2e} < \eta_{6e}$ regime), or, charge-6e type (for $\eta_{6e} < \eta_{2e}$ regime) at low temperatures.

The $\frac{1}{3}$-fractional flux can emerge above the $T_c$ of the PDW order due to the vortex configuration fluctuations [23, 29]. Around each hexagonal vortex/antivortex plaquette $h$, its total vorticity is defined as $\Phi_h = \frac{1}{3} \sum_i T_i$. For a configuration satisfying the color constraint, the vorticity around a plaquette takes the values of 0, ±2. A fundamental topological defect exhibits a domain wall which is a string of bonds with mismatched color variables. The starting and the ending points of a domain wall are located in a plaquette as the core of a $\frac{1}{3}$-vortex, or, a $\frac{1}{3}$-antivortex, respectively. Around the vortex cores, the superconducting phase winding equals to $\pm \frac{2}{3}\pi$, respectively. An external magnetic flux at 1/3 of the fundamental flux generates a $\frac{1}{3}$-vortex such that the superconducting phase across each bond remains matched. Consequently, the free energy will show periodic modulation under the flux change of

$$\Phi_{6e} = \frac{1}{3} \Phi_{2e} = \frac{hc}{6e},$$

exhibiting the charge-6e QLRO and 1/3-fractional flux.

IV. PDW STATES

PDW states exhibiting spatial modulation of the pairing order have been proposed in the FFLO states [32, 33]. The vortex-antivortex model provides a platform for realizing the PDW state as its ground state. In the hexagonal crystal system, PDW state can manifest 6 commensurate wave vectors denoted as $\pm Q_i$ ($i = 1, 2, 3$). The spatial distribution of PDW order parameter is represented as [8],

$$\Delta(r) = \sum_i (\Delta_{+Q_i} e^{iQ_i \cdot r} + \Delta_{-Q_i} e^{-iQ_i \cdot r}),$$

where the gap functions $\Delta_{\pm Q_i}$ ($i = 1, 2, 3$) carry nonzero mass-of-center momentum $\pm Q_i$, respectively. The ground state configuration can be determined by the Ginzburg-Landau free energy [8], presenting several possibilities depending on the microscopic details of the relevant model.

For the previously considered AFM-like vortex-antivortex pattern, it corresponds to an inversion symmetric breaking PDW state with only three wavevectors $Q_1, Q_2$ and $Q_3$. The superconducting order parameter is given by

$$\Delta_{3Q}(r) = \Delta_{3Q} (e^{iQ_1 \cdot r} + e^{iQ_2 \cdot r} + e^{iQ_3 \cdot r}),$$

where the gap functions $\Delta_{\pm Q_i}$ ($i = 1, 2, 3$) carry nonzero mass-of-center momentum $\pm Q_i$, respectively.
with relative phases $\theta_1, \theta_2$ between the three PDW components. This 3Q state forms a honeycomb lattice with alternating vortices/antivortices as illustrated in Fig. 4(a) [8]. On the other hand, when inversion symmetry is preserved, the 6Q PDW order parameter exhibits all the six momenta and a typical chiral PDW pattern is depicted in Fig. 4(b) [8, 31], where a double vortex/anti-vortex appears in plaquette centers. The 3Q state forms a honeycomb lattice, and the color constraint is updated as that of the hexagons.

In the STM experimental detection of the PDW state, the local density of state (LDOS) exhibits extra spatial comb lattice is formed from single vortices or antivortices. This 3Q state forms a honeycomb lattice with double antivortices (or vortices) in the centers of the hexagons.

$\Phi = 2 \pi \chi \frac{\pi}{4} \left( \theta_1 + \theta_2 \right)$

The LDOS measurement from a STM experiment can not distinguish the 3Q PDW state from the 6Q state one up to this order, since both of them exhibit six peaks in the Fourier spectrum of LDOS.

The difference between the LDOS patterns of the 3Q and 6Q states arises from the couplings among PDW components. A pair of two PDW components contribute to the LDOS,

$$N_{Q_i} (r) \propto \Delta_0 \Delta_Q e^{iQ \cdot r} + \Delta_0 \Delta_Q e^{-iQ \cdot r}.$$  

The above idea of frustrated superfluidity can also be extended to three dimensions. The three-coloring model is generalized to the 4-coloring model defined on a diamond lattice, and the color constraint is updated as that all the four bonds connected to a lattice site should be painted by the R, G, B, Y colors without repetition [29]. The R, G, B, Y colors represent the quartic unit roots $1, i, -1, -i$ respectively. The frustration therein could favor a charge-8e state.

It is worth noting that the phenomenon of quantum quartetting, or charge-4e, order has been investigated in the previous study within the context of one-dimensional spin-$\frac{3}{2}$ fermionic system [2]. A ground state featuring charge-4e order driven by quantum fluctuations was explicitly identified in certain parameter regimes. It competes with the charge-2e pairing as tuning interaction parameters. Since the charge-4e ordering can be viewed as pairing of pairs, the transition between charge-2e and 4e orderings is Ising-like, i.e., these two phases are Ising-dual to each other. The model proposed in Eq. (1) could also be augmented to the quantum version, for example, by adding the on-site Bose Hubbard interaction. Typically, “order-from-disorder” mechanism will select the charge-2e ordering state in $(2+1)$-dimensions even in the absence of $J_2$ term. It would be interesting to study

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$N_{Q_i} (r) \propto \Delta_0 \Delta_Q e^{i(Q_i-Q_j) \cdot r} + h.c.$  

For the 3Q state, the LDOS pattern exhibits peaks corresponding to the wavevector $\pm (Q_i - Q_j)$ with $i \neq j = 1, 2, 3$. For the 6Q state, several extra peaks $\pm 2Q_i$ ($i = 1, 2, 3$) different from the 3Q state appear. Such differences could be helpful to distinguish these two PDW states.

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how to further enhance quantum fluctuations to achieve the quantum sextetting order in the future studies.

VI. CONCLUSIONS

We study the phase frustrations in the vortex-antivortex model on the honeycomb lattice by mapping its superconducting phase coherency problem to the three coloring model. The fundamental degrees of freedom are described by the U(1) phases coupled to the discrete vorticity variables. The classical ground states, subject to the color constraints, exhibit a macroscopic degeneracy in terms of coloring patterns. The inclusion of inter-vortex coupling $J_z$ results in a phase coherent ground state pattern that naturally realizes the pair-density-wave state. Above the superconducting transition temperature ($T_c$), the sextetting (charge-6e) order prevails over the pairing (charge-2e) order, persisting until the system enters a disordered regime. Such behavior is further numerically verified through Monte Carlo simulations. The fundamental degrees of freedom are described by the U(1) phases coupled to the discrete coloring model. The quantum sextetting order in the future studies.

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In this appendix, we provide a detailed explanation of the combined loop update method employed in the low-temperature Monte Carlo simulations. At exceedingly low temperatures, the system primarily manifests states within the three-color subspace, adhering to color configurations subjected to color constraints. To effectively capture the system’s behavior in such conditions, we would confine the simulation to the three-color subspace in this temperature regime.

For the implementation of the combined loop update method, we initially define loops within the three-color subspace. These loops are generated by traversing a path with two alternating colors, such as $RGRG\cdots$, as depicted in Fig. A1(b). Importantly, the interchange of the two colors along the loop does not violate the three-color constraint. In the loop update, a specific loop is selected, and colors along its path are interchanged, effectively flipping the chiralities along the loop.

Simultaneous updates of the U(1) phases are also necessary for the sufficiency of the simulation. During the loop update, the chiralities along the loop $L$ are flipped, and the local U(1) phases $\theta_i(i \in L)$ are altered concurrently. It is crucial to note that each site $i \in L$ within the loop possesses one external bond that is not part of the loop, featuring a phase coupling denoted as $\varphi_{i,\text{out}}$ at the bond. The transformation of $\theta_i(i \in L)$ under the update is determined while keeping the external phase $\varphi_{i,\text{out}}$ fixed.

\begin{equation}
\varphi_{i,\nu=\text{out}} = \theta_i^{(0)} + \phi_{i,\text{out}} \tau_i^{(0)} = \theta_i^{(1)} + \phi_{i,\text{out}} \tau_i^{(1)} \quad (A1)
\end{equation}

where $(\theta_i^{(0)}, \tau_i^{(0)})$ and $(\theta_i^{(1)}, \tau_i^{(1)})$ are phase variables and chiralities before and after the loop update. All the chiralities along the loop are flipped, $\tau_i^{(1)} = -\tau_i^{(0)}$, and we can find the updated phase $\theta_i^{(1)}$ as

\begin{equation}
\theta_i^{(1)} = \theta_i^{(0)} + 2\phi_{i,\text{out}} \tau_i^{(0)} \quad (A2)
\end{equation}

In total, for each loop $L$, we use the following updates for the color configuration,

\begin{equation}
i \in L : \begin{cases}
\tau_i^{(0)} & \rightarrow \tau_i^{(1)} = -\tau_i^{(0)} , \\
\theta_i^{(0)} & \rightarrow \theta_i^{(1)} = \theta_i^{(0)} + 2\phi_{i,\text{out}} \tau_i^{(0)} \quad (A3)
\end{cases}
\end{equation}
APPENDIX B. FINITE SIZE EFFECT AND FITTING OF THE SCALING DIMENSIONS

In the Monte-Carlo simulation, the correlation functions for 2e, 4e, and 6e are numerical evaluated at different lattice sizes \( L = 12, 18, 24, 30, 36, 42, 48 \). For each size, the correlation functions are calculated based on the following formula,

\[
\begin{align*}
2e : & \quad F_{2e}(m; L) = \frac{1}{N} \sum_j \langle e^{i\theta_j} e^{-i\theta_{j+m}} \rangle, \\
4e : & \quad F_{4e}(m; L) = \frac{1}{N} \sum_j \langle e^{2i\theta_j} e^{-i2\theta_{j+m}} \rangle, \\
6e : & \quad F_{6e}(m; L) = \frac{1}{N} \sum_j \langle e^{3i\theta_j} e^{-i3\theta_{j+m}} \rangle,
\end{align*}
\]

where \( j \) is the lattice site, \( m \) is the relative distance and \( N = L^2 \) is the total number of lattice sites.

The simulated results \( F(m; L) \) for a fixed size \( L \) suffer strong finite size effect for larger \( m \) close to \( L \) and the directly fitting of exponents would not be good. Instead, for each lattice size \( L \), the value of the correlation functions at the furthest distance, \( r = (L/2, L/2) \), is selected, \( F(r) \equiv F(r; L) \). The correlation exponents are then fitted within \( F(r) \) for different lattice sizes. This approach effectively reduces finite size effect on the calculations when simulating exponents. For example, two typical fitting functions \( F(r) \) at \( T = 0.032 \) and \( T = 0.064 \) for \( J_1 = 1 \) and \( J_2/J_1 = 0.01 \) are depicted in Fig. A2. At smaller temperature \( T = 0.032 \), 2e correlation roughly exhibits larger absolute value and smaller power-law decaying exponent than that of 6e correlation. The system exhibits the 2e-QLRO state. On the contrary, at larger temperature \( T = 0.064 \), the above relations reverse and the system exhibits the 6e-QLRO state.

APPENDIX C. BEHAVIOR OF EXPONENTS AT DIFFERENCE \( J_2 \)

The simulations to the power-law decay exponents related to 2e, 4e and 6e correlations under temperature for \( J_2/J_1 = 0.015, 0.02 \) are depicted in Fig. A3. These results exhibit qualitatively consistent behavior with \( J_2/J_1 = 0.01 \) depicted in Fig. 2. At low temperature below \( T_c \), due to the dominance of energy consideration from \( J_2 \)-term, 2e-QLRO wins over 6e-QLRO, indicated by the lower scaling dimension \( \eta_{2e} < \eta_{6e} \). When \( T_c < T < T_K T \), entropy contribution from color frustration wins over energetic consideration, leading to the 6e-QLRO with dominant instability, \( \eta_{6e} < \eta_{2e} \). The transition temperature \( T_c \) becomes larger as \( J_2 \) increases, due to the increased energy cost deviation from the staggered vortex/antivortex configuration.

When \( J_2 \) surpasses a critical value, the energy cost associated with it could always outweigh the entropy contribution from color frustration across the entire temperature regime, resulting in the absence of intermediate charge-6e QLRO. For example, for \( J_3 = 0.03J \), the calculated scaling dimensions are depicted in Fig. A4. Here, 2e-QLRO exhibits the dominant instability, indicated by the lowest scaling dimension \( \eta_{2e} \) in a broad temperature regime below \( T_K T \). Consequently, only the charge-2e
FIG. A5. A schematic phase diagram of the effective model in Eq. (1) with respect to the $J_2/J_1$ and $T/J_1$. The state is expected to persist above a certain $J_2$ value at low temperature.

The schematic phase diagram of the effective model Eq. (1) is depicted in Fig. A5. In the regime of weak $J_2$, charge-6e order emerges within an intermediate temperature regime, $T_c < T < T_{KT}$. As $J_2$ increases, $T_c$ rises due to the enhanced energy cost resulting from it. The KT temperature $T_{KT}$ also rises with increasing $J_2$. This phenomenon aligns with the prediction of KT transition, where KT temperature is directly proportional to the superfluid stiffness [17]. In the model described by Eq. (1), both the $J_1$-term and $J_2$-term contribute to superfluid stiffness in the low-energy theory, consequently leading to a larger $T_{KT}$ as $J_2$ increases when $J_1$ remains fixed. It’s important to acknowledge the limitations of Monte Carlo simulations in capturing the complexities of frustrated systems. A comprehensive establishment of the phase diagram is left for subsequent investigations, potentially employing advanced techniques such as state-of-the-art tensor-network methods.