Higgs-Leggett mechanism for the elusive 6e superconductivity observed in Kagome vanadium-based superconductors

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Abstract
A recent Little-Parks experiment [1] on Kagome-structured superconductor CsV₃Sb₅ [2–5] demonstrated remarkable resistance oscillations with period \( \phi_0/3 = \hbar c/6e \), one-third of the flux quantization \( \phi_0 \) [6–12]. In the present work, we perform analysis based on a Ginzburg-Landau free energy functional involving bilinear Josephson-type couplings between 2e SC order parameters at the three reciprocal lattice vectors connecting M points of the hexagonal Brillouin zone of the material. In a ring geometry we unveil that, as a series of metastable states, phase of one SC order parameter winds \( 2\pi \) more or less than the other two ones around the ring, which yields local free-energy minima at integer multiples of \( \phi_0/3 \). Intriguingly, these states are stabilized by a Higgs-Leggett mechanism [13,14], which induces domain walls (DW) between domains with \( Z_2 \) chirality defined by phases of the three SC order parameters [14–17]. At low
temperatures DW are expelled from the system resulting in free energy minima only at integer multiples of $\phi_0$. The present theory explains successfully the $6e$ SC observed at intermediate temperatures slightly above the bulk transition point for vanishing zero-field resistance and the re-assertion of $2e$ SC at low temperatures in the recent experiment, and thus provides a reasonable starting point for upcoming exploration on the rich physics taking place in the Kagome vanadium-based superconductors.

**Introduction**

Quantization of magnetic flux $\phi_0 = hc/2e$ is the hallmark of superconductivity (SC) as a global quantum state of electron pair condensation [6–11] (for a textbook see [12]). As shown first by Little and Parks [18], in a ring-shaped superconductor the SC transition temperature and resistance between two electrodes oscillate as functions of applied magnetic flux through the ring with period $\phi_0$. Very recently, in the new Kagome vanadium-based superconductors CsV$_3$Sb$_5$ [2–5,19–24] a Little-Parks experiment demonstrated clear resistance oscillations of period $\phi_0/3$, hinting existence of a fantastic six electrons ($6e$) SC state at intermediate temperatures slightly above the bulk transition point for vanishing zero-field resistance [1]. This remarkable observation raises several questions which may be intimately related with the fundamental understanding on SC [25]: Does the period $\phi_0/3$ of magneto resistance imply a condensate of six electrons ($6e$) distinct from the $2e$ Cooper pairs? How does the Kagome structure play the role in this phenomenon, and is it possible that Cooper pairs at three important reciprocal lattice vectors of the Kagome structure interweave with each other in a unique way to give birth to the never-seen-before period $\phi_0/3$? After all, can and in what sense this phase be considered a SC state, and how does the ordinary $2e$ SC state re-assert itself at lower temperatures where zero-field resistance drops to zero?

Here, we propose a Ginzburg-Landau (GL) free energy functional involving bilinear Josephson-type couplings between $2e$ SC order parameters (see Methods) at the three
reciprocal lattice vectors connecting M points of the hexagonal Brillouin zone of the material. Based on analysis of the free energy functional on a ring geometry mimicking the Little-Parks setup, we find that, in a series of metastable states, phase of one SC order parameter winds $2\pi$ more or less than the other two ones around the ring, which yields local free energy minima at integer multiples of $\phi_0/3$ starting from zero applied magnetic flux, responsible for the magneto resistance oscillations with period of $\phi_0/3$ observed in the recent Little-Parks experiment for CsV$_3$Sb$_5$ [1]. As the mechanism of this remarkable phenomenon, in the competition between the Higgs mode [26,27] and the Higgs-Leggett mode [13,14], the latter wins and stabilizes domain walls (DW) of $Z_2$ chirality domains defined by phases of the three SC order parameters [14–17]. Expelling these DW from the system at low temperatures brings the system to its ground state where the phases of three SC order parameters are locked to each other and behave in the same way as a single SC order parameter, yielding free energy minima only at integer multiples of $\phi_0$ and conventional Little-Parks signals. The DW manifest themselves as dislocations and other types of defects, such as compression and shear distortions, in the pair density wave (PDW) lattices [5,28–32]. The present theory explains successfully the main results of the recent Little-Parks experiment [1], namely the 6e SC state observed at intermediate temperatures slightly above the bulk SC transition point and the re-assertion of 2e SC at low temperatures, and is expected to provide a reasonable starting point for upcoming exploration on the rich physics hosted by the Kagome vanadium-based superconductors.

**Results**

*Theory based on GL free energy functional.* We consider a GL free energy functional of three SC order parameters

$$G = \sum_{j=1,2,3} \left[ |a_j| \psi_j|^2 + \frac{b_j}{2} |\psi_j|^4 + \frac{1}{2m_j}\left(\frac{\hbar}{i} (\mathbf{v} - \frac{2e}{c} \mathbf{A}) \psi_j \right)^2 \right] - \sum_{j,k=1,2,3; j<k} \gamma_{jk} (\psi_j^* \psi_k + c. c.) + \frac{1}{8\pi} (\nabla \times \mathbf{A} - \mathbf{H})^2 \quad (1)$$
where due to the Kagome lattice symmetry $a_j = a$, $b_j = b$, $m_j = m$ and $\gamma_{jk} = \gamma$ for $j, k = 1, 2, 3$, and $a > 0$ ($a < 0$) above (below) the bare transition point of a single SC component, $b, m > 0$ and $\gamma < 0$. The mean-field SC transition point $T_{\text{mf}}$ of the system including the inter-component Josephson couplings is given by $a + \gamma = 0$ [14], below which the order parameters take finite amplitude $|\psi_j| = \sqrt{-(a + \gamma)/b}$, and the two coherence lengths and the penetration depth of magnetic field diverge. In the present work, the three SC order parameters are related to the three reciprocal lattice vectors of the Kagome lattice as discussed for pair density wave (PDW) [28,31,32]. It is worthy noticing that the bilinear Josephson-type couplings including their sign are crucial for the $Z_2$ chiral SC state discussed in systems with three and more components [14–17] (for more works see the review article [33]), which are missing in previous discussions on PDW states.

In order to simulate the Little-Parks experiment we consider a SC ring with magnetic flux penetrating as shown schematically in Fig. 1(a). For clarity we mainly consider the small ring width as compared with the ring radius (similar results are available for wider ring). Performing numerical calculations we look for stable and/or metastable states of the system when the applied magnetic flux is swept below the mean-field transition temperature $T_{\text{mf}}$ (with $a + \gamma < 0$). As displayed in Fig. 1(b), there is a series of states with their free energy minima at integer multiples of flux quantization $\phi_0$, such as [000]$_g$, [111]$_g$, [222]$_g$ and [333]$_g$, where the number $L = 0, 1, 2, \ldots$ refers to the winding numbers of the three SC order parameters around the SC ring. Namely the three SC order parameters wind simultaneously in these ground states although the three phases deviate from each other by $2\pi/3$ due to the repulsive Josephson-like coupling $\gamma < 0$ in Eq. (1). In these states the system responds to the external magnetic field in the same way as single-component superconductors [18]; especially the magneto resistance oscillates with applied magnetic flux in the period $\phi_0$ as seen in the recent Little-parks experiment at low temperatures [1].
Remarkably, we find another series of metastable states at free energy higher than the ground states which exhibit their free energy minima at integer multiples of $\phi_0/3$ starting from the zero applied magnetic flux. Distinguished from the ground states, one phase of the three SC order parameters winds $2\pi$ more than the other two in states [100], [211], [322] etc., or less than the other two in states [011], [122], [233] etc., which intervene each other and yield the free energy minima separated from each other by $\phi_0/3$. We believe these metastable states are responsible for the $\phi_0/3$ magneto resistance oscillations in the recent Little-Parks experiment at temperatures ranging from 2.9K to 2.4K [1].

The phase winding in these metastable states is much richer as compared with the ground states. In state [100] depicted in Fig. 2(a) and 2(b), since the SC order parameter $\psi_1$ acquires $2\pi$ phase around the ring, its phase changes faster than the other two SC order parameters, which renders inevitably two DW: one at the north pole of the ring where the phases of $\psi_1$ and $\psi_2$ across each other, the other one at the south pole of the ring where the phases of $\psi_1$ and $\psi_3$ across each other. As addressed in previous works on SC states with three or more order parameters and $\gamma < 0$ (see Eq. (1)), a $Z_2$ chirality can be defined based on the relative phase differences when the bilinear Josephson coupling takes $\gamma < 0$ in Eq. (1), where the time reversal symmetry is broken [14–17,33]). It is clear that in state [100] the two halves of the SC ring correspond to two domains with opposite chirality as depicted by the red and blue colors in the circle at the ring center in Fig. 2(b). The DW are against the Josephson couplings, which raises the free energy of state [100] above the ground state. The system compromises the energy cost by reducing partially the amplitude of SC order parameter according to the total free energy expression as can be seen in Fig. 2(b) for state [100]. When the winding numbers increase, the three phases interweave in magnificent patterns forming beautiful phase kaleidoscope as displayed in Fig. 2(b).
Metastable states should also take place at integer multiples of $\phi_0$ in order to complete the $\phi_0/3$ series. Comparing metastable states [000], [111], [222] and [333] with same winding numbers in the three SC order parameters in Fig. 2(b) with their counterparts in the ground state [000]$_g$, [111]$_g$, [222]$_g$ and [333]$_g$ in Fig. 2(c), it is clear that in the metastable states there are two DW between same two SC order parameters, such as $\psi_1$ and $\psi_2$, whereas in other metastable states shown in Fig. 2 the two DW involve three SC order parameters and there is no DW in ground states [34]. There are other possible metastable states including four or more DW, which take higher free energies and are thus popular at higher temperatures. Nevertheless, their free energy minima always take place at integer multiples of $\phi_0/3$, same as the metastable states with 2 DW displayed in Fig. 1(b).

Starting from one of the metastable states such as state [100] and increasing the applied magnetic flux, the system remains to the same winding configuration up to a magnetic flux $\phi/\phi_0=0.885$, which is much higher than $\phi/\phi_0=2/3$ where the free energy minimum of state [011] locates. As the price, the system acquires a large free energy which is induced by the large suppression in SC order parameters $\psi_2$ and $\psi_3$ as depicted in Fig. 2(d). Then, upon a tiny increase of magnetic flux to $\phi/\phi_0=0.89$ state [100] loses its metastability and the system jumps to state [111] with a smaller free energy accompanied by the recovery of the amplitudes of $\psi_2$ and $\psi_3$. Obviously, this is nothing but penetration of two vortices into the SC ring, each carried by $\psi_2$ and $\psi_3$. Similarly, around $\phi/\phi_0=2.215$ two vortices penetrate into the SC ring, both carried by $\psi_1$.

Why can states with winding numbers distinct in the three SC order parameters be metastable? In the present system with repulsive bilinear Josephson-type coupling between three SC order parameter ($\gamma < 0$ in Eq. (1)), there are two diverging coherence lengths [14]: $\xi_H = h/\sqrt{-2(a+\gamma)m}$ associated with the Higgs mode, where only the amplitudes of SC order parameters are involved (see Fig. 3(a)) which is essentially same as single-component
SC [26,27]; $\xi_{HL} = \hbar/\sqrt{-(a + \gamma)m}$ associated with a *Higgs-Leggett mode*, which involves both SC amplitudes and phases (see Fig. 3(b)) and is unique in systems with three and more SC order parameters [13,14]. As indicating by $\xi_{HL} > \xi_H$, the free energy cost of distortions in SC order parameters induced by the Higgs-Leggett mode is smaller than that induced by the Higgs mode (the zero-energy Nambu-Goldstone mode corresponds to an infinite coherence length). With the Higgs-Leggett mechanism, the system responds to the applied magnetic flux by enhancing amplitude in one SC order parameter and squeezing phase difference between the two remaining SC order parameters as schematically shown in Fig. 3(b) [14] (see also Fig. 2(d)); when the phase difference shrinks to zero and then changes sign, a DW between $Z_2$ chirality domains appears. This renders states with winding numbers distinct in the three SC order parameters, and thus local free energy minima at integer multiples of $\phi_0/3$, as revealed above (see Fig. 2). In a stark contrast, in the Higgs mechanism, all three SC order parameters carry the same winding number and are suppressed simultaneously (see Fig. 3(a)). In order to accommodate more vortices into the SC ring, all the three SC order parameters have to be suppressed to zero, which costs a large free energy and thus is unfavorable. The Higgs-Leggett mode and thus the coherence length $\xi_{HL}$ are unique for systems with three (or more) SC order parameters, implying that half flux quantization $\phi_0/2$ if any has to be realized in a different mechanism.

**Real-space distribution of PDW.** The behaviors of SC order parameters upon sweeping of the applied magnetic flux unveiled by the above free energy analysis leave direct consequences in the real-space PDW lattice. For the case of three SC order parameters [31,32] one has

$$\Psi_3(r) = \psi_1 e^{iQ_1 \cdot r} + \psi_2 e^{iQ_2 \cdot r} + \psi_3 e^{iQ_3 \cdot r}$$

where $Q_1 = 2 \frac{2\pi}{d\sqrt{3}} \left[ \frac{\sqrt{3}}{2} - \frac{1}{2} \right]$, $Q_2 = 2 \frac{2\pi}{d\sqrt{3}} \left[ 0 \quad 1 \right]$ and $Q_3 = 2 \frac{2\pi}{d\sqrt{3}} \left[ -\frac{\sqrt{3}}{2} \quad -\frac{1}{2} \right]$ with $d$ the lattice constant of the Kagome lattice. A common constant for the three SC order parameters generates a triangular lattice for the maxima of the absolute value of the SC condensate $\Psi_3(r)$,
whereas around the zeros of $\Psi_3(r)$ located at the center of triangles vortex and antivortex of supercurrent with vorticity $\pm 1$ appear alternatingly as depicted in the inset of the left panel of Fig. 4(a) [31,32]; Complex numbers (with the same absolute value) distinct in the three SC order parameters merely slide the PDW lattice in the three corresponding directions given by $Q_1, Q_2$ and $Q_3$; A $2\pi$ phase winding in one order parameter more or less than the other two order parameters, such as $\psi_1 = e^{\pm i \theta}$, introduces a dislocation in the PDW lattice. However, since the dislocation takes place at the origin of coordinate, it is not easy to capture by local techniques in the Little-Parks setup. In contrast, DW in metastable states associated with the $\phi_0/3$ flux quantization induce distortions in the PDW lattice which may be detected experimentally. The perfect PDW lattice associated with the SC condensate $\Psi_3(r)$ in the ground states [000]$g$ and [111]$g$ is shown in Fig. 4(a) for a slice of SC ring. In contrast, the metastable states [100] and [111] with two DW of $Z_2$ chirality domains on the ring are accompanied by distortions in the real-space PDW lattice, where the amplitudes of PDW are suppressed slightly as displayed in Fig. 4(b).

One may also consider the following PDW lattice [32]

$$\Psi_6(r) = 2\psi_1 \cos(Q_1 \cdot r) + 2\psi_2 \cos(Q_2 \cdot r) + 2\psi_3 \cos(Q_3 \cdot r),\quad (3)$$

for which the maxima of the amplitude of SC condensate $\Psi_6(r)$ form a Kagome lattice accompanied by -2 vorticity in hexagons and +1 vorticity in triangles in the ground states (see inset of the left panel of Fig. 4(c)). The metastable states [100] and [111] with two DW of $Z_2$ chirality domains on the ring induce distortions in the real-space PDW lattice as displayed in Fig. 4(d), where the maxima in the amplitude of the SC condensate $\Psi_6(r)$ form stripes of triangles.

**Discussions**

**Mean-field and genuine SC phase transitions.** In the present work, the fractional flux quantization $\phi_0/3$ has been clarified below the transition point $T_{mf}$ determined by the GL free
energy functional Eq. (1): $\alpha + \gamma = 0$. While these states with phase windings distinct in the three SC order parameters accompanied by DW of opposite chirality are metastable in the GL theory, they are stabilized thermodynamically when thermal fluctuations are addressed appropriately. The transition point $T_{mf}$ is around 3K for the CsV$_3$Sb$_5$ sample in the Little-Parks experiment (see Fig. 1c and Fig. 2i in Ref. [1]). It is anticipated that at a lower, genuine transition point $T_c$ these DW should be expelled completely from the system, where magnetic flux quantization takes place only at integer multiples of $\phi_0$ as revealed in the present work, which renders the vanishing bulk zero-field resistance simultaneously. From Fig. 1c and Fig. 2i in Ref. [1], it is estimated $T_c \approx 1$K. Therefore, the fractional magnetic flux quantization is characteristic of an intermediate SC phase in the Kagome vanadium-based superconductors, which is reminiscent of the picture proposed in the previous works [28–32] where melting of the PDW lattice results in a phase of uniform order of $6e$ SC condensate, which is secondary before melting.

About half flux quantization $\phi_0/2$. We have to note that the present theory cannot capture easily the half flux quantization $\phi_0/2$. Actually, we have performed the similar analysis for a system of two SC order parameters by omitting one SC order parameter in Eq. (1). We can only find states with the same winding number in the two SC order parameters and the free energy minima appear at the integer multiples of $\phi_0$, whereas configurations with winding numbers distinct in the two SC order parameters are unstable. We take this as evidence for the importance of the Higgs-Leggett mechanism in the fractional magnetic flux quantization, since the Higgs-Leggett mode is absent in systems with two SC order parameters. As a matter of fact, we notice that in the recent experiment [1] the signals of $\phi_0/2$ are scattered as compared with those of $\phi_0/3$.

Other possible novel phenomena. For the chiral $6e$ SC state implied by the Little-Parks experiment [1] novel SC phenomena are expected in addition to the one discussed in the
present work. For a SC ring with sufficient width where supercurrent is suppressed to zero due to Meissner effect at the center part of superconductor, the magnetic flux trapped by the SC ring should be quantized into integer multiples of $\phi_0/3$ resulting in plateaus in magnetization curve [34]. In a narrow constriction between two bulk CsV$_3$Sb$_5$ crystals, the critical Josephson current is suppressed significantly (to zero in theory) when the two bulks take opposite chirality, as compared to the case where a same chirality occupies the two bulks [14]. An unconventional intermediate SC state characterized by clustering vortices may also be possible [35].

**Methods**

**Numerical simulations.** In order to capture the period-$\phi_0/3$ magneto resistance observed in the Kagome vanadium-based superconductor, we consider the GL free energy functional with three SC order parameters $\psi_j (j = 1,2,3)$ at the three reciprocal lattice vectors connecting M points of the hexagonal Brillouin zone of the material as given in Eq. (1). Performing the variational analysis with respect to $\psi_j^*$, we obtain the GL equations:

$$a \psi_j + b |\psi_j|^2 \psi_j + \left( \frac{\nabla}{i} - A \right)^2 \psi_j - \sum_{k=1,2,3; j \neq k} \gamma \psi_k = 0 \quad (4)$$

with $j = 1,2,3$. These equations are given in dimensionless form for convenience of numerical calculation: length in units of $\xi_1 = \hbar/\sqrt{-20m a}$, order parameter $\psi_j$ in units of $\psi_0 = \sqrt{-10a/b}$, $A$ in units of $\hbar c/2e\xi_1$, and free energy in units of $G_0=(10a)^2/b$. For numerical calculations in this work, we take $a = -0.1$, $b = 1$ and $\gamma = -0.24$.

The simulation is implemented using finite difference method [36,37] for ring geometry of the sample with polar coordinate. For rings with small width as compared to the penetration depth, the demagnetization effect can be neglected. The gauge $\nabla \cdot A = 0$ is taken with $A = e \phi H r/2$ for the uniform magnetic field $H$. The boundary conditions for $\psi_j$ correspond to zero total current density normal to the sample surface.
Starting from suitable initial configurations of SC order parameters, we obtain ground state solutions and metastable solutions by the iterative relaxation method. To find the metastable states with two DW carrying integer multiples of $\phi_0/3$, we should put initial guesses within the attractive basins of the final solutions. As an example, in order to obtain state [100] which carries magnetic flux of $\phi_0/3$ we set $\psi_1(\varphi) = \sqrt{n_0}e^{i\varphi}$, $\psi_2(\varphi) = \sqrt{n_0}e^{i2\pi/3}$ and $\psi_3(\varphi) = \sqrt{n_0}e^{i4\pi/3}$ where $n_0$ is the ground state amplitude in absence of the applied magnetic flux. Two DW separating two different chiral domains are formed after a few iteration steps from the initial guess during simulations. The algorithm is iterated until changes in SC order parameters between two steps are less than $10^{-6}$.

Next, we sweep slightly up/down the applied magnetic flux and recalculate the distribution of amplitudes and phases of SC order parameters with those of the previous solution as the initial guess, and reach the new solution. Repeating this process gives one of the parabolic curves in the diagram of free energy functional. Although they are not the ground states, we find that the solutions with two DW and winding numbers distinct in the three SC order parameters are stable against small perturbations. As the applied magnetic flux deviates largely from the value where a local minimum of free energy locates, the solution jumps discontinuously to a new one with a different set of winding numbers. This results in the series of metastable states with local free energy minima at integer multiples of $\phi_0/3$.

References

[1] J. Ge, P. Wang, Y. Xing, Q. Yin, H. Lei, Z. Wang, and J. Wang, Discovery of Charge-4e and Charge-6e Superconductivity in Kagome Superconductor CsV$_3$Sb$_5$, arXiv:2202.20352 (2022).

[2] B. R. Ortiz, L. C. Gomes, J. R. Morey, M. Winiarski, M. Bordelon, J. S. Mangum, I. W. H. Oswald, J. A. Rodriguez-Rivera, J. R. Neilson, S. D. Wilson, E. Ertekin, T. M.
McQueen, and E. S. Toberer, New Kagome Prototype Materials: Discovery of KV_3Sb_5, RbV_3Sb_5, *Phys. Rev. Mater.* **3**, 094407 (2019).

[3] B. R. Ortiz, S. M. L. Teicher, Y. Hu, J. L. Zuo, P. M. Sarte, E. C. Schueller, A. M. M. Abeykoon, M. J. Krogstad, S. Rosenkranz, R. Osborn, R. Seshadri, L. Balents, J. He, and S. D. Wilson, CsV_3Sb_5: A Z_2 Topological Kagome Metal with a Superconducting Ground State, *Phys. Rev. Lett.* **125**, 247002 (2020).

[4] H. Zhao, H. Li, B. R. Ortiz, S. M. L. Teicher, T. Park, M. Ye, Z. Wang, L. Balents, S. D. Wilson, and I. Zeljkovic, Cascade of Correlated Electron States in the Kagome Superconductor CsV_3Sb_5, *Nature* **599**, 216 (2021).

[5] H. Chen, H. Yang, B. Hu, Z. Zhao, J. Yuan, Y. Xing, G. Qian, Z. Huang, G. Li, Y. Ye, S. Ma, S. Ni, H. Zhang, Q. Yin, C. Gong, Z. Tu, H. Lei, H. Tan, S. Zhou, C. Shen, X. Dong, B. Yan, Z. Wang, and H.-J. Gao, Roton Pair Density Wave in a Strong-Coupling Kagome Superconductor, *Nature* **599**, 222 (2021).

[6] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Theory of Superconductivity, *Phys. Rev.* **106**, 162 (1957); **108**, 1175 (1957).

[7] A. A. Abrikosov, Magnetic Properties of Superconductors of the Second Group, *Sov. Phys. JETP* **5**, 1174 (1957).

[8] Y. Nambu, Quasi-Particles and Gauge Invariance in the Theory of Superconductivity, *Phys. Rev.* **117**, 648 (1960).

[9] F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1950).

[10] N. Byers and C. N. Yang, Theoretical Considerations Concerning Quantized Magnetic Flux in Superconducting Cylinders, *Phys. Rev. Lett.* **7**, 46 (1961).

[11] L. Onsager, Magnetic Flux Through a Superconducting Ring, *Phys. Rev. Lett.* **7**, 50 (1961).

[12] M. Tinkham, *Introduction to Superconductivity*, 2nd ed (McGraw Hill, New York, 1996).
[13] A. J. Leggett, Number-Phase Fluctuations in Two-Band Superconductors, *Prog. Theor. Phys.* **36**, 901 (1966).

[14] X. Hu and Z. Wang, Stability and Josephson Effect of Time-Reversal-Symmetry-Broken Multicomponent Superconductivity Induced by Frustrated Intercomponent Coupling, *Phys. Rev. B* **85**, 064516 (2012).

[15] V. Stanev and Z. Tešanović, Three-Band Superconductivity and the Order Parameter That Breaks Time-Reversal Symmetry, *Phys. Rev. B* **81**, 134522 (2010).

[16] Y. Tanaka and T. Yanagisawa, Chiral Ground State in Three-Band Superconductors, *J. Phys. Soc. Jpn.* **79**, 114706 (2010).

[17] J. Carlström, J. Garaud, and E. Babaev, Length Scales, Collective Modes, and Type-1.5 Regimes in Three-Band Superconductors, *Phys. Rev. B* **84**, 134518 (2011).

[18] W. A. Little and R. D. Parks, Observation of Quantum Periodicity in the Transition Temperature of a Superconducting Cylinder, *Phys. Rev. Lett.* **9**, 9 (1962).

[19] F. H. Yu, T. Wu, Z. Y. Wang, B. Lei, W. Z. Zhuo, J. J. Ying, and X. H. Chen, Concurrence of Anomalous Hall Effect and Charge Density Wave in a Superconducting Topological Kagome Metal, *Phys. Rev. B* **104**, L041103 (2021).

[20] Y.-X. Jiang, J.-X. Yin, M. M. Denner, N. Shumiya, B. R. Ortiz, G. Xu, Z. Guguchia, J. He, M. S. Hossain, X. Liu, J. Ruff, L. Kautzsch, S. S. Zhang, G. Chang, I. Belopolski, Q. Zhang, T. A. Cochran, D. Multer, M. Litskevich, Z.-J. Cheng, X. P. Yang, Z. Wang, R. Thomale, T. Neupert, S. D. Wilson, and M. Z. Hasan, Unconventional Chiral Charge Order in Kagome Superconductor KV₃Sb₅, *Nat. Mater.* **20**, 1353 (2021).

[21] Z. Liang, X. Hou, F. Zhang, W. Ma, P. Wu, Z. Zhang, F. Yu, J.-J. Ying, K. Jiang, L. Shan, Z. Wang, and X.-H. Chen, Three-Dimensional Charge Density Wave and Surface-Dependent Vortex-Core States in a Kagome Superconductor CsV₃Sb₅, *Phys. Rev. X* **11**, 031026 (2021).
[22] S.-Y. Yang, Y. Wang, B. R. Ortiz, D. Liu, J. Gayles, E. Derunova, R. Gonzalez-Hernandez, L. Šmejkal, Y. Chen, S. S. P. Parkin, S. D. Wilson, E. S. Toberer, T. McQueen, and M. N. Ali, Giant, Unconventional Anomalous Hall Effect in the Metallic Frustrated Magnet Candidate, KV₃Sb₅, Sci. Adv. 6, eabb6003 (2020).

[23] C. Mielke, D. Das, J.-X. Yin, H. Liu, R. Gupta, Y.-X. Jiang, M. Medarde, X. Wu, H. C. Lei, J. Chang, P. Dai, Q. Si, H. Miao, R. Thomale, T. Neupert, Y. Shi, R. Khasanov, M. Z. Hasan, H. Luetkens, and Z. Guguchia, Time-Reversal Symmetry-Breaking Charge Order in a Kagome Superconductor, Nature 602, 245 (2022).

[24] H.-S. Xu, Y.-J. Yan, R. Yin, W. Xia, S. Fang, Z. Chen, Y. Li, W. Yang, Y. Guo, and D.-L. Feng, Multiband Superconductivity with Sign-Preserving Order Parameter in Kagome Superconductor CsV₃Sb₅, Phys. Rev. Lett. 127, 187004 (2021).

[25] C. M. Varma, Flux Quantization Cubed, DOI:10.36471/JCCM_March_2022_03 (2022).

[26] D. Pekker and C. M. Varma, Amplitude/Higgs Modes in Condensed Matter Physics, Annu. Rev. Condens. Matter Phys. 6, 269 (2015).

[27] P. W. Anderson, Coherent Excited States in the Theory of Superconductivity: Gauge Invariance and the Meissner Effect, Phys. Rev. 110, 827 (1958).

[28] D. F. Agterberg and H. Tsunetsugu, Dislocations and Vortices in Pair-Density-Wave Superconductors, Nat. Phys. 4, 639 (2008).

[29] E. Berg, E. Fradkin, and S. A. Kivelson, Charge-4e Superconductivity from Pair-Density-Wave Order in Certain High-Temperature Superconductors, Nat. Phys. 5, 830 (2009).

[30] L. Radzihovsky and A. Vishwanath, Quantum Liquid Crystals in an Imbalanced Fermi Gas: Fluctuations and Fractional Vortices in Larkin-Ovchinnikov States, Phys. Rev. Lett. 103, 010404 (2009).

[31] D. F. Agterberg, M. Geracie, and H. Tsunetsugu, Conventional and Charge-Six Superfluids from Melting Hexagonal Fulde-Ferrell-Larkin-Ovchinnikov Phases in Two Dimensions, Phys. Rev. B 84, 014513 (2011).
[32] S. Zhou and Z. Wang, Doped Orbital Chern Insulator, Chern Fermi Pockets, and Chiral Topological Pair Density Wave in Kagome Superconductors, arXiv:2110.06266 (2021).

[33] Y. Tanaka, Multicomponent Superconductivity Based on Multiband Superconductors, *Supercond. Sci. Technol.* **28**, 034002 (2015).

[34] Z. Huang and X. Hu, Fractional Flux Plateau in Magnetization Curve of Multicomponent Superconductor Loop, *Phys. Rev. B* **92**, 214516 (2015).

[35] Y. Takahashi, Z. Huang, and X. Hu, H–T Phase Diagram of Multi-Component Superconductors with Frustrated Inter-Component Couplings, *J. Phys. Soc. Jpn.* 83, 034701 (2014).

[36] M. V. Milošević and R. Geurts, The Ginzburg–Landau Theory in Application, *Physica C Supercond.* **470**, 791 (2010).

[37] B. J. Baelus, F. M. Peeters, and V. A. Schweigert, Vortex States in Superconducting Rings, *Phys. Rev. B* **61**, 9734 (2000).

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Fig. 1. Free energy minima at integer multiples of one-third flux quantization $\phi_0/3$ in the Little-Parks setup for the Kagome-structured superconductor CsV$3$Sb$5$. (a) Schematic setup for the Little-Parks experiment where $Z_2$ chirality domains defined by phases of three SC order parameters are separated by DW, which are associated with distortions in the real-space PDW lattice shown schematically by color. (b) Free energy as a function of applied magnetic flux penetrating through the superconductivity ring. In the ground states, such as $[000]_g$, $[111]_g$, $[222]_g$ and $[333]_g$, the three SC order parameters wind in the same way around the ring, which yields free energy minima at the integer multiples of $\phi_0$. In the metastable states one phase of the three SC component winds one more time than the other two in states such as $[100]$, $[211]$, $[322]$ etc., or one less time than the other two in states such as $[011]$, $[122]$, $[233]$ etc., which intervenes each other and yields the free energy minima separated from each other by $\phi_0/3$. The metastable states are triply degenerate. Parameters are taken as $a = -0.1$, $b = 1$, and $\gamma = -0.24$. The radius of the ring is taken as $R = 8\xi_1$. 


Fig. 2. Kaleidoscope of phases winding for the three SC order parameters in the Little-Parks setup at integer multiples of $\phi_0/3$. (a) Schematic diagram of free energy for the metastable states with circles and squares denoting the values of applied magnetic flux for which the detailed wavefunctions are displayed in (b) and (d), respectively. (b) Distribution of phases of the three SC order parameters around the SC ring in the metastable states at the applied magnetic flux marked by circles in (a) where, for example, [211] refers to the state with phase winding 2, 1 and 1 in the SC order parameter $\psi_1$, $\psi_2$ and $\psi_3$, respectively. (c) Same for (b) except for the ground states at applied magnetic flux of integer multiples of $\phi_0$. (d) Distributions of the amplitudes of the three SC order parameters around the SC ring in the metastable states at the applied magnetic flux marked by squares in (a), where the winding numbers jumps corresponding to penetration of vortices into the SC ring in one or two out of three SC order parameters. Parameters are taken same as Fig. 1.
Fig. 3. Possible modes in systems with three SC order parameters and repulsive bilinear Josephson-like inter-component couplings. (a) Higgs mode where only variations in amplitudes of the three SC order parameters are involved, which is essentially same for SC state with a single SC order parameter. (b) Higgs-Leggett mode where variation in amplitude of one SC order parameter is accompanied by variation of phase difference between the two remaining SC order parameters. The three colored arrows indicate the three complex SC order parameters associated with a $Z_2$ chiral state induced by repulsive bilinear Josephson-like coupling.
Fig. 4. Real-space distribution of SC condensate of the pair density wave (PDW) \( \Psi_3(\mathbf{r}) \) and \( \Psi_6(\mathbf{r}) \) (see text for detailed definition). (a) Amplitude of PDW \( |\Psi_3(\mathbf{r})| \) on a slice of ring in the ground states [000]_g and [111]_g. The maxima of amplitude \( |\Psi_3(\mathbf{r})| \) form a triangular lattice, and the zeros of \( \Psi_3(\mathbf{r}) \) locate at the center of triangles around which +1 vorticity and -1 vorticity of supercurrent appear alternatingly as depicted in the inset of the left panel. (b) Same as (a) except for the metastable states [100] and [111] with two DW of \( Z_2 \).
chirality domains on the ring, which induce shear distortions in the PDW lattice, where the amplitudes of PDW are suppressed slightly. (c) Same as (a) except that the local maxima of $|\Psi_6(r)|$ form a Kagome lattice, and the zeros of $\Psi_6(r)$ locate at the centers of triangles and hexagons around which $+1$ vorticity and $-2$ vorticity of supercurrent appear as depicted in the inset of the left panel. (d) Same as (b) except for the metastable states [100] and [111] with two DW of $Z_2$ chirality domains on the SC ring, where the maxima of $|\Psi_6(r)|$ form stripes of triangles.