Fractional Quantization of the Magnetic Flux in Cylindrical Unconventional Superconductors

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The magnetic flux threading a conventional superconducting ring is typically quantized in units of $\Phi_0 = hc/2e$. The factor of 2 in the denominator of Φ_0 originates from the existence of two different types of pairing states with minima of the free energy at even and odd multiples of Φ_0. Here we show that spatially modulated pairing states exist with energy minima at fractional flux values, in particular, at multiples of $\Phi_0/2$. In such states, condensates with different center-of-mass momenta of the Cooper pairs coexist. The proposed mechanism for fractional flux quantization is discussed in the context of cuprate superconductors, where $hc/4e$ flux periodicities were observed.

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In superconducting (SC) rings, flux quantization in units of $\Phi_0 = hc/2e$ is usually attributed to the charge $2e$ of Cooper pairs carrying the supercurrent. This connection has been anticipated by Fritz London [1] long before the experimental confirmation of the SC flux quantum Φ_0 by Doll and Näbauer [2] and by Deaver and Fairbank [3] in 1961. In the same year, Φ_0 was derived from the BCS pairing theory as a consequence of the existence of two classes of SC wave functions with energy minima at even or odd multiples of Φ_0 [4–6].

The formation of a pair condensate alone does not inevitably imply that Φ_0 is the unit of flux quantization. In fact, this conclusion is valid only for a uniform condensate of noninteracting Cooper pairs, as assumed in BCS theory. Normal persistent currents can as well sustain a $hc/2e$ flux periodicity in special geometries [7] or due to electron-electron interactions [8,9]. Correlations between Cooper pairs may also lead to fractional flux quanta. Such effects were first discussed by Little in 1964 in the context of fractional flux periodicities in the critical temperature of conventional SC cylinders [10]. More recently, unusual flux periodicities were reported in superconducting quantum interference device (SQUID) experiments with high-T_c cuprates [11–13]. An example for $\Phi_0/2$ oscillations of the critical current is shown Fig. 1. The low magnetic field data indicate a $\sin 2\varphi$ Josephson relation. As one likely origin of the fractional periodicity, multiple Andreev scattering in grain-boundary Josephson junctions was proposed [11,12]; however, the abrupt disappearance of the $\Phi_0/2$ periodicity beyond a threshold field has remained unexplained. In the search for an alternative origin, interactions between Cooper pairs and quartet formation have been investigated theoretically [14–17]. Yet, their influence on flux quantization is unresolved.

Here we formulate a conceptually distinct mechanism for fractional flux quanta in superconductors with a spatially modulated SC order parameter (OP), in particular, for $\Phi_0/2$ flux periodicity and a $\sin 2\varphi$ Josephson relation in SQUIDs. The proposed concept rests on the coexistence of pair condensates with different center-of-mass momenta (COMM) of Cooper pairs and is unique for superconductors with unconventional pairing symmetries.

Consider the uniform OP $\Delta(\theta) = \Delta e^{i\theta(q-\Phi/\Phi_0)}$ for electron pairs with quantized COMM $\hbar q$ and angular coordinate θ on a hollow cylinder threaded by the magnetic flux Φ. In this geometry, q refers to the phase-winding number of the OP upon circulating the cylinder once. The free energy F of this system has a minimum for a vanishing phase gradient, i.e., at the flux value $\Phi = q\Phi_0$. Since q is an integer, the fluxoid threading the cylinder is quantized in units of Φ_0 [18,19].

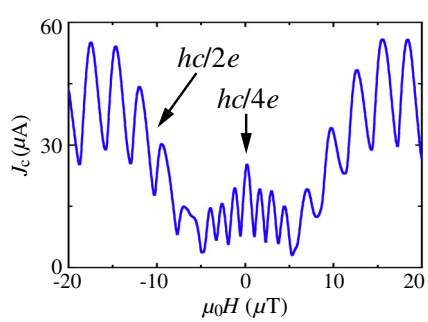

FIG. 1 (color online). Critical current $J_c(H)$ through a 24° YBCO grain-boundary SQUID at $T = 4.2$ K (data from Ref. [12]). Clearly visible is the abrupt change of periodicity at $\mu_0 H \approx \pm 5$ μT. For the SQUID size in this experiment, a flux of Φ_0 is achieved for a magnetic field of 2.7 μT. Data reprinted with permission from C. W. Schneider.

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If instead the magnitude Δ of the OP oscillates around the cylinder, Cooper pairs with different COMMs exist. An example for a sign-changing OP is

$$\tilde{\Delta}(\theta) = (\Delta_1 e^{iq_1\theta} + \Delta_2 e^{iq_2\theta}) e^{-i\theta\Phi/\Phi_0} \xrightarrow{\Delta_1=\Delta_2} 2\Delta_1 \times \cos(\theta[q_1 - q_2]/2) e^{i\theta([q_1+q_2]/2-\Phi/\Phi_0)}. \quad (1)$$

In such a state, pairs with COMM q_1 coexist with pairs with q_2; its phase-winding number is $[q_1 + q_2]/2$, which implies that F has a minimum at $\Phi/\Phi_0 = [q_1 + q_2]/2$. Since $q_1 + q_2$ can be both even or odd, the flux is hence quantized in units of $\Phi_0/2$.

The above state generalizes the time-reversal symmetric "pair-density wave" (PDW) concept with $q_1 = -q_2$ (see, e.g., Refs. [20,21], and references therein). This state bares similarities to Fulde-Ferrell-Larkin-Ovchinnikov states in an external magnetic field [22,23]. Because of its concomitant charge-density modulations, the PDW was proposed as a candidate ground state of striped cuprate superconductors [21,24]. For unconventional pairing symmetries with gap nodes [25], it was indeed verified that the PDW state has a finite range of stability. Topological considerations furthermore demonstrated that the PDW state can sustain a $\Phi_0/2$ vortex phase [20,26,27]. In fact, analogous arguments were introduced earlier in the context of "stripe fractionalization," where a sign-changing antiferromagnetic OP was considered [28,29]. The melting of the PDW state may even give rise to a state with charge-4e superconductivity, which also leads to a $\Phi_0/2$ periodicity in a SQUID geometry [27].

Another system where the generalized PDW concept may lead to $\Phi_0/2$ flux quanta is the spin-triplet superconductor Sr_2RuO_4. Here it is the two-component order parameter $\{\Delta^{\uparrow\uparrow}_{q_1}, \Delta^{\downarrow\downarrow}_{q_2}\}$ for the $s_z = 1$ and the $s_z = -1$ condensates which allows for the coexistence of different COMMs [30]. Indeed, experimental evidence was reported that the flux through a Sr_2RuO_4 ring is quantized in units of $\Phi_0/2$ [31]. The rare observations of fractional flux quanta have been encountered only for unconventional superconductors. This is not an accident, because it is the existence of gap nodes, in particular, for the d-wave pairing symmetry of the cuprates, which allows for the emergence of a PDW ground state [25].

In the following, we apply the concept of the generalized PDW with arbitrary combinations of COMMs q_1 and q_2 to flux-threaded hollow cylinders. We show that a nonlocal pairing interaction allows for a PDW ground state. Furthermore, for sufficiently large cylinders, the ground state energy $E(n\Phi_0/2)$ (with integer n) is minimized by a combination of two COMMs q_1 and q_2 fulfilling $(q_1 + q_2)/2 = n/2$, and, consequently, the flux is quantized in units of $\Phi_0/2$. The energy minima for different n become degenerate in the thermodynamic limit, leading to a $\Phi_0/2$ periodicity of the ground state energy and the supercurrent.

We start from a pairing Hamiltonian on a square lattice (the lattice constant is set to 1) with periodic boundary conditions, and N_θ sites in the θ direction, N_z sites in the z direction [32]:

$$\mathcal{H} = \sum_{\mathbf{k},s} \epsilon_{\mathbf{k}}(\varphi) c^\dagger_{\mathbf{k}s} c_{\mathbf{k}s} + \frac{1}{N_\theta N_z} \sum_{\mathbf{q}} \sum_{\mathbf{k},\mathbf{k}'} \sum_{s,s'} V(\mathbf{k},\mathbf{k}',\mathbf{q}) c^\dagger_{\mathbf{k}s} c^\dagger_{-\mathbf{k}+\mathbf{q}s'} c_{-\mathbf{k}'+\mathbf{q}s'} c_{\mathbf{k}'s} \quad (2)$$

with a general attractive interaction $V(\mathbf{k},\mathbf{k}',\mathbf{q})$. Here, $\mathbf{k} = (k_\theta, k_z)$, where $k_\theta = 1, \ldots, N_\theta$ enumerates the angular momenta $\hbar k_\theta$ for motion around the cylinder and $k_z = 1, \ldots, N_z$ the momenta $\hbar k_z$ in the z direction. The dispersion in the z direction is required for an unconventional OP, but it is otherwise not essential for the flux periodicity. The single-electron dispersion takes the form

$$\epsilon_{\mathbf{k}}(\varphi) = -2t\left[\cos\left(\frac{2\pi(k_\theta - \varphi)}{N_\theta}\right) + \cos\left(\frac{2\pi k_z}{N_z}\right)\right] \quad (3)$$

with nearest-neighbor hopping amplitude t. The magnetic flux enters in $\epsilon_{\mathbf{k}}(\varphi)$ through $\varphi = \Phi e/hc = \Phi/2\Phi_0$.

For a superconducting state with singlet pairing, we use the BCS mean-field decoupling scheme and approximate $\langle c^\dagger_{\mathbf{k}\uparrow} c^\dagger_{-\mathbf{k}+\mathbf{q}\downarrow} c_{-\mathbf{k}'+\mathbf{q}\downarrow} c_{\mathbf{k}'\uparrow}\rangle \to \langle c^\dagger_{\mathbf{k}\uparrow} c^\dagger_{-\mathbf{k}+\mathbf{q}\downarrow}\rangle\langle c_{-\mathbf{k}'+\mathbf{q}\downarrow} c_{\mathbf{k}'\uparrow}\rangle$. The Heisenberg equations of motion for the spin-independent imaginary time Green's function $\mathcal{G}(\mathbf{k},\mathbf{k}',\tau) = -\langle T_\tau c_{\mathbf{k}s}(\tau) c^\dagger_{\mathbf{k}'s}(0)\rangle$ and the anomalous propagators $\mathcal{F}(\mathbf{k},\mathbf{k}',\tau) = \langle T_\tau c_{\mathbf{k}s}(\tau) c_{-\mathbf{k}'s'}(0)\rangle$ and $\mathcal{F}^*(\mathbf{k},\mathbf{k}',\tau) = \langle T_\tau c^\dagger_{-\mathbf{k}s}(\tau) c^\dagger_{\mathbf{k}'s'}(0)\rangle$ for $s \neq s'$ then lead to the Gor'kov equations [33]:

$$\mathcal{G}(\mathbf{k},\mathbf{k}',\omega_n) = \mathcal{G}_0(\mathbf{k},\omega n)\left[\delta_{\mathbf{k}\mathbf{k}'} - \sum_{\mathbf{q}} \Delta(\mathbf{k},\mathbf{q}) \mathcal{F}^*(\mathbf{k}-\mathbf{q},\mathbf{k}',\omega_n)\right] \quad (4)$$

and

$$\mathcal{F}(\mathbf{k},\mathbf{k}',\omega_n) = \mathcal{G}_0(\mathbf{k},\omega_n) \sum_{\mathbf{q}} \Delta(\mathbf{k},\mathbf{q}) \mathcal{G}(-\mathbf{k}',-\mathbf{k}+\mathbf{q},-\omega_n), \quad (5)$$

where $\mathcal{G}_0(\mathbf{k},\omega_n) = [i\omega_n - \epsilon_{\mathbf{k}} + \mu]^{-1}$ is the Green's function in the normal state and $\omega_n = (2n-1)\pi k_B T$ is the fermionic Matsubara frequency. The average charge density ρ is controlled by the chemical potential μ. The order parameter $\Delta(\mathbf{k},\mathbf{q})$ represents electron pairs with center-of-mass momentum $\hbar\mathbf{q}$ and is determined self-consistently as shown below.

At this point, the form of the interaction is crucial. As the simplest ansatz that allows for unconventional pairing, we choose an attraction between electrons on neighboring

sites. With the restriction to singlet pairing, only the extended s- and the d-wave channels remain, which leads to the interaction $V(\mathbf{k}, \mathbf{k}', \mathbf{q}) = V_+(\mathbf{k}, \mathbf{k}', \mathbf{q}) + V_-(\mathbf{k}, \mathbf{k}', \mathbf{q})$ with

$$V_\pm(\mathbf{k}, \mathbf{k}', \mathbf{q}) = V g_\pm(\mathbf{k} - \mathbf{q}/2) g_\pm(\mathbf{k}' - \mathbf{q}/2), \quad (6)$$

where $g_\pm(\mathbf{k}) = \cos(2\pi k_\theta/N_\theta) \pm \cos(2\pi k_z/N_z)$. The OP thus becomes

$$\Delta(\mathbf{k}, \mathbf{q}) = \Delta_s(\mathbf{q}) g_+(\mathbf{k} - \mathbf{q}/2) + \Delta_d(\mathbf{q}) g_-(\mathbf{k} - \mathbf{q}/2) \quad (7)$$

with a nonvanishing extended s-wave contribution $\Delta_s(\mathbf{q})$ if $\mathbf{q} \neq 0$ [25]. Inserting $V(\mathbf{k}, \mathbf{k}', \mathbf{q})$ from (6) into (4) generates a set of coupled self-consistency equations for $\Delta_s(\mathbf{q})$ and $\Delta_d(\mathbf{q})$:

$$\Delta_{s,d}(\mathbf{q}) = -\frac{k_B T V}{N_\theta N_z} \sum_{\mathbf{k}} g_\pm(\mathbf{k} - \mathbf{q}/2) \sum_n \mathcal{F}(\mathbf{k} - \mathbf{q}, \mathbf{k}, \omega_n). \quad (8)$$

In an ansatz for a self-consistent solution, we choose Q trial vectors $\mathbf{q}_1, \ldots, \mathbf{q}_Q$ and set $\Delta(\mathbf{k}, \mathbf{q}) = 0$ for all $\mathbf{q} \neq \mathbf{q}_i$. Thereby we test selected combinations of $\mathbf{q}$ vectors. This procedure and its solutions without magnetic flux are discussed in Ref. [25]. Cooper pairs moving in the flux-threaded cylinder acquire angular momentum, and we therefore choose $\mathbf{q} = (\pm q, 0)$ along the θ direction. Solutions of this kind are generally in competition with states with different combinations of COMMs, e.g., in the z direction. Since such COMMs do not affect flux quantization, we do not include them in our calculations. In the context of flux quantization, two notions are important: (i) If instead of Eq. (6) conventional s-wave pairing originating from an on-site interaction is considered, the ground state is the standard BCS superconductor with one COMM and therefore no fractional flux quanta occur; (ii) for the pairing interaction (6), the ground state for zero flux is either a d-wave BCS or a PDW superconductor with the two COMMs $\mathbf{q}$ and $-\mathbf{q}$, i.e., $Q = 2$, depending on the interaction strength V. If V exceeds a critical value V_c, a first-order transition from the BCS to a PDW state occurs. This phase transition originates from the competition between the cost in kinetic energy E_{kin} and a gain in condensation energy E_{con} arising from finite COMMs in the presence of gap nodes. The d-wave superconductor has four nodal points in $\mathbf{k}$ space where unpaired electrons remain at the Fermi energy. In the PDW state, these electrons can be paired by $\Delta(\mathbf{k}, \pm\mathbf{q})$. The condensation energy is therefore optimized by the vector $\mathbf{q}$ for which the nodes in $\Delta(\mathbf{k}, \mathbf{q})$ and $\Delta(\mathbf{k}, -\mathbf{q})$ are furthest apart [25].

In the following, we investigate the flux dependence of the generalized PDW and its total energy $E_{q_1,q_2}(\varphi)$ for the state with the two COMMs $\{q_1, q_2\}$ at $T = 0$. In particular, we search for the combinations $\{q_1, q_2\}$ for which $E_{q_1,q_2}(\varphi)$ is minimal for flux values $\varphi = n/2$, and we test whether combinations $\{q_1, q_2\}$ exist for which $E_{q_1,q_2}(\varphi)$ is minimal at $\varphi = n/4$. The charge density is fixed to $\rho = 0.8$, and an additional next-nearest-neighbor hopping $t' = -0.3t$ is included. For these values and $\varphi = 0$, a PDW state is realized for $V > V_c \approx 2.1t$ with $q_1 = -q_2 = q$ as the integer closest to N_θ/A, where the value of A is obtained by minimizing the total energy over q (here, $A \approx 6$). The value of A depends on ρ and typically shrinks with increasing interaction strength V [25]. At the flux value $\varphi = 1/4$ we expect a realization of COMMs with $(q_1 + q_2)/2 = 2\varphi = 1/2$. In particular, the combination $\{q, -q + 1\}$ is a likely candidate for the ground state. However, especially for small cylinders, the favored combination of COMMs may vary with system size; therefore, different sets $\{q_1, q_2\}$ are separately compared for each system size.

Figure 2 shows the energy $E_{q,-q+1}(1/4)$ as a function of the circumference N_θ of the cylinder for $V_1 = 2.2t$ and, for comparison, $E_{q,-q}(0)$ and $E_{q,-q}(1/4)$. The ground state COMM q is the integer closest to $\sim N_\theta/A$ and switches to the next integer when N_θ increases by A. Since A is typically not commensurate with the lattice, there is a sensitive size dependence for small cylinders ($N_\theta \lesssim 300$)

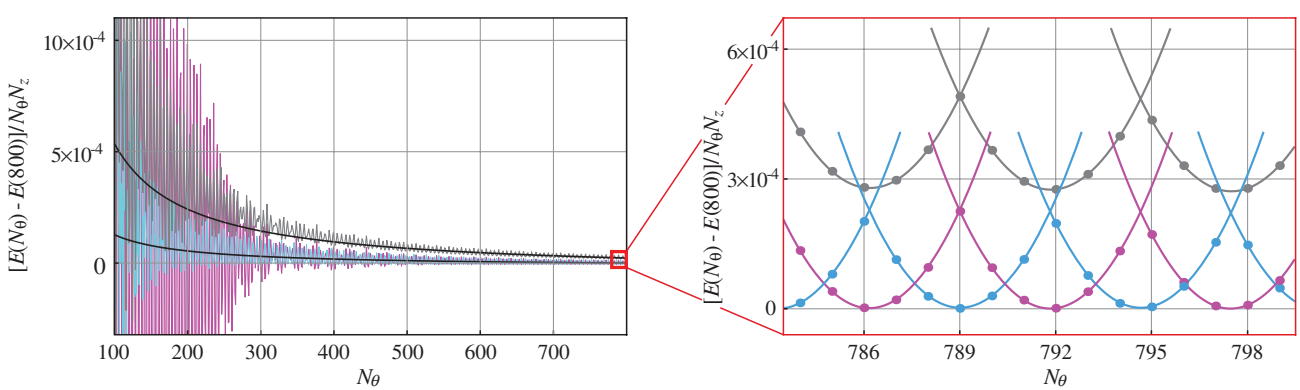

FIG. 2 (color). Energies $E_{q,-q+1}(1/4)/(N_\theta N_z)$ (blue), $E_{q,-q}(0)/(N_\theta N_z)$ (pink), and $E_{q,-q}(1/4)/(N_\theta N_z)$ (gray) per lattice site as a function of the circumference N_θ of the cylinder, for $N_z = 80$, $V_1 = 2.2t$, $\rho = 0.8$, and $t' = -0.3t$. Although $E_{q_1,q_2}(\varphi)$ oscillates with the cylinder's radius, for large enough cylinders $E_{q,-q}(0) = E_{q,-q+1}(1/4)$ is always smaller than $E_{q,-q}(1/4)$.

where the existence of an energy minimum at $\varphi = 1/4$ is not certain. For large cylinders, the total energy oscillates in N_θ with periodic minima at $N_\theta = nA$ and $N_\theta = nA/2$. The amplitude of the oscillations decreases with $1/N_\theta$, and in the limit $N_\theta \to \infty$ one finds that $E_{q,-q}(0) = E_{q,-q+1}(1/4)$ is always smaller than $E_{q,-q}(1/4)$.

The SC ground state around $\varphi = 1/4$ is therefore realized for the combination $\{q, -q+1\}$ and $E_{q,-q+1}(\varphi)$ is indeed minimal at $\varphi = 1/4$. Upon approaching $\varphi = 1/2$, the ground state switches to the combination $\{q+1, -q+1\}$ with an energy minimum at $\varphi = 1/2$ and to $\{q+1, -q+2\}$ around $\varphi = 3/4$ (see Fig. 3). For an arbitrary flux value, the combination of COMMs minimizing the energy is given by

$$q_1 + q_2 = \mathrm{floor}(4\varphi + 1/2), \tag{9}$$

where floor(x) is the largest integer smaller than x. The new energy minima correspond to fractional flux values $n\varphi/4$ and therefore lead to flux quantization in units of $\Phi_0/2$. This result remains valid also for thick-walled, irregular, or disordered cylinders, which can be modeled by averaging the total energy over various channels with different circumferences. In the limit $N_\theta \to \infty$, the amplitude of the flux oscillations vanishes proportional to the inverse radius of the cylinder [34]; the energy difference between the minima at $\phi = n/4$ and at $\phi = n/2$, however, vanishes with a higher order of the inverse radius. Consequently, also the flux periodicity of thermodynamic properties like the supercurrent is $\Phi_0/2$. This situation is illustrated in Fig. 3 for $N_\theta = 796$, for which $E_{q,-q}(0)$ and $E_{q,-q+1}(1/4)$ are almost equal. The ground state energy as a function of φ forms a series of parabolas with minima at $\varphi = n/4$. The supercurrent is obtained as

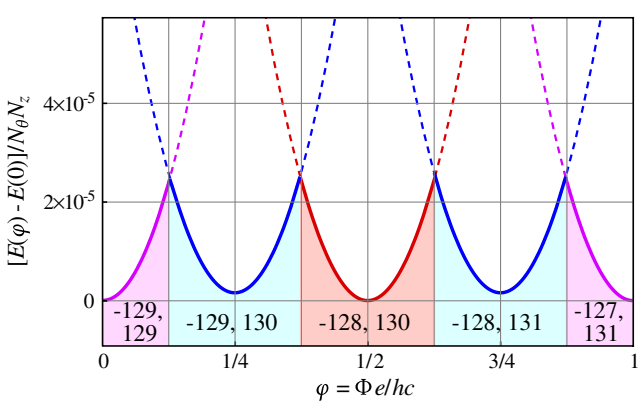

FIG. 3 (color online). Ground state energy $E(\varphi) = \min_{q_1,q_2} E_{q_1,q_2}(\varphi)$ for the same model parameters as in Fig. 2 with $N_\theta = 796$. The numbers in the shaded areas indicate the COMMs q_1 and q_2 in the corresponding flux regimes. The energy difference between the minima at $\varphi = 1/4$ and at $\varphi = 1/2$ corresponds to the difference between the blue and pink dots at $N_\theta = 796$ in Fig. 2. Whether the fluxless state is formed by two odd or two even COMMs depends on the system size N_θ.

$$J(\varphi) = -\frac{e}{hc} \min_{q_1,q_2} \frac{\partial E_{q_1,q_2}(\varphi)}{\partial \varphi} \tag{10}$$

and vanishes whenever the ground state energy has a minimum. Therefore, the formation of a SC state with half-integer valued phase-winding number $(q_1 + q_2)/2$ compensates the current induced by the half-integer magnetic flux $\Phi = \Phi_0(q_1 + q_2)/2$ and thereby minimizes the total energy of the system.

In finite systems, SC states with even and odd phase-winding numbers are distinct, and their energy minima at the corresponding flux values differ [35]. The flux periodicity is therefore strictly hc/e. However, the flux is always quantized in units of $hc/4e$, since the flux values where the energy minima occur do not depend on the system size. We thus identify three distinct classes of pair states on a flux-threaded cylinder (see Fig. 3): superpositions of two odd (purple) or two even (red) COMMs with energy minima at integer multiples of Φ_0 and even-odd (blue) combinations with energy minima at half-integer multiples of Φ_0. These pair states correspond to integer or half-integer phase-winding numbers of the SC order parameter. In principle, states with more than two different COMMs are possible as well. Such states will allow for fractional flux quanta of magnitude Φ_0/Q, but they are not realized as ground states in the parameter range of our model Hamiltonian. In alternative Hamiltonians, the prerequisite will remain the existence of an instability towards nodal superconductivity, which allows for fractional flux quantization due to coexisting COMMs.

In a real flux-threaded SC ring, flux always penetrates into the superconductor itself. The system is then no longer periodic in Φ, and the spatial variations of the magnetic field induce additional COMMs. Therefore, a unidirectional PDW state is expected to break down even in weak magnetic fields, leaving a standard d-wave superconducting ground state. Above this threshold field, $\Phi_0/2$ flux quantization will then disappear abruptly, returning to the flux quantum Φ_0. As described above (cf. Fig. 1), in YBCO SQUIDs the periodicity of the supercurrent changes indeed abruptly at a threshold magnetic field [12]. The low-field $\Phi_0/2$ oscillations were also reported to vanish close to T_c [12]. While the solutions of our model do not change qualitatively with temperature far below T_c, the PDW is also replaced by a d-wave BCS state upon approaching T_c.

Whether a PDW state occurs in cuprate superconductors is unsettled. However, uniaxial charge and spin modulations in coexistence with superconductivity were verified for 214 cuprates [36–39], which inevitably imply spatial modulations in the pair density [40–44]. In the typical rectangular geometry of the SQUID experiments, the orientation of an anticipated PDW would differ from the cylinder geometry discussed above. Although the emergence of SC states with half-integer phase-winding numbers and $\Phi_0/2$ periodicity will inevitably occur, too, the explicit solution of a model Hamiltonian in a SQUID

geometry is more involved—especially for a finite mismatch angle between the lattices on both sides of the Josephson junctions. A possible analysis of this geometry will require numerical simulations.

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