$hc/4e$ oscillations in a model for (100)/(110) SQUIDs of d-wave superconductors

L. M. León Hilario and A. A. Aligia
Centro Atómico Bariloche and Instituto Balseiro,
Comisión Nacional de Energía Atómica, 8400 S. C. de Bariloche, Argentina
(Dated: March 23, 2022)

We use a model of hard-core bosons to describe a SQUID built with two crystals of $d_{x^2−y^2}$-superconductors with orientations (100) and (110). Across the two faceted (100)/(110) interfaces, the structure of the superconducting order parameter leads to an alternating sign of the local Josephson coupling, and the possibility of quartet formation. Using a mapping of the boson model to an $XXZ$ model, we calculate numerically the energy of the system as a function of the applied magnetic flux, finding signals of $hc/4e$ oscillations in a certain region of parameters. This region has a large overlap to that at which binding of bosons exists.

PACS numbers: 74.20.Rp; 85.25.Dq; 85.25.Cp

I. INTRODUCTION

The interference between two superconducting condensates results in a wide range of interesting phenomena. In particular, it has been suggested that in the cuprates, the Josephson effect provides a direct probe of the angular dependence of the superconducting order parameter [1], and several type of phase sensitive experiments have been performed, which confirm the $d_{x^2−y^2}$-symmetry of the superconducting state in high-$T_c$ cuprates [2].

If two crystals of cuprate superconductors are connected at an interface, which is perpendicular to the (100) or (010) direction in both crystals, one can have a conventional Josephson junction (0-junction) or one with a sign reversal of the Josephson coupling (so called π-junction) depending on the mutual orientations of the $d_{x^2−y^2}$-wave superconducting order parameter [1] and [3]. In principle (neglecting multiple Andreev reflections [4]), for a perfectly flat (100)/(110) interface between two crystals of cuprate superconductors, the CuO2 lattices meet at 45°, such that a lobe (antinodal direction) of the $d_{x^2−y^2}$-order parameter of one superconductor points towards a nodal direction of the other, and therefore the Josephson current vanishes by symmetry. However, microscopic roughness allows for local Josephson tunneling across the interface facets, with the sign of the coupling depending on the orientation of each facet [5]. This leads to a variety of interesting effects like spontaneous supercurrent loops [5], locally time-reversal symmetry breaking phases [6], [7] and [8], or anomalous field dependencies of the critical current density [9].

A particularly interesting experimental observation in superconducting quantum interference devices (SQUIDs) with (100)/(110) interfaces is the $hc/4e$ periodicity of the critical current with applied magnetic flux [10]. This corresponds to periodicity of half a flux quantum. Since the natural explanation for the usual periodicity of one flux quantum $\Phi_0 = hc/2e$ is that the object that tunnels (a Cooper pair) has charge $2e$, a possible explanation of the periodicity of $hc/4e$ is that electrons tunnel in quartets, with total charge $4e$. Although at first sight this possibility seems exotic, it has been proposed before in nuclear physics, where a formation of a four-particle condensate was predicted at low density [11]. Furthermore, it has been proposed that pairing-quartetting competition is expected to be a general feature of interacting fermion systems [11]. Motivated by the dominance of the second harmonics (periodicity of $hc/4e$) of the dependence of the current with flux, Hlubina et al. [12] studied a model for fluctuations of the phase in an array of Josephson junctions. They conclude that the cuprates are close to an exotic phase with quartet condensation. Furthermore, while in principle one might think that a more conventional explanation of half flux periodicity is the vanishing of the first harmonic in a phenomenological Hamiltonian for Josephson junctions [10], the work of Hlubina et al. suggests that this is related with quartet formation. Thus, the microscopic origin of the dominance of the second harmonic seems to be the quartet condensation.

Recently, the alternating sequence of superconducting 0- or π-junctions (corresponding to a faceted (100)/(110) interface) was modeled by a bosonic lattice Hamiltonian with a staggered sign for the hopping amplitude across the facets [13]. Each boson represents a Cooper pair and the boson hopping through the facet is the Josephson coupling energy. As a consequence of partial frustration of the kinetic energy, the tendency towards boson pair formation in the presence of a weak attractive interaction is strongly enhanced and in some region of parameters the boson hopping through the facet is the Josephson coupling energy. A consequence of partial frustration of the kinetic energy, the tendency towards boson pair formation in the presence of a weak attractive interaction is strongly enhanced and in some region of parameters the boson hopping through the facet is the Josephson coupling energy. As a consequence of partial frustration of the kinetic energy, the tendency towards boson pair formation in the presence of a weak attractive interaction is strongly enhanced and in some region of parameters the boson hopping through the facet is the Josephson coupling energy. As a consequence of partial frustration of the kinetic energy, the tendency towards boson pair formation in the presence of a weak attractive interaction is strongly enhanced and in some region of parameters the boson hopping through the facet is the Josephson coupling energy. A consequence of partial frustration of the kinetic energy, the tendency towards boson pair formation in the presence of a weak attractive interaction is strongly enhanced and in some region of parameters the boson hopping through the facet is the Josephson coupling energy. As a consequence of partial frustration of the kinetic energy, the tendency towards boson pair formation in the presence of a weak attractive interaction is strongly enhanced and in some region of parameters the boson hopping through the facet is the Josephson coupling energy.
the amplitude of the superconducting order parameter are integrated out [14] and [15], as stated above, one might expect that the approaches of Refs. [12] and [13] are related and that the appearance of higher harmonics in the Josephson current is directly connected with quartet formation.

In this work, we calculate the energy as a function of the flux $E(\Phi)$ for a simple model describing the motion of Cooper pairs in a SQUID with two (100)/(110) interfaces, as in the experimental situation [10] (see Fig. 1). Since the current through the ring $j(\Phi)$ is proportional to $\partial E/\partial \varphi$ for a given geometry, the oscillations in $E(\Phi)$ are directly related to those of the current. While some ideas were reported before [13], the calculation of pairings was limited to the dilute case of very few bosons in the system, and $E(\Phi)$ was not calculated. In Section 2 we explain the model and its mapping to a spin $1/2$ model. The results of the numerical solution of this model are presented in Section 3. Section 4 contains a short summary and discussion.

$\mathbf{II. \hspace{1em} THE \hspace{1em} MODEL}$

We describe the Cooper pairs as bosons moving on a lattice [13]. At the interfaces, the boson hopping across a facet represents the Josephson coupling energy of the quantum phase Hamiltonian.

The Hamiltonian is [13] (see Fig. 1)

$$H = H_1 + H_2 + H_3$$

$$H_1 = \sum_{\alpha i} (-t(-1)^i a^\dagger_{\alpha i+1} a_{\alpha i} - t' a^\dagger_{\alpha i+2} a_{\alpha i} + H.c.),$$

$$H_2 = \sum_{\alpha i} [V a^\dagger_{\alpha i} a_{\alpha i} a^\dagger_{\alpha i+1} a_{\alpha i+1} + U (a^\dagger_{\alpha i} a_{\alpha i} - 1) a^\dagger_{\alpha i} a_{\alpha i}],$$

$$H_3 = -t_\perp \sum_j (a^\dagger_{ij} a_{2j} e^{i\varphi(-1)^j/2} + H.c.)$$

where $a^\dagger_{\alpha i}$ creates a boson (Cooper pair) at site $i$ of the interface $\alpha = 1, 2$. The first term $H_1$ describes the kinetic energy of the bosons within each interface. The hopping with amplitude $t$ describes Josephson tunneling across the (100)/(110) interface and has an alternating sign due to the roughness of the surface and the different change of phase of the superconducting order parameter. The term in $t'$ describes motion along the interface without crossing it. The second term $H_2$ represents an on-site $U$ and a nearest-neighbor $V$ interaction. The last term $H_3$ describes in the simplest possible way the motion of Cooper pairs from one interface to the other. The phase $\varphi$ is related to the magnetic flux threading the ring by $\varphi = 2\pi \Phi/\Phi_0$, where $\Phi_0 = \hbar c/2e$ is the flux quantum. For simplicity we take $U \rightarrow +\infty$. This implies hard-core bosons and allows to map the model into a spin-1/2 model as described below. The nearest-neighbor interaction is assumed attractive ($V < 0$) unless otherwise stated. Its origin can be the same as the magnetic pairing interaction in the cuprates [13]. From results of the equivalent spin-1/2 XXZ model for one interface [16], we expect that inclusion of second nearest-neighbor attraction does not change essentially the physics. The values of the parameters were estimated as $V \sim -250$ K, $t' > t \sim 100$ K [13]. These parameters favor quartet formation at one (100)/(110) interface, but not in conventional interfaces with 0-junctions only [13].

The alternating sign in the first term of Eq. (1) can be eliminated through an adequate change of phases of the boson operators,

$$b^\dagger_{\alpha 4i} = -a^\dagger_{\alpha 4i}, \quad b^\dagger_{\alpha 4i+1} = -a^\dagger_{\alpha i+1},$$

$$b^\dagger_{\alpha 4i+2} = a^\dagger_{\alpha i+2}, \quad b^\dagger_{\alpha 4i+3} = a^\dagger_{\alpha 4i+3}$$

which transforms the first term into

$$H_1 = \sum_{\alpha i} [-t b^\dagger_{\alpha i+1} b_{\alpha i} + t' b^\dagger_{\alpha i+2} b_{\alpha i} + H.c.]$$

while the other terms retain the same form with the replacement $a_{\alpha i} \rightarrow b_{\alpha i}$.

Using a boson-spin transformation

$$S^+_{\alpha i} = (-1)^i b^\dagger_{\alpha i}, \quad S^-_{\alpha i} = (-1)^i b_{\alpha i}, \quad S^z_{\alpha i} = b^\dagger_{\alpha i} b_{\alpha i} - \frac{1}{2}$$

the model is mapped into the following spin-1/2 model

$$H_S = \sum_{\alpha i} [J_1 (S^x_{\alpha i} S^x_{\alpha i+1} + S^y_{\alpha i} S^y_{\alpha i+1} + \Delta S^z_{\alpha i} S^z_{\alpha i+1})$$

$$+ J_2 (S^x_{\alpha i} S^x_{\alpha i+2} + S^y_{\alpha i} S^y_{\alpha i+2})]$$

$$+ J_3 \sum_j (e^{i\varphi(-1)^j/2} S^+_{ij} S^-_{2j} + H.c.)/2$$

FIG. 1: Sketch of a SQUID with two faceted (100)/(110) interfaces and the simple Hamiltonian Eq. (1) used to describe it.
with the spin exchange coupling constants $J_1 = 2t$, $J_2 = 2t'$, $J_3 = 2t$, and the anisotropy parameter $\Delta = V/2t < 0$. For $J_3 = 0$, the model describes two uncoupled XXZ chains representing the interfaces. If periodic or antiperiodic boundary conditions are used for each chain, the Hamiltonian Eq. (5) can be thought as describing two XXZ rings, one on top of the other with a complex spin-flip interaction $J_3$ between them, the phase of which alternates sign between odd and even sites of the rings. The spin-1/2 XXZ chain has been studied before in the context of metamagnetic transitions and results for magnon binding (bosons in the original language) and phase separation were obtained in specific parameter regimes [16].

The region of parameters for which binding of two bosons occur, and clustering in more than two bosons or phase separation takes place has been studied in more detail in Ref. [13]. Taking $t = 1$ as the unit of energy, the region most favorable for pairing of bosons can be defined roughly as $t' > 1$ and $1 < -V < 2t' + 2$. In the following section, we report our search for signals of $hc/4e$ oscillations in $E(\Phi)$ in parameter space and for a finite concentration of bosons.

For this model, the current in the loop is given by [17]

$$j(\Phi) = \frac{2e}{\hbar} \frac{\partial \langle H \rangle}{\partial \varphi} = \frac{2eN_s}{\hbar} \frac{\partial E(\Phi)}{\partial \varphi}$$

(6)

where $N_s = 2L$ is the number of sites in the system, and $L$ is the number of sites in one ring.

### III. RESULTS

We have studied by numerical diagonalization the equivalent spin Hamiltonian Eq. (5) for two rings of $L$ sites each and a number $N$ of up spins (bosons in the original language) in a background of down spins. The total spin projection $S_z = N - L/2$ is a conserved quantity in Eq. (5). In general, we have taken $8 \leq L \leq 12$. For small $N$ some calculations were done also for $L = 14$ (28 sites counting both rings). We have chosen for each ring the boundary conditions (BC) periodic or antiperiodic, which lead to the minimum ground state energy per site as a function of flux $E(\Phi)$. In all studied cases, these BC did not change as the flux $\Phi$ was varied. Actually in general, and particularly in the region of binding, the BC which lead to the minimum energy are twisted [16] and [19]. However, the deviation from periodic or antiperiodic BC is small for $t' \geq t$ and we believe that our choice does not affect the results.

The energy per site $E(\Phi)$ is periodic in one flux quantum: $E(\Phi) = E(\Phi + \Phi_0)$. We have investigated the tendency towards a periodicity of half a flux quantum in $E(\Phi)$. This is reflected in the presence of two relative minima in $E(\Phi)$ for $\Phi = 0$ and $\Phi = \Phi_0/2$. For $N = 2$ and $N = 4$, we have explored the region of parameters $-3 \leq t'/t \leq 3, -8 \leq V/t \leq 0$, examining in more detail the region of binding determined previously [13] (roughly $t'/t > 1$ and $1 < -V/t < 2t' + 2, t > 0$). For only two bosons in the system ($N = 2$), the region of binding has been determined analytically in the thermodynamic limit [13] and [16]. Rather surprisingly, we do not find two minima for $N = 2$ in all the explored region of parameters. Instead, for $N = 4$, we do find clear signals of a periodicity in $\Phi_0/2$, and only inside the region of binding.

An example is shown in Fig. 2 for $t'/t > 1$ and $L = 8$. Similar results are obtained for larger system sizes and moderately larger values of $t'/t$. However, keeping $N = 4$ and increasing $L$ up to $L = 14$, the relative minimum of $E(\Phi)$ at $\Phi = 0$ becomes less pronounced, suggesting that it disappears and there is no periodicity of $E(\Phi)$ in $\Phi_0/2$ in the thermodynamic limit for dilute systems $N/L \to 0$. In any case, the dilute limit is not expected to be relevant to the experimental situation. To be able to analyze how $E(\Phi)$ evolves with system size at a finite fixed concentration of bosons, we calculated $E(\Phi)$ for increasing $L$, keeping $N = L$ and the other parameters fixed. Note that due to the symmetry of the spin Hamiltonian Eq. (5) under a change of sign $S_z$, the problems with $N$ or $2L - N$ bosons are equivalent. Therefore, the case with $N = L$ is that of the greatest possible total kinetic energy in absence of the interaction $V$. In Fig. 3 we show how $E(\Phi)$ changes as $L$ increases from 8 to 12 (in even steps to avoid frustration) and other parameters as in Fig. 2. The absolute minimum is at $\Phi = 0$ and there is a relative minimum at $\Phi = \Phi_0/2$. This minimum is rather shallow for $L = 8$, but becomes more pronounced as $L$ increases. This is consistent with a periodicity of $E(\Phi)$ in half a flux quantum in the thermodynamic limit. For a reasonable value of the Josephson coupling energy $t \sim 10^{-2}$ eV, one has $4eIt/\hbar \approx 10\mu A$. Since the number of facets in a real system is of the order of $L = 10^3 - 10^4$ [18], from Eq. (6), the current extrapolated to the thermodynamic limit is

$$j \sim 1 - 10\mu A.$$  

This in rough agreement with the experimental order of magnitude $j \sim 10 - 100\mu A$ [10]. Since roughly $j \sim t_{\perp}$ (see below) the agreement improves if $t_{\perp}$ is increased.

The evolution of $E(\Phi)$ with size for larger $t'$ is shown.
FIG. 3: Ground state energy per site as a function of the applied flux for \( t'/t = 1, V/t = -2, t_\perp/t = 0.1, N = L \) and different system sizes \( L \).

FIG. 4: Same as Fig. 3 for \( t'/t = 2 \). The dashed line at the bottom is the result for \( t_\perp/t = 0.2 \) multiplied by a factor 0.2.

in Fig. 4. In the spin language, these parameters correspond to a next-nearest-neighbor antiferromagnetic interaction in the \( x \) and \( y \) directions \( J_2 \) two times larger than the corresponding one for nearest neighbors \( J_1 \). In this situation, in the classical case, an antiferromagnetic order between next-nearest neighbors is favored and to avoid frustration of this order in a ring, the number of sites \( L \) should be multiple of 4. As a consequence, although this frustration is only partial in the quantum case, we believe that the results for \( N = L = 10 \) are not reliable for \( t' > t \). This is the case of the middle panel in Fig. 4. Comparing the other two cases represented in Fig. 4, although none of them shows two well defined minima, the results are not inconsistent with a double periodicity in the thermodynamic limit because \( E(\Phi) \) has a rather sharp maximum for \( N = L = 8 \) at \( \Phi = 0.5\Phi_0 \) that becomes rather flat for \( N = L = 12 \), suggesting the development of a minimum as \( L \) is further increased. Note that for \( t_\perp/t = 0.2 \), a relative minimum in \( E(\Phi) \) at \( \Phi = 0.5\Phi_0 \) already exists for \( N = L = 12 \) (dashed line in Fig. 4). We have also analyzed the effect of varying hopping between interfaces \( t_\perp \). As \( t_\perp \) increases, the tendency towards an additional periodicity increases slightly up \( t_\perp/t \sim 0.2 \) and for larger \( t_\perp \), it is weakened. This is expected, since the additional kinetic energy tends to break the bound pairs. For small \( t_\perp \), from perturbation theory, the structure of \( E(\Phi) \) is dominated by several terms of order \( t_\perp^2 \). This is in rough agreement with our numerical results for \( t_\perp/t \sim 0.2 \), although the increase in the amplitude of \( E(\Phi) \) with \( t_\perp \) seems faster than quadratic (see for example the bottom of Fig. 4).

Most of the above-mentioned results correspond to the half filled case \( N = L \). We have also looked for signals of an extra periodicity in \( E(\Phi) \) in the quarter filled case \( N = L/2 \), comparing the flux dependence of the energy for \( L = 8 \) and \( L = 12 \). The results suggest that no double periodicity is present in the thermodynamic limit.
Finally, we also study the shape of $E(\Phi)$ for repulsive interaction $V > 0$ in the half filled case. Surprisingly, we find a tendency for double periodicity for positive $t'$ (see Fig. 5). We have studied the binding energy of an isolated ring $\Delta_b = L[E_1(N + 2) + E_1(N) - 2E_1(N + 1)]$ for different parameters, where $E_1(N)$ is the energy per site of one ring for $N = L/2$ particles. The region of binding coincides roughly with that obtained previously for $N = 0$ [13] and [16] and $\Delta_b > 0$ for positive $V$. The absence of a negative binding energy for positive $V$ also persist when $t_\perp$ is included. The factor of 10 increase in the magnitude of the oscillations as $L$ is increased form 8 to 12 points to particularly large finite-size effects. In all cases we do not find signs of extra periodicity for negative $t'$. We remind the reader that negative $t'$ corresponds to the case of ordinary junctions in the facets (all 0-junctions), for which no extra periodicity was experimentally observed.

**IV. SUMMARY AND DISCUSSION**

We have calculated numerically the energy per site as a function of the applied magnetic flux $E(\Phi)$ in a simplified model for hard-core bosons (representing Cooper pairs) for a SQUID containing two (100)/(110) interfaces. In spite of the limitations of the size of the systems studied, the finite-size scaling suggests $hc/4e$ periodicity for parameters for which binding of bosons at one interface take place [13]. This fact and the magnitude of the resulting current is consistent with the experiments of Schneider et al. [10]. This periodicity is absent for usual interfaces ($t' < 0$ in the model).

We also find signals of $hc/4e$ oscillations in $E(\Phi)$ for repulsive interactions for which no binding exists. The reason of the extra periodicity in this case is unclear, and needs further study.

For simplicity the motion of Cooper pairs inside one crystal from one interface to the other has been represented by only one hopping parameter $t_\perp$. This is a rather crude description which allowed us to include more sites at the interfaces, keeping the total amount of sites below 32 due to computer limitations. In any case, we expect that the essential physics is retained, at least for attractive interaction between Cooper pairs. The physical picture can be the following: quartets (bound pairs of bosons) are formed at the interfaces as a consequence of the competition between attraction and the reduced kinetic energy there [13]. The energy scale of the motion perpendicular to the interface ($t_\perp$ in our model) is responsible for the coherence and the observed flux dependence. However, if it is too large, it tends to unbind the quartets. If this image is correct, and quartets live only near the interface, the observation of $hc/4e$ oscillations should depend on the size of the system. They should be weaker or disappear if the coherence length in the direction normal to the interface is much smaller than the size of the system.

A more realistic model for the description of the motion of Cooper pairs between both interfaces should include one or more layers of intermediate sites between the interfaces.

**Acknowledgments**

A.A.A. wants to thank A. Kampf, T. Kopp and J. Mannhart for useful discussions. This work was sponsored by PICT 03-13829 of ANPCyT. L.M.L.H is a fellow of the IB-ICTP Diploma Program. A.A.A. is partially supported by CONICET.
Kirtley, P.J. Hirschfeld and J. Mannhart, Europhys. Lett. 68 (2004), p. 86.
11 G. Rpke, A. Schnell, P. Schuck and P. Nozières, Phys. Rev. Lett. 80 (1998), p. 3177.
12 R. Hlubina, M. Grajcar, J. Mrz, preprint cond-mat/0304213 in press.
13 A.A. Aligia, A.P. Kampf and J. Mannhart, Phys. Rev. Lett. 94 (2005), p. 247004.
14 M.P.A. Fisher and G. Grinstein, Phys. Rev. Lett. 60 (1988), p. 208.
15 M.P.A. Fisher, P.B. Weichman, G. Grinstein and D.S. Fisher, Phys. Rev. B 40 (1989), p. 546.
16 A.A. Aligia, Phys. Rev. B 63 (2000), p. 014402 therein.
17 A.A. Aligia, Phys. Rev. B 66 (2002), p. 165303.
18 J. Mannhart, private communication.
19 A.A. Aligia, C.D. Batista and F.H.L. Eler, Phys. Rev. B 62 (2000), p. 3259.