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A quantum interferometer for quartets in superconducting three-terminal Josephson junctions

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An interferometric device is proposed in order to analyze the quartet mode in biased three-terminal Josephson junctions (TTJs), and to provide experimental evidence for emergence of a single stationary phase, the so-called quartet phase. In such a quartet-Superconducting Quantum Interference Device (quartet-SQUID), the flux sensitivity exhibits period $hc/4e$, which is the fingerprint of a transient intermediate state involving two entangled Cooper pairs. The quartet-SQUID provides two informations: an amplitude that measures a total "quartet critical current", and a phase lapse coming from the superposition of the following two current components: the quartet supercurrent that is odd in the quartet phase, and the phase-sensitive multiple Andreev reflection (phase-MAR) quasiparticle current, that is even in the quartet phase. This makes a TTJ a generically "θ-junction". Evidence for phase-MARs plays against conservative scenarii involving synchronization of AC Josephson currents, based on "adiabatic" phase dynamics and RSJ-like models.

Introduction: Multiterminal Josephson junctions (MTJs) [1–4] appear as a very fertile evolution in the field of superconductivity. While unbiased MTJs offer prospects as platforms for controllable topological properties [5–23], biased MTJs reveal new channels for both superconducting phase-sensitive and quantum mechanical DC currents, as predicted by theory [22, 24–42] and confirmed in experiments [43–55]. A paradigm of multiterminal Josephson junction [26] involves three superconductors biased at the opposite voltages $0,V,-V$, this making the junction host Cooper quartets [26]. Those transient quartets are made of entangled pairs of Cooper pairs and flowing from the unbiased terminal towards the two others simultaneously. This voltage configuration ensures energy conservation, a necessary condition for having DC Josephson currents. The quartet mechanism goes together with emergence of a stationary phase combination of the three terminal phases, the so-called quartet phase φ_Q. At the microscopic level, the minimal process appearing in perturbation theory in the tunnel amplitudes consists of four Andreev reflections. Quartets (as well as higher-order multipairs such as sextets, octets, ...) therefore constitute a genuine quantum mechanical mesoscopic phenomenon, not occurring in simple classical Josephson arrays but instead in truly multiterminal junctions.

Besides this quartet supercurrent, another current component happens to depend on the quartet phase. It originates from multiple Andreev reflections (MAR), which promote quasiparticles across the superconducting gap 2Δ with the help of Cooper pair transfers, each one gaining energy $2eV$ [56]. New channels open in a three-terminal Josephson junction (TTJ) [34], where all pairs of terminals are simultaneously involved. Among those, specific processes involve emission of quartets at zero energy but with phase φ_Q: the energy cost for promoting a quasiparticle between two terminals, say S_1, S_0 (with $V_1-V_0=V$), instead of transferring a pair between terminals S_1, S_0, can be provided by transferring a pair between terminals S_0, S_2 (with $V_0-V_2=V$) plus absorbing simultaneously a quartet from (S_1, S_2) to S_0 (Figure 1). This quartet carries a phase φ_Q and these MAR processes become phase-dependent subgap quasiparticle currents [27]. Detailed calculations about the phase and voltage sensitivities of both quartet and phase-MAR currents can be found in Refs. 31 and 57. While quartet supercurrents are truly nondissipative, the phase-MAR currents are dissipative. Both of them depend on the control variables (φ_Q,V) but with different symmetries [27, 31]. Owing to time inversion symmetry, the quartet and phase-MAR currents have to be antisymmetric with respect to inverting both variables φ_Q and V. The quartet current is antisymmetric in phase and symmetric in voltage, but the phase-MAR current is instead symmetric in phase and antisymmetric in voltage. This duality is reminiscent of the tunnel junction treated by Josephson [58] in his seminal work, concerning the DC current and the phase-sensitive quasiparticle current. The latter is AC in a two-terminal junction, but can become DC in a multiterminal one.

Regarding experiments, an important question is about the interpretation of transport anomalies observed when a TTJ is biased at the voltages $0,V,-V$ [43, 45, 47, 53–55]. A conservative explanation involves the synchronization of AC Josephson currents flowing across each of the junctions polarized at V and $-V$ respectively [59]. This mechanism is electromagnetic in nature and it involves the impedance (or the photon modes) of the whole circuit including the junction.

Minimal models involve an adiabatic dependence of the currents with time-dependent phases, in a way similar to the standard treatment of Shapiro resonances [60]. This can be done in the presence of an external environment described by a circuit impedance, that includes the resistive part of the junction itself, within RSJ-related models [59]. This qualitatively accounts for the DC-current features observed in TTJs [46, 48–50, 53], but is not a proof of the physical relevance of such a description. For instance, the zero-frequency current-current cross-correlations [45] can hardly receive interpretation in terms of the RSJ model, and quite specific frequency dependence of the device external circuit impedance should be advocated to interpret a recent four-terminal experiment as originating from a RSJ model [47]. Still, complementary experiments would be important to ascertain the mesoscopic nature of multipair processes.

A first requisite is the control over the quartet phase, which can be used to prove coherence of the multipair supercurrent. Such a phase coherence might be present in an extrin-

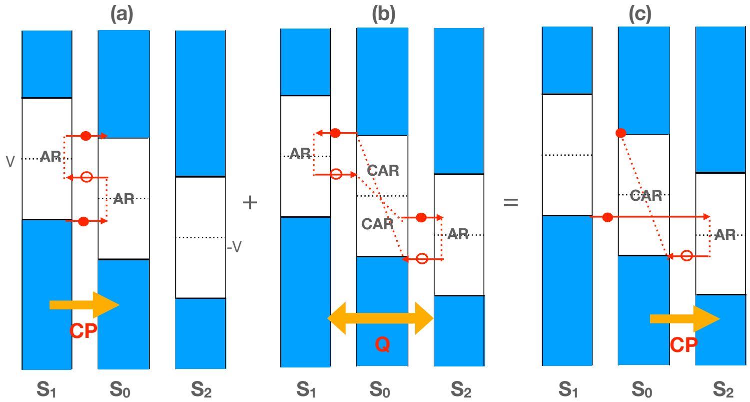

FIG. 1. Diagram (a) pictures a quasiparticle promoted through the gap and a Cooper pair (CP) transferred from S_1 to S_0, via two Andreev reflections (AR). Diagram (b) pictures a quartet (Q) formed of two entangled Cooper pairs, transferred from S_0 to S_1 and S_2 simultaneously, with two AR and two crossed Andreev reflections (CAR). Diagram (c) pictures a quasiparticle promoted through the gap, transferred from S_1 to S_0, while a Cooper pair is instead transferred from S_0 to S_2. Diagram (c) can be formally obtained by superimposing the lines of diagrams (a) and (b), showing that phase-dependent MARs involve quartets.

sic synchronization scenario, although hampered by decoherence mechanisms due to the environment itself. On the contrary, phase coherence of the quartets is expected to be much more robust. To go further in the discrimination between extrinsic and intrinsic mechanisms, one must take into account the high transparency of the junctions, necessary to produce a mesoscopic multipair transport. The consequence is the existence of MAR processes, which in the standard two-terminal case have no explanation but with the help of subgap Andreev reflections, and thus go well beyond a phenomenological RSJ modeling. Specifically, in MTJs, the observation of the phase-sensitive MARs can be taken as evidence for truly mesoscopic processes involving quartets, thus disproving any classical synchronization scenario.

In this work, we propose an interferometric scheme able to control the quartet phase and, at the same time, reveal the phase-MAR component, thus proving both the phase coherence of multipair processes and their truly subgap mesoscopic nature.

Following Josephson's discovery that a current must flow in an unbiased junction and depends on the phase difference between the contacts [58], SQUID setups were invented in order to control and analyze this phase sensitivity [60]. The flux dependence exhibits period $hc/2e$ that directly proves supercurrents carried by Cooper pairs with charge $2e$. Similarly, one expects that interferometry also helps elucidating the mechanism of quartets in TTJs, in particular proving that they carry a charge 4e. Yet, this simple expectation meets a difficulty : a TTJ involves three terminals, two of them being biased. This prevents from building a trivial generalization of the original two-terminal SQUID which is fully equipotential. Such a device must necessarily be different from those already proposed for multijunctions at equilibrium [28, 44].

In this work, we describe a four-terminal scheme building a true quartet-SQUID. The clue is to connect two TTJs in parallel by their unbiased as well as their biased terminals, in order to close them in a double-TTJ loop. Cooper pairs injected in the quartet-SQUID at voltage $V = 0$ can cross either TTJ as quartets, picking up the quartet phase of each TTJ, and recombine in the common outputs at voltages V and $-V$. The design encloses two loops instead of one. Generalizing the standard SQUID argument in the presence of magnetic flux shows that this imposes a difference between the quartet phases of the two TTJs, thus achieving a perfect parallel with an ordinary SQUID.

This scheme allows analyzing the sensitivity of the quartet mode on voltage, as a new control parameter for a DC supercurrent. Microscopic models show that it is not monotonous, owing to nonadiabatic transitions between Andreev levels. Moreover it can switch from a generic π-junction behavior (perturbative and low voltage case) to a 0-junction one. Such an evidence goes beyond classical synchronization scenarii unless assuming ad hoc an unlikely voltage (i.e. AC Josephson frequency) dependence of the circuit impedance.

The proposed quartet-SQUID also allows exploiting the interplay between quartets and phase-sensitive MARs. Separation between those two distinct processes could in principle be achieved in ideally symmetrical TJJs. More generally, the different phase symmetry of quartet and phase-sensitive MAR currents results in a phase lapse in the periodic flux response of the quartet-SQUID. Measuring this phase lapse quantifies the presence of phase-MARs in transparent enough junctions. Phase-MARs are mesoscopic and they involve quartet excitation amplitudes, therefore they bring the necessary proof of a truly new physics being involved in TTJs.

Three-terminal junction quartet-SQUID: The principle of the quartet-SQUID is to make two TTJs interfere with each other by joining their biased arms in a secondary circuit, as pictured in Figure 2. The two TTJs thus enclose a secondary loop with two branches respectively at the voltages V (hereafter denoted as "L-branch"), $-V$ (hereafter denoted as "R-branch"), threaded by a flux ϕ. The main loop is threaded by a flux Φ. Both loops are separated by the L branch (see Figure 2). The total current is injected as $I_{tot} = I_{1M} + I_{2M}$ where I_{1M} and I_{2M} are the currents entering each of the TTJs from the unbiased branch, and eventually exiting in the biased branches

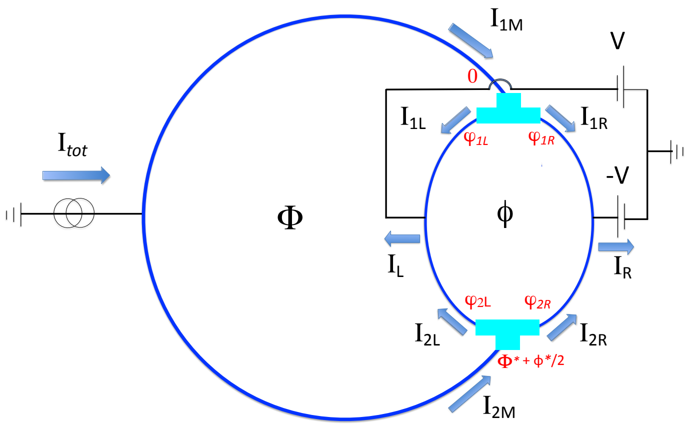

FIG. 2. Scheme of a quartet-SQUID based on two TTJs, with a main loop threaded by a flux Φ and a secondary loop threaded by a flux ϕ. The current is injected into the large loop such as to make the two TTJs interfere with each other. The current exits through the biased leads at voltages $V, -V$ of the branches L, R of the secondary loop. The phases are mentioned in red within a simple gauge convention, see text. Here $\varphi_{1M} = 0$ and $\varphi_{2M} = \Phi^* + \phi^*/2$.

(second circuit) as I_L and I_R. Current conservation reads:

$$I_{tot} = I_{1M} + I_{2M} = I_L + I_R \tag{1}$$

Let us define the phases at the unbiased branch of TTJ1 and TTJ2 as $\varphi_{1M}, \varphi_{2M}$ respectively, and the phases at the biased branches of the TTJs as $(\varphi_{1L}, \varphi_{1R})$ and $(\varphi_{2L}, \varphi_{2R})$ respectively. From previous works [26] one knows that the stationary quartet phase components are

$$\varphi_{Qi} = \varphi_{iL} + \varphi_{iR} - 2\varphi_{iM}, \tag{2}$$

while the oscillating phase components (at frequency $4eV/\hbar$) are $\varphi_{iL} - \varphi_{iR}$ $(i = 1, 2)$. Let us define the normalized fluxes between 0 and 2π as $\Phi^* = (2\pi/\phi_0)\Phi$ and $\phi^* = (2\pi/\phi_0)\phi$, with $\phi_0 = hc/2e$. The fluxoid argument is applied to the main loop containing the L branch, then to the main plus secondary loop containing the R branch. This is perfectly allowed, in spite of the main loop and the biased branches not being at the same potential. In fact, the fluxoid argument takes care of the phase variation inside each superconductor, whatever its potential. The supercurrent circulation in the bulk of each superconductor is assumed to be zero as for a thick superconductor, see Ref. 60. The presence of voltage biases between the different superconductors only enters in the phase difference at the junctions, that can depend on time in the present scheme, with frequency $2eV/\hbar$. The fluxoid argument [60] amounts to equating on both paths the sum of the phase differences at the junctions to the normalized flux in the loop (modulo 2π), which yields:

$$\Phi^* = \varphi_{1L} - \varphi_{1M} + \varphi_{2M} - \varphi_{2L} \tag{3}$$
$$\Phi^* + \phi^* = \varphi_{1R} - \varphi_{1M} + \varphi_{2M} - \varphi_{2R}. \tag{4}$$

Taking the difference between these two equations, one obtains a relation between the oscillating phases components at the two TTJs:

$$(\varphi_{1R} - \varphi_{1L}) - (\varphi_{2R} - \varphi_{2L}) = \phi^*, \tag{5}$$

expressing that these time-dependent components are perfectly synchronized. On the other hand, taking the sum of Eqs. (3)-(4) yields a relation between the quartet phases of the two TTJs [see Eq. (2)]:

$$\varphi_{1Q} - \varphi_{2Q} = 2(\Phi^* + \phi^*/2) \tag{6}$$

This central result shows that, like an ordinary SQUID, the interferometer imposes a phase difference between the stationary quartet phases at the two TTJs. Because of the (L, R) symmetry of the quartet current, the corresponding flux is the arithmetic mean of the fluxes delimited by the L (i.e. Φ^*) and the R (i.e. $\Phi^* + \phi^*$) branches.

Interestingly, if the TTJs are symmetric by exchanging their contacts to branches L, R, the currents $I_{1,2M}$ entering the TTJs are pure quartet currents i.e. $I_{1M} = I_{1Q}(\varphi_{1Q}), I_{2M} = I_{2Q}(\varphi_{2Q})$. In turn, a pure MAR current $I_L - I_R$ flows between branches L and R thus in the secondary circuit, and one can write:

$$I_L = \frac{1}{2}\left[I_{1Q}(\varphi_{1Q}) + I_{2Q}(\varphi_{2Q})\right] + I_{1MAR}(\varphi_{1Q}) + I_{2MAR}(\varphi_{2Q}) \tag{7}$$
$$I_R = \frac{1}{2}\left[I_{1Q}(\varphi_{1Q}) + I_{2Q}(\varphi_{2Q})\right] - I_{1MAR}(\varphi_{1Q}) - I_{2MAR}(\varphi_{2Q}). \tag{8}$$

In this case, a Cooper pair current circulates in the main loop, while the secondary one contains a superposition of a quartet current – flowing as parallel Cooper pair currents in the L and R branches, thus insensitive to the flux ϕ^* – and a circulating MAR current – sensitive to ϕ^* via its phase-MAR component.

In the general case of asymmetric TTJs, all currents $I_{1,2M}, I_{L,R}$ contain components of both quartet and MAR origins. One can carry the analysis further in the simplifying case of weak transparencies. Noting that the quartet and MAR components are respectively odd and even in the quartet phases, and one can write:

$$I_{1M} = I_{Qc1}(V)\sin\varphi_{1Q} + \bar{I}_{MAR1}(V) + I_{MARc1}(V)\cos\varphi_{1Q} \tag{9}$$
$$I_{2M} = I_{Qc2}(V)\sin\varphi_{2Q} + \bar{I}_{MAR2}(V) + I_{MARc2}(V)\cos\varphi_{2Q} \tag{10}$$

where the first terms in Eqs. (9)-(10) are the quartet currents, with "critical currents" I_{Qci}. The second terms are the phase-averaged MAR components, including the phase-independent two-terminal MAR processes, and the last terms contain the phase-sensitive MAR components, with "critical currents" I_{MARci}. The critical current values defining the amplitude of the phase oscillations of the quartet and MAR currents are voltage-sensitive, and have in general nonmonotonous variations with V [31, 37, 38, 42, 57]. The sine and cosine dependences of the respective quartet and MAR currents stem from their symmetry in phase. Such expressions can be checked by microscopic calculations in the low transparency case [31].

From Eqs. (1), (6), (9) and (10), the total current injected in this quartet-SQUID can be written as (omitting the voltage sensitivities):

$$I_{tot} = \bar{I}_{MAR1} + \bar{I}_{MAR2} + I_{c1}\sin(\varphi_{1Q} + \alpha_1) \tag{11}$$
$$+ I_{c2}\sin\left(\varphi_{1Q} - 2(\Phi^* + \phi^*/2) + \alpha_2\right), \tag{12}$$

with $(i = 1,2)$:

$$I_{ci} = \sqrt{I_{Qci}^2 + I_{MARci}^2}$$
$$\tan(\alpha_i) = I_{MARci}/I_{Qci}. \tag{13}$$

The total current appears as the sum of (i) a phase-independent MAR current and (ii) a typical SQUID current, which depends on the quartet phase φ_{1Q}, and on the effective flux $\Phi^* + \phi^*/2$, with phase lapses $\alpha_{1,2}$ that measure the ratio of phase-sensitive MAR currents to quartet currents. As in a usual SQUID, maximizing the total current with respect to the (quartet) phase yields the following expression for the critical current:

$$I_{tot} = \bar{I}_{MAR1} + \bar{I}_{MAR2} \tag{14}$$
$$+ \left[I_{c1}^2 + I_{c2}^2 + 2I_{c1}I_{c2}\cos\left(2(\Phi^* + \phi^*/2) + \alpha_1 - \alpha_2\right)\right]^{1/2}.$$

This relation achieves the goal of building a quartet-SQUID. As a first result, the factor 2 in the flux sensitivity, that results in a $hc/4e$ periodicity, manifests the fact that quartets are made of two entangled Cooper pairs and carry charge $4e$. Second, the phase lapses $\alpha_{1,2}$ directly contain the information about the presence or not of phase-MARs. These phase lapses disappear in the case of TTJs with symmetric branches $V, -V$ ($\alpha_{1,2} = 0$) or in the unlikely case of identical TTJs ($\alpha_1 = \alpha_2$).

In experiments performed at low voltage and in incoherent diffusive regimes, the MAR currents are negligible, and the quartet-SQUID gives direct access to the pure quartet currents.

The above discussion is not restricted to harmonic dependences of the quartet and MAR current with phase. In resonant dot models, nonharmonic behavior is easily obtained and the quartet current can be quite large, actually comparable to the ordinary Josephson current of a two-terminal junction in the same conditions [27, 31].

Exploring the voltage dependence: from "0−" to "π−" junction: Having a quartet-SQUID in hands allows a thorough study of the dependence of the quartet (and phase-MAR) currents with voltage, as a new control parameter for DC Josephson currents. Focusing on the quartet current, different models, suited to different types of junctions (single or many level quantum dot, or diffusive metallic) lead to the same conclusions: the quartet current-phase characteristics changes sign several times with voltage, owing to nonadiabatic transitions between Andreev levels, triggered by the voltage via the running phase $(\varphi_L - \varphi_R)(t)$ at frequency $4eV/\hbar$ [31, 37, 38, 42, 57]. This means that, in terms of the quartet current component, a TTJ can be either a "0−" or a "$\pi-$" junction, with respect to the quartet phase φ_Q. The same occurs with the phase-MAR current component that also changes sign but at different voltages. Superposition of quartet and phase-MAR components actually makes a generic TTJ a "θ-junction".

More generally, the characteristics of a TTJ (transparency, asymmetries between the three contacts, degree of decoherence) all conspire to shift or even suppress the sign changes. For instance, if the couplings to the biased terminals are much smaller than the one to the unbiased terminal, and the quartet TTJ current keeps a "$\pi-$" junction character at low and intermediate voltages [57]. Focusing on quartets only, in the case where backgates allow to separately control the transparency of the different contacts, one can reach a situation where, for a given voltage, the pair of TTJs of the SQUID can be both "0−" (or both "$\pi-$") junctions, or one being a "0−" and the other a "$\pi-$" junction. This strongly reminds the experiments performed with carbon nanotubes [61] (nanoSQUID) where the mechanism for "0−" to "$\pi-$" transition is instead the Coulomb interaction and the gate control of the nanotube junctions. In addition, "0−" to "$\pi-$" transitions have also been observed in superconductor-ferromagnet-superconductor Josephson junctions [62].

To illustrate the possibilities of such a quartet-SQUID, let us assume that TTJ1 is fully symmetric and resonant, with high quartet critical currents and several sign changes as V is increased from 0 to 2Δ. On the contrary, TTJ2 couples weakly but equally to the L, R terminals. This suppresses the MAR component in the SQUID current, and leaves us with a very asymmetric quartet-SQUID, with (neglecting the anharmonicity in this example):

$$I_{tot}(V) = I_{c1}(V)\sin(\varphi_{1Q}) + I_{c2}(V)\sin\left(\varphi_{1Q} - 2(\Phi^* + \phi^*/2)\right) \tag{15}$$

and $|I_{c1}(V)| >> |I_{c2}(V)|$. As said above, TTJ2 remains a "$\pi-$" junction so that $I_{c2} < 0$, while the sign of I_{c1} depends on V. Following the classical argument of an asymmetric SQUID, one first maximizes $I_{tot} \sim I_{c1}\sin(\varphi_{1Q})$ with respect to φ_{1Q}, which yields $\varphi_{1Q} \sim \pi/2$ if $I_{c1} > 0$ and $\varphi_{1Q} \sim 3\pi/2$ if $I_{c1} < 0$. Inserting this value into the – small – second term of Eq. (15) yields

$$I_{tot} \sim |I_{c1}| \pm I_{c2}\cos 2(\Phi^* + \phi^*/2), \tag{16}$$

with $\pm$ sign depending on the "0−" or "$\pi-$" character of TTJ1. First, this reconstructs the current-phase relation of TTJ2, including the sign of I_{c2}. Second, as V is swept upwards from 0, the sign changes of TTJ1 reflect themselves in $\pi-$ shifts in the flux dependence of I_{tot}.

As another example, Eq. (14) shows that phase-MARs can be investigated in TTJ1 only, provided TTJ2 is symmetric in (L, R) thus $\alpha_2 = 0$. The relative amplitude of phase-MARs and quartets in TTJ1 reflects directly in the shift α_1 of the total current vs flux dependence. In the generic case of non-symmetric TTJs, one instead measures the difference $\alpha_1 - \alpha_2$ of the two TTJ phase lapses.

An additional piece of information can also be gained by measuring the currents in the biased loop, i. e. the combination $I_L - I_R$ which, contrarily to I_{tot}, eliminates the quartet components at low transparency and is thus sensitive to MAR currents only.

Conclusion: We have proposed a quartet-SQUID generalizing the standard SQUID geometry to make quartet and phase-sensitive MAR current interfere under control of a magnetic flux. The periodicity in the flux dependence of the total critical current through the SQUID reflects the quartet charge $4e$. In addition, the distinguishing phase symmetries of both current components imply a phase lapse in the flux sensitivity of the critical current of the interferometer, which allows to quantify the phase-MARs with respect to the quartet current. Finally, phase-MARs are a consequence of both quartet emission and coherent subgap transport. Thus, they provide evidence against scenarii based on extrinsic synchronization via the outer circuit of junctions described by an adiabatic current-phase relation. A full quantitative analysis and comparison with future experiments requires to inject into the present description the expressions for quartet and MAR currents of each TTJ, obtained from microscopic theories.The principle of the present quartet-SQUID can obviously be generalized to higher order multipair transport in MTJs with four or more terminals.

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