Non-local Super-current of Quartets in a Three-Terminal Josephson Junction

One Sentence Summary:
Observation of a new coherent and nonlocal super-current, carried by four entangled electrons originating from two independent Cooper pairs.

Authors:
Yonatan Cohen†, Yuval Ronen†, Jung-Hyun Kang, Moty Heiblum#, Denis Feinberg2,3, Régis Mélin2,3, and Hadas Shtrikman

Affiliation:
1 Braun Center for Submicron Research, Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 76100, Israel
2 CNRS, Institut NEEL, F-38042 Grenoble, France
3 Université Grenoble-Alpes, Institut NEEL, F-38042 Grenoble, France

† Equal contributions
# Corresponding Author (moty.heiblum@weizmann.ac.il)

Abstract
We report observation of a new non-dissipative and nonlocal Josephson current - carried by quartets – each made of four entangled electrons originating from a pair of Cooper pairs. This supercurrent is a result of a new kind of Andreev bound states, formed between three biased superconducting terminals deposited on a semiconducting nanowire. Sensitive cross-correlation measurements of current fluctuations, accompanied by non-local conductance measurements, proves the non-local nature of the supercurrent and points to its origin. In addition, a rich sub-gap structure, resulting from non-local multiple Andreev reflections, was observed. An extensive and detailed theoretical study is intertwined with the experimental results.
Introduction

Superconductivity is one of modern physics’ triumphs, manifesting a macroscopic phenomenon governed by quantum mechanics (1) and stressing the significance of the ‘phase’ of a macroscopic wave function. Most striking is the DC Josephson effect (2): In response to a phase difference between two superconductors (SCs) connected by a ‘weak link’, a non-dissipative supercurrent flows through the junction (Fig. 1A). Moreover, biasing the junction makes the phase evolve with time, leading to an oscillatory current - the AC Josephson effect. Here, we demonstrate a new type of DC Josephson current, observed in a biased triple-superconductor-junction (as illustrated in Fig. 1B). This current is a consequence of binding pairs of Cooper pairs - forming the so-called quartets (3).

In an equal-potential three-terminal junction, an ordinary supercurrent of Cooper pairs can flow from one terminal to any other. In contrast, when the terminals are biased with different potentials, the usual two-terminal DC supercurrent vanishes; however, new types of supercurrent can emerge. The simplest one occurs when $V_L = -V_R$ (with $S_M$ grounded). In this case it is predicted that two Cooper pairs emerging simultaneously, one from $S_R$ and one from $S_L$, entangle themselves inside the junction to form a quartet four-electron state (3-6). It is also predicted that a reversed process occurs, where two Cooper pairs in $S_M$ are entangled in the junction thereby exiting simultaneously in $S_L$ and $S_R$, leading to a non-local supercurrent. Noting that while the entanglement may not exist beyond the coherence length of the superconductor, simultaneous bursts of charges can be looked for in order to point to the non-local nature of the process, similar to Cooper pair splitting experiments (7).

Here, we show results of non-local conductance measurements, accompanied by positive cross-correlation (CC) of current fluctuations at the two biased SCs ($S_R$ and $S_L$). In addition to the observed quartets, higher non-dissipative processes, leading also to supercurrents (such as sextets (4, 5); as well as dissipative quasiparticle channels due to local and non-local multiple Andreev reflections (MAR) (8-10), have been observed. In the past, a 3-terminal non-coherent metallic junction (6) showed also a conductance signature of the quartets; however, this work did not incorporate non-local or cross-correlation measurements which pinpoint the microscopic mechanism responsible for the quartet’s formation, leaving the quartet interpretation debatable.
Quartet super-current and quantum noise

Let us look at the microscopic picture of the supercurrent flow. In a short two-terminal Josephson junction the equilibrium supercurrent is carried by a resonant Andreev bound state (ABS, Fig 1C), and is given by

\[ I_c = -\frac{2e}{\hbar} \frac{dE}{d(\Delta \phi)} \]

with \( E \) the energy of the bound state and \( \Delta \phi \) the phase difference between the two superconductors (11). Extending to three terminals (12-14), the ABS energy \( E(\phi_q, \chi) \), is a function of two independent phases: \( \phi_q = \phi_L + \phi_R - 2\phi_M \) and \( \chi = \phi_L - \phi_R \) where \( \phi_L, \phi_R \) and \( \phi_M \) are the phases of the left, right and middle terminals. At the quartet’s biasing condition, \( V = V_R = -V_L \), the phase \( \phi_q \) stays stationary while the phase \( \chi \) is continuously changing, \( \chi = \frac{4e}{\hbar} Vt \). In a semi-classical picture, one may average the energy \( E(\phi_q, \chi) \) over time, yielding a \( \phi_q \)-dependent effective energy \( E_{\text{eff}}(\phi_q) \), with a supercurrent

\[ I_{c_{\text{quartet}}} = -\frac{2e}{\hbar} \frac{dE_{\text{eff}}}{d\phi_q} \]

This new kind of bound state which is formed, connecting all three phases, is illustrated in Fig. 1D (Supplementary Information, S1 A, B). Hence, in a similar fashion to the two-terminal Josephson supercurrent, the quartet supercurrent is governed by the stationary phase \( \phi_q \).

In general, depending on the voltage \( V \) and the temperature, quantum transitions between the occupied quartet ABS and its excited particle-hole partner may take place, like Landau-Zener (LZ) transitions (15) or as induced by microwave (16) (shown in Fig. 1E). Void of such transitions, the Josephson current is expected to be noiseless; however, the transitions introduce stochasticity in the occupation of the ABS (17, 18), thus leading to strong fluctuations in the quartet supercurrent (SI, S1 A&B). As in the case of ‘Cooper pair splitting’ (7, 19-23), it is expected that the resulting current fluctuations in the left and right terminals will be positively correlated (22). Modeling the non-equilibrium dynamics in a similar structure that has two quantum dots, at resonance, imbedded in the normal arms, finds: (24) (Supplementary Information, S1 B): (i) The cross-correlation (CC) of current fluctuations in the biased left and right terminals depends on the stationary quartet phase in a characteristic non-monotonic fashion. Its variation with the voltage \( V \) reflects the LZ transitions. (ii) The CC is positive at small \( V \).
where non-local MAR processes are suppressed, and exceeds the value expected from non-local MAR processes. Our device, described in Fig. 1F, emulates the theoretical model.

Experimental setup

Two configurations of the three-terminal Josephson junction were realized by coupling the superconducting contacts to an InAs nanowire: A single nanowire configuration (d1, Fig. 1F), and a Y-shape nanowire configuration (d2, Fig. 4A). The nanowires were grown by gold assisted MBE process, using the well-established vapor-liquid-solid growth technique. Growth was initiated on a non-patterned (100) InAs substrate, where both, single wires and merged (Y-shape) intersections were formed. Devices were fabricated on an oxidized P+ doped Si wafer (oxide: 150nm SiO$_2$), with superconducting contacts and local gates made by depositing 5nm/120nm Ti/Al. The measurement setup allowed measurements of the differential conductance and the ‘zero frequency’ CC of current fluctuations ($S_L$, $S_R$) (SI, S2). We define $G_{L/R} = \frac{dI_{L/R}}{dV_M}$, where $I_{L/R}$ is the current in $S_L$ or $S_R$, and $V_M$ is a small AC signal applied to the central contact. The DC bias to $S_L$ and $S_R$, for the CC measurements, was applied on a 5Ω resistor at the source (SI, S2). The induced superconducting energy gap in the nanowire was $2\Delta \approx 180\mu$eV.

Results and Discussion

Conductance measurements

Figures 2A & 2B present a color plot of $G_L$ and $G_R$ as function of the applied voltages $V_L$ and $V_R$ (in d1 configuration). A pronounced high conductance peak is observed for $V_L=-V_R$, agreeing with the expected quartet resonance. Trace cuts at $V_R = -16\mu$V show both $G_L$ and $G_R$ as a function of $V_L$ in Fig. 2C. The sharp peak at $V_L=+16\mu$V emphasizes the difference between the quartet conductance peak and those peaks that are due to dissipative MAR processes (much wider and smoother). Moreover, this conductance peak is accompanied by two dips at its sides, much like the ubiquitous ‘zero-bias-conductance-peak’ of the two-terminal Josephson junction (see Fig. 2C, and SI, S1 C) (24). Mapping the phase dynamics close to the quartet peak onto an ‘effective « quartet » RSJ model’ (SI, S1 B) allows accessing a typical ‘quartet energy’ extracted from the peak’s width, $E_q \approx \frac{\hbar C}{2e} \approx 2\mu$eV, leading to a critical quartet current of ~0.6nA. In addition, other
non-dissipative processes are manifested by conductance lines with different slopes (Figs. 2A & 2B) also crossing the origin. For example, the non-dissipative process corresponding to the conductance line at \( V_R = -2V_L \) (and \( V_L = -2V_R \)), involves two Cooper pairs coming from the left (right) and one coming from the right (left). This represents a six-electron entangled state (sextet resonance) (4,5).

Cross Correlation
Concentrating on the quartet conductance line (\( V_L = -V_R \)), the CC of current fluctuations in the two biased SCs are shown in Fig. 3A as well as \( G_L \) and \( G_R \). At \( V_L \sim 15\mu V \) a clear positive CC peak, coinciding with the quartet conductance peak, is seen as expected from the quartet tunneling process. Note that this excludes a MAR interpretation as discussed above. The small negative CC background is due to various MAR processes (20,25-27). The CC measurement was extended along the quartet resonance line (Figs. 3B & 3C – upper panel), where a non-monotonous behavior was observed; being consistent with LZ transitions (see SI, S1 B). Noting that the evolution of the CC signal with voltage depends on experimental details (not necessarily known), and thus it is not surprising that the data does not fully replicate the theoretical estimate (Fig. 3C - lower panel). Interestingly, the large CC signal agrees with the expected one, being on the order of \( \frac{e^2}{h} \Delta = 7 \times 10^{-28} A^2 / Hz \).

Quartet Non-Locality
Quite like in experiments of Cooper pair splitting (21), it is expected that the probability of forming a quartet in \( S_M \) by a Cooper pair arriving from the left depends on the probability of its partner arriving from the right. This means that suppressing \( G_R \) along the quartet line should suppress also \( G_L \) along this line. Pinching the right arm (with \( V_{GR} \)), the quartet conductance peaks disappeared simultaneously in left and right sides (Fig. 3D). Needless to say, the CC signal followed the behavior of the conductance (Fig. 3D).
Dissipative MAR - Non Locality

The MAR processes in a three-terminal Josephson junction consist of local and non-local MAR processes \((9)\). They can be categorized in two families: \((L1)\) \(mV_L+nV_R=2\Delta\), and \((L2)\) \(mV_L+nV_R=0\), where \(m\) and \(n\) are integers. If, in addition, the transport is coherent, interference between different MAR processes will further modify the expected MAR behavior, thus obtaining the so-called phase MAR processes \((6)\).

In order to observe non-local MAR we switched to the d2, Y-shape configuration, with an enhancement in the direct transmission from left to right (Fig. 4A). Plotting \(G_L\) as a function of \(V_L\) and \(V_R\) reveals a rich sub-gap structure, with certain lines belonging to the \(L1\) family (guide lines in Fig. 4B). Some of those lines correspond to the well-known local MAR process; such as the \((-1,0)\) line – being the MAR between \(S_M\) and \(S_L\), and the \((-1,1)\) line – being the MAR between \(S_L\) and \(S_R\). Other lines correspond to non-local MAR processes such as the \((-2,1)\) and \((-3,2)\) lines; involving crossed-AR (see Fig. 4C & 4D). Lines belonging to the \(L2\) family are not observed in the experiment and sextet lines are clearly seen for certain transmission configurations (see SI -S5). Identifying the origin of a differential conductance line is straightforward. First, as explained above (Fig. 2C for d1, and SI - S5 for d2), by comparison with the supercurrent conductance peaks (SI, S1 - C). Second, a non-local MAR in the bias range of interest should have been a \(10^{th}\) order process, which is unlikely (due to dephasing and overlap of adjacent MAR peaks).

Summary

Here, we present a new phenomenon, which takes place in a multi-terminal Josephson junction. Due to the phase coherence of the junction, new quantum states form, as Andreev bound resonances carrying a non-dissipative DC quartet (and higher order) current. Using sensitive, cross-correlation measurements of current fluctuations, combined with non-local conductance measurements, we reveal the non-locality of the effect and distinguish it from the ubiquitous multiple Andreev reflections. Moreover, the observed large cross-correlation of current fluctuations when the quartet's condition was fulfilled is theoretically justified, in its sign and magnitude, by resonant Landau-Zener transitions between the ground and excited Andreev bound states. This is therefore the first clear-cut experimental signature of the coherent resonant
quartet state, being a non-local, phase-dependent, quartet supercurrent. Finally, we also observed a very rich subharmonic structure in the conductance attributed to non-local multiple Andreev reflection processes.
References and Notes:


12. B. van Heck, S. Mi, A. R. Akhmerov, Single fermion manipulation via 
superconducting phase differences in multiterminal Josephson junctions, Phys. Rev. B 
90, 155450 (2014).

R.-P. Riwar, M. Houzet, J. S. Meyer, Yu. V. Nazarov, Multi-terminal Josephson 
junctions as topological materials, Nature Comm. 7, 11167 (2016); E. Strambini, S. 

Nazarov, Closing the proximity gap in a metallic Josephson junction between three 

Urbina, Supercurrent in atomic point contacts and Andreev states, Phys. Rev. Lett. 85, 
170 (2000).

15. F. S. Bergeret, P. Virtanen, T. T. Heikkilä, J. C. Cuevas, Theory of microwave-assisted 

16. A. Martín-Rodero, A. Levy Yeyati, F. J. García-Vidal, Thermal noise in 

76, 3814 (1996).

18. J. C. Cuevas, A. Martín-Rodero, A Levy Yeyati, Shot noise and coherent multiple 
charge transfer in superconducting quantum point contacts, Phys. Rev. Lett 82, 4086 
(1999).

19. R. Cron, M. F. Goffman, D. Esteve, C. Urbina, Multiple-charge-quanta shot noise in 

20. L. Hofstetter, S. Csonka, J. Nygård, C. Schönberger, Cooper pair splitter realized in a 


Acknowledgement:
D. F. and R. M. acknowledge support from ANR Nanoquartets 12-BS-10-007-04 and the CRIANN computing centre. M.H. acknowledges the partial support of the Israeli Science Foundation (ISF), the Minerva foundation, the U.S.-Israel Bi-National Science Foundation (BSF), the European Research Council under the European Community’s Seventh Framework Program (FP7/2007-2013)/ERC Grant agreement No. 339070, and the German Israeli Project Cooperation (DIP). H.S. acknowledges partial support by ISF grant number 532/12, and IMOST grants #0321-4801 & #3-8668.
Fig. 1. Non-dissipative current in two- and three-terminal Josephson junctions.

A) Schematic illustration of a two-terminal Josephson junction. B) Schematic illustration of the formation of a quartet by entangling two separate Cooper pairs. C) Schematic illustration of the two terminals resonance process of an Andreev bound state, enabling Josephson super-current flow. D) Schematic illustration of the three terminal quartet ABS, leading to non-local super-current. E) The dependence of the ABS on the complementary phase $\chi = \varphi_L - \varphi_R$. Evolution of the phase in time leads to Landau-Zener transitions. F) SEM image of device d1. Gates (yellow) were used to form barriers or quantum dots in the bare part of the nanowire.
Fig. 2. Differential conductance of the left\right contact in the $V_L$-$V_R$ plane.  
A) $G_L$ as function of $V_L$ and $V_R$. The solid line is a guide for Fig. 3A (upper panel). The dashed 
square is a guide for Fig. 3B. The c-quartet line indicates quartets emitted from $S_L$ towards $S_R$ 
and $S_M$ (S4) instead of $S_M$ to $S_L$ and $S_R$. B) $G_R$ as a function of $V_L$ and $V_R$. C) $G_L$ (blue) and $G_R$ 
(red) as a function of $V_L$. The shape of the quartet peak is shown in the upper right corner with 
the quartet energy, $E_q$, indicated.
Fig. 3. Cross-correlation of current fluctuations and non-local conductance measurement.  
A) Upper panel: Differential conductance cuts of $G_L$ and $G_R$ along the solid line in figures 2A & 2B. Lower panel: Cross-correlation (CC) of current fluctuations at the left and right terminals. B) CC as a function of $V_L$ and $V_R$ in the region defined by the dashed square of figure 2A, 2B & 2C. C) Upper panel: CC along the quartet line. Lower panel: Theoretical calculation of the CC. The maximum is due to Landau-Zener resonances. Inset: zoom-out in the bias voltage range. It should be noted that the measured CC in the experiments also drops after 20µV. D) Upper & middle panel: $G_L$ and $G_R$, respectively, as function of the left contact bias, $V_L$, and the right gate voltage, $V_{GR}$. Lower panel: The CC as a function of $V_{GR}$. 

Fig. 3
Figure 4. Local and non-local Multiple Andreev Reflections (MAR) in a three terminal Y-shaped Josephson junction. A) An SEM image of device d2. B) $G_L$ as a function of $V_L$ and $V_R$. The lines labeled (-1,0) and (-1,1) correspond to first order local MAR. The lines labeled (-2,1) and (-3,2) correspond to non-local MAR.
and (-3,2) are second and third order \textit{non-local} MAR processes. C) Schematic illustration of the (-2,1) process – second order (single Andreev reflection). D) Schematic illustration of the (-3,2) process – third order (two Andreev reflections).
Non-local Super-current of Quartets in a Three-Terminal Josephson Junction

Authors:
Yonatan Cohen††, Yuval Ronen††, Jung-Hyun Kang†, Moty Heiblum‡, Moty Heiblum‡, Denis Feinberg², Régis Mélin², and Hadas Shtrikman†

Affiliation:
† Braun Center for Submicron Research, Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 76100, Israel
‡ CNRS, Institut NEEL, F-38042 Grenoble, France
³ Université Grenoble-Alpes, Institut NEEL, F-38042 Grenoble, France

†† Equal contributions
‡‡ Corresponding Author (moty.heiblum@weizmann.ac.il)

Methods and Supplementary Information:
In this Supplementary Section we add details to the main text. We include a brief review of the theoretical background as well as the simulation method and results, as well as more information on the conductance and noise measurements.
A. General

Let us consider a normal region connected to three superconducting terminals. When terminals $S_{L,R}$ are biased respectively at voltages $V_{L,R}$ with respect to terminal $S_M$, a coherent stationary motion of Cooper pairs occurs when $nV_L + mV_R = 0$, where $(n,m)$ are integers. This involves $n$ pairs crossing from $S_M$ to $S_L$ and $m$ pairs crossing from $S_M$ to $S_R$ in a single quantum process $(1, 2)$. This multi-pair process unveils a phase combination $\varphi_{n,m} = n\varphi_L + m\varphi_R - (n + m)\varphi_M$ which, owing to the Josephson relation, $\frac{d\varphi_i}{dt} = \frac{2eV_i}{\hbar}$; $(i=L,R,M)$, is a constant of motion. The main anomaly reported in the experiment along the line $V_L + V_R = 0$ corresponds to a quartet (a pair of pairs) crossing from $S_M$ towards $S_{L,R}$, revealing the stationary phase $\varphi_q = \varphi_{1,1} = \varphi_L + \varphi_R - 2\varphi_M$. Sextet lines are also visible, though fainter, where, $(n,m) = (1,2)$ or $(2,1)$. These DC modes manifest static phase coherence despite the non-equilibrium conditions. Due to energy conservation, multi-pair processes are non-dissipative, contrary to the usual quasiparticle multiple Andreev reflections (MAR). Along the line $V_L = -V_R = V$, theory predicts that the quartet current $I_q(\varphi_q, V)$ is odd in phase and even in voltage. $I_q$ is similar to a DC Josephson supercurrent but it depends on $V$ as a new control parameter. It involves equal and perfectly correlated currents flowing through $S_L$ and $S_R$.

Choosing $\varphi_q = \varphi_L + \varphi_R - 2\varphi_M$ and $\chi = \varphi_L - \varphi_R$, as canonical variables one may as a first step begin with the Andreev bound state (ABS) energies at equilibrium $E_{ABS}(\varphi_q, \chi)$ which can be computed in a suitable model. Subsequently one can use a semiclassical approximation and average out the drifting phase $\chi(t)$. This can be formally done by expanding $E_{ABS}(\varphi_q, \chi)$ in Fourier series in both variables keeping only the zeroth component in $\chi(t)$. This leads to an effective energy $E_{eff}(\varphi_q)$, which is a function of $\varphi_q$ only. Then the average quartet current is found to be $I_{quartet}^{SC} = -\frac{2e}{\hbar} \frac{dE_{eff}}{d\varphi_q}$. This rough procedure reduces a set of two-dimensional ABS, valid at equilibrium, to a set of one-dimensional effective ABS. Yet, it neglects the quantum nature of the non-equilibrium processes, which take place as multiple Andreev reflections at the
junction interface of all three superconductors. In the limit where the Josephson junction frequency \( \omega_0 = \frac{2eV}{\hbar} \) is much smaller than the separation between the effective ABS, one obtains Landau-Zener transitions between the latter. Non-equilibrium Green’s function calculations confirm this picture (see below and Figure S1a) and demonstrate that those transitions indeed induce a strong quartet noise.

B. Results from non-equilibrium Green’s function theory

The picture above is semi-phenomenological and a full non-equilibrium theory of transport is necessary. Such a theory is indeed available along the line \( V_L = -V_R = V \); it involves the calculation of the Keldysh Green’s function matrix \( G(E, n) \), where \( E \) is the energy and \( n \) the index of the harmonics of the Josephson frequency \( \omega_0 = \frac{2eV}{\hbar} \). Voltages down to 0.1\( \Delta \) can be reached with about 100 harmonics. Mapping the full \((V_L, V_R)\) plane is out of reach, as independent Josephson frequencies \( \omega_L, \omega_R \) would require much too large matrices. Results concerning a single dot model are found in Ref. (3). The model used to describe the present experiment, instead, involves two single-level quantum dots \( D_L \) and \( D_R \) with energy levels \( \varepsilon_L \) and \( \varepsilon_R \) coupled to the terminals by couplings (broadening in the normal state) \( \Gamma_L, \Gamma_M \) (for dot \( D_L \)), and \( \Gamma_R, \Gamma_M \) (for dot \( D_R \)). For the purpose of interpreting the experiment, \( \varepsilon_L \) and \( \varepsilon_R \) are taken to be zero (resonant dots). Interactions are neglected owing to the large transparency. Figure S1a shows the quartet current flowing in terminal M and the cross-correlation noise \( S_{LR} \). The \( \Gamma \)’s are taken as \( \Gamma_L = \Gamma_R = 1.5\Delta \) and a smaller \( \Gamma_M = 0.3\Delta \), owing to the finite width of the central superconducting finger that limits the crossed Andreev reflection.

Panels S1a, b show the quartet current and the crossed noise as a function of the quartet phase, fixing \( eV = 0.15\Delta \) and taking into account a very small inelastic broadening \( \eta = 10^{-6} \) in the superconductors. A very strong resonance appears as marked dips at specific values of \( \varphi_q \), that can be interpreted as resonant Landau-Zener transitions between two symmetrical ABS formed at zero voltage, triggered by the Josephson frequency \( \omega_0 \). This indeed resembles the effect of microwave irradiation on a quantum point contact (4). Spectacularly, the cross correlation noise exhibits sharp peaks at the same phase values as the current dips (Fig S1a, Panel B). These peaks
can be very high, signaling “trains” of quartets, in a way similar to the thermal noise due to transitions between a single junction ABS (5, 6). Fig S1a, Panel C & D shows a broadening and an amplitude decrease in the current and the noise anomalies when increasing the inelastic parameter, where $\eta = 10^{-3}$. Panel S1a, E shows the variation with $V$ of the value of the cross correlation noise, calculated along the line $(V, -V)$ by taking into account thermal fluctuations (see Section C). First, one finds that the noise is positive (it becomes negative at larger $V$ values). Second, its behavior is not monotonous, the first maximum being indeed due to the above Landau-Zener resonance. The maximum noise is much larger than $\frac{e^2 \Delta}{\hbar}$, indicating large bursts of quartets emitted within the Landau-Zener resonances. Those trends are also found in the experiment, where a non-monotinous variation of the maximum noise is obtained as well (Figure 3c, main text). No quantitative fit is attempted here, because the details of the current and noise variations with phase and voltage are very sensitive to the location of the resonances. In particular, i) the non-monotinous variation with $V$, with huge oscillations, and ii) the anharmonic phase variation, with dips reflecting Landau-Zener transitions, are characteristic of such resonances and point towards the phase coherence of the quartet dynamics. Here, the parameters of the model are chosen to illustrate the main trends in a somewhat dramatic case. We also emphasize the extreme sensitivity of the quartet noise to the inelastic time, a parameter unknown in the experiment. As a last remark, measuring the charge $4e$ of quartets would require low transparency, making the detection much more difficult.

C. Phase diffusion model close to the quartet line

Here we present a semi-phenomenological picture which is capable of describing transport in the vicinity of the quartet line $(V, -V)$, where no full microscopic solution is available anymore. In a voltage-biased junction, the Josephson supercurrent is probed indirectly through the shape of the conductance anomaly manifesting a rounded Josephson plateau in the $V(I)$ characteristics. Its double-well shape can be described by an overdamped RSJ model (7). The same is true here for the conductance anomaly, as a function of two voltages $V_L, V_R$. Transport by a quartet supercurrent is witnessed by a rounded plateau, centered on the quartet line. One can proceed and adiabatically describe the dynamics close to this line in the same spirit as the overdamped Josephson junction close to $V=0$, by means of an effective « quartet » RSJ model. This involves
two branches in parallel: a quartet branch, non-dissipative and dependent on the phase \( \varphi_q \), and a resistive branch. Setting, \( V_L + V_R = v \ll |V_L|, \quad |V_R| \approx V \), the phase \( \varphi_q = \varphi_{q0} + \frac{2e}{\hbar} vt \) is a slow variable, while \( \varphi_L - \varphi_R = \frac{4e}{\hbar} Vt \) is a fast one. The phase \( \varphi_q \) evolves in an effective potential, which is determined here from the non-equilibrium Green’s function calculation, by integrating the calculated quartet current \( I_q(f_q) = \frac{2e}{\hbar} \frac{dU_{\text{eff}}}{d\varphi_q} \). Notice that this self-consistent procedure goes beyond the time-averaging procedure explained in section B1: One uses the microscopically exact solution on the quartet line to extrapolate to the slow adiabatic motion in its vicinity.

For this purpose, one can apply the theory of phase diffusion in the « washboard » potential formed by the quartet phase potential \( U_{\text{eff}}(\varphi_q) = -\frac{\hbar I_q}{2e} \), where \( I_q \) is the average quartet current. Application of the Ambegaokar-Halperin phase diffusion model (5) yields the phase thermal probability distribution \( p(\varphi_q) \) as well as the quartet current-voltage \( I_q(v) \) characteristics. The conductance calculated from this scheme has the classical shape found in the usual Josephson effect and also in the present experiment on the quartet line (Fig S1b). It only depends on a single parameter \( \gamma = \frac{\hbar I_{\text{qc}}}{2e k_B T} \), that can here be estimated from the universal shape of the anomaly to be about 1-2 at 30mK.

This argument confirms that the conductance anomaly across the quartet line underlies the quartet phase, and allows to evaluate a typical quartet energy to be about 60-100mK. This model also allows calculating the thermally averaged value of the crossed noise at the center of the anomaly \( (v = 0) \). It is plotted in Figure S1a Panel e and can be much larger than \( \frac{e^2 \Delta}{h} \). Yet, this model does not allow to fully calculate the crossed correlation noise anomaly across the quartet \( (V, -V) \) line, owing to the strong non-adiabatic character of the quartet noise which dramatically depends on Landau-Zener transitions. Those transitions are not correctly described by the model described above.
D. Nonlocal multiple Andreev reflections vs quartets.

The only phenomenon that might compete with the quartet mechanism along the line \((V, -V)\) is the zero-energy nonlocal MAR. First, the order of magnitude of the noise obtained from such MAR’s is not larger than \(\frac{e^2\Delta}{h}\), as in two-terminal junctions. It could reach such value only if the MAR stays coherent and elastic down to voltages as low as \(0.16\mu eV\), which would correspond to MAR of order \(N>10\), and more non-local MAR are involved. In the region of the \(V_L-V_R\) plane, where the noise anomaly is measured, MARs are instead not present and a careful investigation shows that the vertical lines featured in Figure 3b (main text) at low voltages are not due to MAR, and that MARs in this sample are limited to order \(N=3-4\). This in turn strongly limits the size of the cross correlation noise which should be much smaller than \(\frac{e^2\Delta}{h}\). This is what is seen in the experiment where the marked quartet noise anomaly emerges from a weakly negative MAR background.

S2 - Measurement Setup
The experimental setup is shown in Fig. S2. Resonance frequencies, of the LC circuits, were matched in order to enable the cross-correlation measurements and were \(\sim 705\text{KHz}\).

Differential conductance measurements:
As described in the main text, differential conductance was measured by applying an input ac signal of \(0.8\mu\text{Vrms}\) at 705 KHz to the middle contact, \(S_M\), while measuring the differential voltages, \(V_L\) and \(V_R\), on the left and right contacts, \(S_L\) and \(S_R\), respectively. The \(500\Omega\) load resistors were chosen to be significantly lower than the typical values of the sample resistance so that they serve as effective drains pulling most of the current to the ground. We then define

\[
G_L = \frac{dI_L}{dV_M} \\
G_R = \frac{dI_R}{dV_M}
\]

Where \(I_L=V_L/500\Omega\) and \(I_R=V_R/500\Omega\).
Cross correlation of current fluctuations measurements:

In the cross correlation of current fluctuations measurement no ac signal is applied. DC bias voltages, however, produce current fluctuations, \(dI_L\) and \(dI_R\) (ac component at relative low frequencies \(\sim 705\text{kHz}\)). We are interested in the cross correlation of the current fluctuations \(<dI_LdI_R>\). The current fluctuations introduce voltage fluctuations \(dV_L=dI_L \times 500\Omega\) and \(dV_R=dI_R \times 500\Omega\) at the inputs of a home-made, cold (1K) amplifier (the gains of which were measured in advance to be \(g_L=6.12\) and \(g_R=5.77\)). Another amplification stage was used at the output of the dilution fridge using NF amplifiers each with a gain of 200. Both signals are multiplied and amplified by a home-made cross correlator with a central frequency of 730KHz, resolution band width of \(RBW=100\text{KHz}\) and gain of \(g_{CC}^2=10^7\). Finally, the cross correlator signal undergoes an RC filter. The CC can be estimated by:

\[
CC_{\text{tot}} = \left\langle (dI_L \times 500 \times g_L \times g_{NF} \times g_{CC}) \times (dI_R \times 500 \times g_R \times g_{NF} \times g_{CC}) \right\rangle \times RBW
\]
\[
= \langle dI_L dI_R \rangle \times [500^2 \times g_L \times g_R \times g_{NF}^2 \times g_{CC}^2 \times RBW] \]
\[
= \langle dI_L dI_R \rangle \times \alpha
\]

However, parasitic effects such as RF picked up by both output lines, cross talk coming from capacitance between the output lines etc., add an independent “background” cross correlation,

\[
CC_{\text{tot}} = \langle dI_L dI_R \rangle \times \alpha + CC_0
\]

Since the load resistor was chosen to be very small (500Ω) relative to the sample resistance, the voltage signal is very small, relative to the background cross correlation. Hence, the background must be calibrated and subtracted as explained in the next section.

S3 - Cross correlation calibration

To calibrate the background, before each measurement of \(V_L\) where we scan the cross correlation (as we move through the quartet line), we perform the same measurement at a high magnetic field of \(B=200\text{mT}\) (above the critical field of the SCs so that all contacts are in the normal state). At zero bias voltages, no current flows through the device and we expect the voltage fluctuations \(dV_L\) and \(dV_R\) to be uncorrelated. Hence, we take the cross correlation measured at this high
magnetic field and at zero bias as our background cross correlation. An example of such cross correlation measurement is shown in Fig. S3

**S4 – Negative Cross correlation on the complementary quartet lines**

As mentioned in the main text we expect to observe a positive cross correlation of the current fluctuations, between the left and right terminals, along the quartet conductance line. As a sanity check we measured the CC along different processes where we expect to get negative cross correlation.

In Fig S4A we illustrated a quartet process which is named complementary quartet process, which is merely a permutation of the terminals from the process described in the main text. In this process Cooper pairs from the left and middle contact enter the right contact and in the process they are entangled between themselves. This process is thus called a complementary quartet process. In Fig S4B we sketched the complementary quartet ABS which is the mechanism for the creation of a quartet in the right contact.

The results of this process are shown in Fig S4C. We placed the right terminal at a bias of $V_R = -13(\mu V)$ and measured the differential conductance in the left and right contact (upper and middle panels) and the cross correlation of the current fluctuation in the left and right terminals.

Concentrating on the blue shaded region (where the complementary quartet process occurs) a clear reduction in the cross correlation is observed – originating from a negative contribution of the process. Concentrating now on the red shaded area, which is the region where a trivial supercurrent flows from the left to the right terminal; once again a clear reduction of the cross correlation is observed.

**S5 – MAR and quartet in d2 (T-shaped nanowire device)**

Fig. S5 shows conductance measurements on device d2 as a function of $V_L$ and $V_R$. Both MAR lines as well as supercurrent lines can be seen. The figure emphasizes the similarity of the quartet line ($V_L=-V_R$) to the other supercurrent lines ($V_L=0$, $V_L=V_R$) and the difference from MAR lines (e.g. $V_L=-2\Delta$).
**Figure Captions**

**Fig. S1a.** Two-dot model.

NEFG calculation of the quartet current $I_0$ (in the central terminal M) and of the current cross correlation (CC) $S_{LR}$, as a function of the quartet phase $\varphi_q$ (panels A-D) and of the voltage (panel E), with $eV = 0.15\Delta$, the inelastic parameter $\eta_s = 10^{-6}\Delta$ (panels A, B) and $\eta_s = 10^{-3}\Delta$ (panels C, D, E). Panel A shows two resonances corresponding to Landau Zener transitions between the adiabatic quartet states. At the same phase values, the CC displays sharp maxima with very high values. Both trends are still present with a larger inelastic parameter (panels C, D) but broader and reduced amplitude. Panel E shows the non-monotonous variation of the CC at the center of the quartet line but sweeping the voltage $V$, showing that the resonances occur at
certain V values. Thermal fluctuations are taken into account with gamma = 0.5 (see text).

Fig. S1b. Differential conductance of the quartet

Differential conductance as a function of voltage v, taken from the center of the quartet line, in units of the maximum quartet current, the normal state conductance being taken to one. It is calculated from the effective phase diffusion model, plugging in it the results of the Green’s function calculation for IQ(φ_Q)
Fig. S2. Devices and Measurement Setup.

A) D1 configuration. Three superconducting contacts are placed on a single nanowire. The central contact is made much narrower than the coherence length of the superconductor so that crossed Andreev reflection is allowed. B) D2 configuration. Three superconducting contacts are placed on the three branches of a T-shaped nanowire. Compared to the d1 configuration, here there is much higher probability for a direct transport of quasiparticles and Cooper pairs from the left contact to the right contact. C) Our measurement setup. The differential conductance and the cross correlation of current fluctuations were measured using this setup as described above. The device configuration is schematically illustrated at the top right hand corner.
Fig. S3. Cross correlation calibration.

Cross correlation as a function of $V_L$, measured at $B=200\text{mT}$, well above the critical magnetic field of aluminum. At zero bias, there is no current and therefore we expect zero cross correlation. It can be seen that even at a finite bias of 50microV the cross correlation is essentially the same as at zero bias (within an uncertainty that is significantly smaller than the cross correlation measured at $B=0$ on the quartet resonance). This is due to the fact that in configuration d1, the central contact disconnects the two sides of the device, or in other words most of the current from the left/right contact flows to the central contact and not to the other side.
Fig. S4. Complementary quartet lines.

A) Schematic illustration of the ‘right’ (right contact) complementary quartet line. B) Schematic illustration of the 3-terminal complementary quartet ABS. C) The upper and middle panels show the differential conductance in the left and right terminals. The lower panel displays the cross correlation of the current fluctuation, showing a negative peak in the location of the complementary quartet.
Fig. S5. MAR and quartet.
Conductance measurements of d2 as a function of $V_L$ and $V_R$. Both the MAR lines as well as the supercurrent lines can be seen. The figure emphasizes the similarity of the quartet line ($V_L=-V_R$) to the other supercurrent lines ($V_L=0$, $V_L=V_R$) and the clear difference from MAR lines (e.g. $V_L=-2\Delta$).