Positive noise cross-correlation in hybrid superconducting and normal-metal three-terminal devices

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Non-local entanglement is a key ingredient to quantum information processing. For photons, entanglement has been demonstrated¹, but it is more difficult to observe for electrons. One approach is to use a superconductor, where electrons form spin-entangled Cooper pairs, which is a natural source for entangled electrons. For a three-terminal device consisting of a superconductor sandwiched between two normal metals, it has been predicted that Cooper pairs can split into spin-entangled electrons flowing in the two spatially separated normal metals^{2–5}, resulting in a negative non-local resistance and a positive current–current correlation^{6,7}. The former prosperity has been observed^{8,9}, but not the latter. Here we show that both characteristics can be observed, consistent with Cooper-pair splitting. Moreover, the splitting efficiency can be tuned by independently controlling the energy of the electrons passing the two superconductor/normal-metal interfaces, which may lead to better understanding and control of non-local entanglement.

Entanglement of electrons may arise in the spatial degree of freedom (orbital entanglement) or the spin degree of freedom (spin entanglement). Recently, orbital entanglement in a fermionic Hanbury Brown and Twiss two-particle interferometer was observed using current cross-correlation measurements^{10,11}, but further investigation is still required to verify the entangled states¹². Spin-entanglement has been predicted to exist at the superconductor/normal-metal (SN) interface^{2,3} and can be understood in the context of Andreev reflection¹³, in which a low-energy electron in the normal metal impinges on the SN interface and a hole is retroreflected whereas a Cooper pair is created in the superconductor.

When two normal metals are coupled to a superconductor with spatial separation comparable to the superconducting coherence length (ξ_S), roughly the size of a Cooper pair, it is predicted that electrons in the two normal metals can also be coupled by means of a non-local analogue of Andreev reflection called crossed Andreev reflection^{6,7} (CAR). As a Cooper pair splits into two coupled electrons with opposite spin orientation that are then injected into the two normal-metal leads, instantaneous currents of the same sign are generated across the two SN interfaces, giving rise to a negative non-local resistance as well as a positive current–current correlation between the SN junctions. Previous experimental attempts focused on non-local resistance measurements^{8,9,14,15}, but the observation of CAR is complicated by another non-local process called elastic cotunnelling, in which electrons in the normal-metal leads tunnel across the superconductor with the help of Cooper pairs, resulting in a positive non-local resistance and a negative current–current

correlation. The electron keeps its spin orientation during the tunnelling. In terms of the conductance, theoretical studies have found that these two non-local processes tend to cancel each other exactly in the lowest order approximation in the tunnelling limit¹⁶, but experimental results and further theoretical investigation show that exact cancellation does not occur if the normal-metal leads are ferromagnetic⁸, if the NS interfaces are highly transparent^{14,17} or if there is strong Coulomb interaction^{9,18}. For tunnelling junctions it was shown that Coulomb interaction may lead to a transition from elastic cotunnelling to CAR with increasing voltage bias^{9,18}.

The noise cross-correlation measurement is an especially powerful tool to probe the correlation between charge carriers in mesoscopic systems¹⁹; for example, recently it has been used for probing orbital entanglement of electrons from independent sources¹¹. For a three-terminal NSN device, it was also predicted that current–current correlation can be used to directly probe CAR and elastic cotunnelling without the drawbacks of the resistance measurement^{20,21}. However, despite a considerable amount of theoretical work, no experimental observation has been reported. Below we describe both resistance and noise cross-correlation measurements, and in each type of measurement a tunable CAR is observed. It is worth noting that observation of a positive correlation alone is not proof of CAR and Cooper pair splitting^{22,23}, but the observation of both a negative non-local resistance and a positive cross-correlation for our devices provides compelling evidence of CAR, which involves spin-entanglement according to current theoretical understanding.

In Fig. 1 we show a scanning electron micrograph of a hybrid device, as well as a schematic of the measurement circuit (see the Methods section for sample fabrication and measurements). Two normal-metal/insulator/superconductor (NIS) junctions are formed between Al and Cu leads, with room-temperature resistances in the range of 10–20 k Ω . The advantage of using tunnelling junctions instead of junctions with highly transparent interfaces is the following. First, the large tunnelling resistance ensures that there is little leakage current from one junction to the other; that is, currents from different sides flow only to the superconducting lead (ground), so the bias across the two junctions can be varied independently. Second, for a highly transparent interface, elastic cotunnelling is expected to dominate over CAR at all temperatures and at energies below the gap voltage^{14,17}, whereas for tunnelling junctions CAR can dominate elastic cotunnelling at high bias^{9,15,18}. Third, the Coulomb interactions between the electrons in the normal metals may lead to a ‘dynamical’ Coulomb blockade effect (DCB), which prevents both electrons from being injected into the same normal-metal lead and thus facilitates the separation

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Figure 1 | Schematic of the cross-correlation measurement. Scanning electron micrograph with false colour enhancement of a three-terminal SN device. The scale bar represents 1 μm . The area coloured in dark purple is Al film and the area coloured in gold is Cu film. The thin white stripes are due to Al film deposited on the sidewall of the lithography mask. The two Al/AIO_x/Cu junctions are each biased with a d.c. current source (I_A and I_B) and the a.c. voltage fluctuations across each junction (V_A and V_B) are measured simultaneously. The right inset shows a schematic of the CAR process.

Figure 2 | Junction characterization. **a**, Current–voltage characteristics of a NIS junction at bath temperatures: 0.1 K (cyan), 0.2 K (red), 0.3 K (green) and 0.4 K (blue). Inset: bias dependence of the differential resistance measured at 0.4 K (note the log scale of the y axis). The solid lines are fits using the semiconductor model, with a slightly elevated electron temperature. **b**, Bias dependence of the voltage noise power of a NIS junction at 4.2 K (red) and 0.4 K (blue). Inset: zoomed-in regime for the 0.4 K data near zero bias. The solid lines are fits to the shot noise expression (Fano factor equals 1) with background noise from the measurement set-up also taken into account. The diverging behaviour near zero bias results from the large differential resistance as shown in **a**.

of the two spin-entangled electrons³. Note that the DCB in the superconducting lead may favour elastic cotunnelling instead of CAR because the number of electrons in the superconductor changes by two for CAR (ref. 18).

Our NIS junctions have no pin holes in the thin insulating layer, as indicated by good fitting of the current–voltage characteristics at different temperatures in Fig. 2a using the simple semiconductor model (see Supplementary Notes 1 for the details of the fitting). When there is no pin hole, current flowing across the NIS junction consists of well-separated events of quantum mechanical tunnelling of electrons (no temporal correlation and inelastic scattering). In this case, the power of the current fluctuation is proportional to the magnitude of the current. This so-called shot noise reflects the particle nature of the charge carriers¹⁹. Here the voltage noise power S_V is measured and it is related to the current noise power S_I by $S_V = S_I \cdot (dV/dI)^2$, with

$$S_I = \frac{4(1-F)k_B T}{R} + F \cdot 2eI \coth\left(\frac{eV}{2k_B T}\right) \quad (1)$$

where dV/dI is the differential resistance at bias voltage V , R is the resistance, k_B is the Boltzmann constant, T is the temperature, e is the electron charge, and F is the Fano factor, which equals 1 for full shot noise^{19,24}. At $eV \gg k_B T$, the hyperbolic cotangent

term approaches 1 and S_I is proportional to I , the hallmark of shot noise, as shown in Fig. 2b. The fitted value $F = 1$ again indicates that there are no pin holes. Below the superconducting transition temperature, Andreev reflection may lead to doubled shot noise near zero bias, as has been observed for short diffusive NS junctions with transparent interfaces²⁵. However, for typical NIS junctions such as ours, the probability of Andreev reflection is very small^{26,27}; thus, the subgap current resulting from Andreev reflection is also small. As the shot noise is proportional to the current, it is difficult to verify the doubled shot noise for NIS junctions²⁸. As for CAR, although in principle its presence can also be inferred from shot noise (autocorrelation) measurements²⁰, it is even harder because CAR is expected to be dominated by the Andreev reflection process^{14,20}. Another practical issue for autocorrelation measurements is that the extrinsic current noise from the measurement set-up (back action) can be much larger than the intrinsic shot noise.

Fortunately, with the non-local configuration shown in Fig. 1, the cross-correlation between V_A and V_B is only determined by non-local processes, and it is much less affected by any extrinsic current noise, as there is no circuit segment shared between the two channels except a short piece of superconducting Al wire, along which no voltage drop is expected to occur. In addition, the large differential resistance near zero bias (see Fig. 2) leads

Figure 3 | Bias current dependence of voltage noise power by the cross-correlation measurement. a–f, The measured voltage noise power at three different bath temperatures: 0.4 K (**a**), 0.3 K (**c**), 0.25 K (**e**) and the respective theoretical prediction at each temperature (**b,d,f**), with the assumption $A^{\text{CAR}} = A^{\text{EC}}$ in equation (2).

to a large non-local voltage–voltage correlation signal, which greatly helps the observation of the small non-local current–current correlation. However, the large differential resistance here also limits the bandwidth used for the cross-correlation measurement because of the capacitance of the electrical wiring (see the Methods section), which prevents us from making a reliable measurement below about 0.2 K.

Figure 3 shows the voltage noise power measured by cross-correlation, S_V , for our device at three different temperatures: 0.4 K, 0.3 K and 0.25 K. For NIS junctions in the absence of the Coulomb interaction in the leads, it was shown that when $e|V|$, $k_B T \ll \Delta$, where Δ is the superconducting gap, the current–current correlation between junction A and junction B is²⁰

$$S_{AB} = S^{\text{CAR}} - S^{\text{EC}} = 2eG_Q \left[(V_A + V_B) \coth \left(\frac{eV_A + eV_B}{2k_B T} \right) A^{\text{CAR}} - (V_A - V_B) \coth \left(\frac{eV_A - eV_B}{2k_B T} \right) A^{\text{EC}} \right] \quad (2)$$

where $G_Q = 2e^2/h$; $A^{\text{CAR}}(A^{\text{EC}})$ is the amplitude of CAR(elastic cotunnelling), determined by the distance between the two SN junctions and the properties of the interfaces. For simple tunnelling

junctions, $A^{\text{CAR}} = A^{\text{EC}}$ is predicted¹⁶. We note that S^{CAR} and S^{EC} have a similar form to the shot noise in equation (1). As the precise value of A^{CAR} is not known, a normalized S_{AB} is plotted in Fig. 3 to compare with experimental results. It is easy to see that by setting $V_A = V_B$, the elastic cotunnelling term in the brackets of equation (2) reduces to a bias-independent term $2k_B T A^{\text{EC}}$, and the CAR term is maximized. As current sources instead of voltage sources were used in the experiment, current–voltage characteristics from the semiconductor model (see Fig. 2) were used to convert $S_I(V_A, V_B)$ to $S_V(I_A, I_B)$ for comparison.

At all three temperatures, when the bias of the two junctions is of the same polarity, there is a clearly positive correlation, consistent with CAR. When the bias of the two junctions is of opposite polarity, equation (2) predicts a negative S_V , whereas in the experiment, S_V changes from slightly positive to slightly negative as the temperature decreases, suggesting a reduced A^{EC} , which indicates that the assumption $A^{\text{CAR}} = A^{\text{EC}}$ is oversimplified. As $V_A, V_B \rightarrow 0$, although S_{AB} is predicted to be $2k_B T(A^{\text{CAR}} - A^{\text{EC}})$, the experimental data show a positive correlation at 0.4 K that evolves to a sharp negative dip at 0.25 K, which is again not consistent with the assumption $A^{\text{CAR}} = A^{\text{EC}}$.

To understand this discrepancy, the Coulomb interactions in the leads need to be considered^{3,18,21}. We note that the charging

Figure 4 | Bias current dependence of local and non-local differential resistance. **a,b**, The local differential resistance at both temperatures 0.4 K (**a**) and 0.3 K (**b**), obtained by adding a small a.c. current modulation i_A to I_A and measuring the a.c. voltage modulation V_A . **c,d**, The non-local resistance is obtained at the same time by measuring the a.c. voltage modulation v_B , at 0.4 K (**c**) and 0.3 K (**d**). The black dashed lines indicate a strong reduction of the local differential resistance, approximately where the d.c. bias voltage is the same for the two junctions (the normal-state resistances of the two junctions are 15 and 19 k Ω respectively).

effect of solitary junctions has a strong temperature and bias dependence²⁹. As the Coulomb energy associated with charging the superconducting lead and charging the normal-metal leads changes, A^{CAR} and A^{EC} also change and this may lead to a non-monotonic temperature and bias dependence of S_{AB} . Note that here the junction capacitance is much smaller than that of the planar NISIN devices studied before⁹. As the charging energy is $E_C = e^2/2C$, where C is the capacitance of the tunnelling junction, a much stronger DCB effect is expected. The effect of DCB on noise correlation was recently considered with a renormalization group approach²¹, where the presence of Coulomb interactions in the normal-metal leads is treated as a reduction of interface transparency, resulting in a change of the sign and magnitude of the correlation. However, this approach does not seem to be applicable here as the Coulomb interaction in the superconducting lead was not taken into account. Further experiments with a modified local impedance near the normal-metal leads that enhances the DCB effect may facilitate comparison with theory^{3,21}. We also note that in most theoretical work only pair tunnelling (Andreev reflection, CAR and higher-order processes) is considered in the zero-temperature limit, whereas in the experiment, quasiparticle tunnelling dominates at finite temperatures because a reasonable differential resistance is required for measurement.

The local and non-local differential resistance corroborate the observation from the noise cross-correlation measurements: they also demonstrate that CAR clearly dominates, as shown in Fig. 4. (In Supplementary Notes 2, more details are given to show that both the reduction of local resistance and the negative non-local resistance are consistent with the noise measurements.) Surprisingly, even for the local differential resistance, there is clearly a reduction of resistance when the d.c. bias $V_A \approx V_B$, as indicated by the dashed line in Fig. 4. This strong reduction is not expected as the differential resistance is mainly determined by quasiparticle tunnelling, and

even if there is a small probability of pair tunnelling, direct Andreev reflection is expected to dominate CAR and elastic cotunnelling^{16,20}. However, this might suggest that DCB indeed enhances the splitting of a Cooper pair, especially when the electrons passing the two SN interfaces are of the same energy, in agreement with predictions³.

Figure 4b shows that at 0.4 K, the non-local resistance is negative in the subgap regime for both junctions, consistent with CAR. As the temperature decreases to 0.3 K, a non-local resistance peak evolves near zero bias. On further cooling to 0.25 K, the non-local resistance at the peak becomes positive, see Supplementary Information S1. Remarkably, if we take a one-dimensional cut at $I_B = 0$, the bias dependence of the non-local resistance is close to that previously reported where only one junction was biased^{9,15}. This confirms that the non-local resistance measured in the current set-up may have the same origin; although a more complete picture is obtained here with the two-dimensional scan. Further experiments with spin-selective normal-metal leads and Bell-inequality-type measurements^{21,30} will provide a conclusive check of non-local spin entanglement.

Methods

The devices were fabricated using standard two-angle electron-beam lithography and electron-gun evaporation. A polymethyl methacrylate/polydimethyl glutarimide bilayer was spin-coated on Si substrates with a 300 nm SiO_2 insulating layer for patterning the devices. For the particular device reported here, a 27 nm 99.999% pure Al film was deposited first in an electron-gun evaporator with a base pressure of 2.2×10^{-7} torr at a 40° angle and at a rate of 0.1 nm s^{-1} . After deposition, 0.2 torr pure oxygen gas was allowed into the chamber for about 5 min to create a thin layer of oxide. Then a 50 nm layer of Cu film was deposited at a normal angle. Devices from three batches were measured in an Oxford KelvinOx 100 or an Oxford KelvinOx 300 dilution refrigerator. The results obtained on these devices were consistent with each other and here we concentrate on a single device with the most complete data. As shown in Fig. 1, the junction size is about $0.3 \mu\text{m}$ by $0.45 \mu\text{m}$, and the distance between the two junctions is about $0.28 \mu\text{m}$, comparable to the superconducting coherence length ξ_S of diffusive Al.

The junction resistance is much larger than that of the normal-metal lead, which ensures a sensitive measurement of current across the junction, as voltage is the quantity actually measured here.

The current sources shown in Fig. 1 were realized by putting a large ballast resistor (1 G Ω) in series with a filtered voltage source. The a.c. voltage signals from the junctions were amplified using home-made battery-powered low-noise amplifiers. The voltage signals after amplification were sent to data acquisition cards and were analysed by a computer after digitization. To reduce noise coupled to devices at low temperature, π -filters on top of the cryostat were used. The measurement signals from the cryostat were amplified inside a μ -metal enclosure close to the cryostat. The bandwidth of measurement was limited by the roll-off effect because of the capacitance of the π -filter (about a few nanofarads).

The autocorrelation data shown in Fig. 2b at 4.2 K were measured in the frequency range from 500 to 1,000 Hz, whereas at 0.4 K different frequency ranges, from 200 to 600 Hz and from 120 to 130 Hz, were used and similar results were obtained. The voltage noise power S_V is fitted to

$$S_V = 2eI \coth\left(\frac{eV}{2k_B T}\right) \left(\frac{dV}{dI}\right)^2 + 2\left(I_n \frac{dV}{dI}\right)^2 + V_n^2$$

where I_n is the extrinsic current noise and V_n is the extrinsic correlated voltage noise. A good fit with data was found by using $I_n = 30$ fA (Hz) $^{-1/2}$ and $V_n = 0.7$ nV (Hz) $^{-1/2}$. These values are consistent with standard junction field-effect transistor amplifiers. Hence, in the subgap regime, the extrinsic current noise dominates the intrinsic shot noise.

The cross-correlation data shown in Fig. 3 were obtained in the frequency range from 2 to 6 Hz to avoid the roll-off effect and the side lobe of the 60 Hz line frequency peak. The efficiency of the cross-correlation measurement at this frequency range was compromised by the $1/f$ noise of the amplifiers. Nevertheless, owing to the large differential resistance, the bias dependence of S_{AB} is clearly visible.

Received 16 October 2009; accepted 12 April 2010;
published online 16 May 2010

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Acknowledgements

We thank M. Büttiker, W. Belzig and A. Levy Yeyati for comments on our preprint. This work was sponsored by the National Science Foundation through grant no. DMR-0604601.

Author contributions

J.W. fabricated samples and carried out measurements and analysis. J.W. and V.C. prepared the manuscript.

Additional information

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