## Production of Nonlocal Quartets and Phase-Sensitive Entanglement in a Superconducting Beam Splitter

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Three BCS superconductors  $S_a$ ,  $S_b$ , and S and two short normal regions  $N_a$  and  $N_b$  in a three-terminal  $S_a N_a S N_b S_b$  setup provide a source of nonlocal quartets spatially separated as two correlated pairs in  $S_a$  and  $S_b$ , if the distance between the interfaces  $N_a S$  and  $S N_b$  is comparable to the coherence length in S. Low-temperature dc transport of nonlocal quartets from S to  $S_a$  and  $S_b$  can occur in equilibrium, and also if  $S_a$  and  $S_b$  are biased at opposite voltages. At higher temperatures, thermal excitations result in correlated current fluctuations which depend on the superconducting phases  $\phi_a$  and  $\phi_b$  in  $S_a$  and  $S_b$ . Phase-sensitive entanglement is obtained at zero temperature if  $N_a$  and  $N_b$  are replaced by discrete levels.

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Regarding the manipulation of entangled states, quantum nanoelectronics is on the way to address the same fundamental issues with electrons as quantum optics does with photons. An entangled quantum state has a density matrix distinct from that of any "hidden-variable" theory. Two-particle entanglement can be probed [1] via the violation of the Bell inequality [2]. Multiparticle entanglement also has a high potential, for instance it [3] can be used to implement error correction codes.

Concerning superconductivity, two-particle entanglement can be generated at normal metal-superconductor N-S-N interfaces, by extracting a split Cooper pair from the BCS condensate of electron pairs [4-7]. We show in this Letter that a nanoscale three-terminal superconducting setup can produce nonlocal quartets separated as two pairs in different electrodes, therefore opening a route for a new generation of entanglers which could be controlled by an electromagnetic field. One must stress that here quartets are absent in the bulk superconductors which instead carry ordinary BCS pairing. This is in contrast with the destruction of the "ordinary" pair condensate in certain arrays of Josephson junctions [8], and its relationship with topological quantum computation [9,10]. Microscopically, nonlocal quartet transmission appears here as a generalization of so-called Cooper pair splitting at a double N-S-N interface, and it can be characterized by interference and noise. Recall that an Andreev pair in a normal metal electrode N results from the emission of a charge 2e from S at a N-S interface, by the process of Andreev reflection (AR). At a double N-S-N interface, spin-entangled [11] or energyentangled [12] pairs can be produced through crossed (or nonlocal) Andreev reflection (CAR) [13-15] involving evanescent quasiparticle states in S, on the coherence length  $\xi_s$ . CAR coexists with normal transmission through S without electron-hole conversion (elastic cotunneling EC) [16]. CAR or EC can be selected by their different Coulomb interaction energy [17], by their spin sensitivity [6,16], by their distinguishing energy dependence [18], or by their different signature in the nonlocal conductance and in the zero-frequency shot noise cross-correlations [5,11,19,20]. The new effects considered here do not require more advanced technology than the experiments on split pairs already realized with metallic structures [13] or with quantum dots [14].

In this Letter, a route to the production of nonlocal quartets is proposed on the basis of bunching of two Andreev pairs in a superconducting beam splitter made of conventional BCS superconductors. Indeed, two Josephson junctions separated by a distance  $d_S$  of the order of the coherence length  $\xi_S$  of S can be coupled by nonlocal coherent effects [21]. Here we study microscopically all the possible nonlocal effects and discuss their physical consequences. Remarkably, in a three-terminal  $S_a N_a S N_b S_b$  structure, nonlocal quartets can be separately transmitted as two correlated pairs in  $S_a$  and  $S_b$ . Nonlocal quartet transmission proceeds through double crossed Andreev reflection (dCAR), which coexists with double elastic cotunneling (dEC). The latter process yields Cooper pair transmission between  $S_a$  and  $S_b$  [22]. The phasesensitive dCAR is a new coherent nonlocal quantum channel, which has no direct analog for incoherent multiple Andreev reflections [23]. The elementary charges involved in dCAR and dEC are doubled as compared to CAR and EC. The four processes of CAR, EC, dCAR and dEC (see Fig. 1) will be treated on an equal footing [24], as well as AR at each of the  $S_a$ - $N_a$ -S or S- $N_b$ - $S_b$  interfaces which transfers Cooper pairs between S and  $S_a$  or  $S_b$ .

Cooper pair splitting in a  $S_a N_a S N_b S_b$  structure with arbitrary interface transparency can be described by generalizing the Andreev–Kulik–Saint-James bound states (ABSs) [25–28]. Those states are coherent superpositions of electrons and holes, forming in a short single channel



FIG. 1 (color online). Panel (a) shows a  $S_aS_b$  structure, where  $N_a$  and  $N_b$  have not been represented for clarity. The processes taking place in three-terminal  $S_aN_aSN_bS_b$  [panel (b)] and  $S_anSpS_b$  [panel (c)] structures are as follows: double crossed Andreev reflection (dCAR, red long dashed lines) producing a nonlocal quartet (a spatially separated pair of pairs), double elastic cotunneling (dEC, dotted black lines) exchanging pairs between  $S_a$  and  $S_b$ , and crossed Andreev reflection (CAR), thermally activated above the gaps of  $S_a$  and  $S_b$ . In addition, elastic cotunneling (EC) and local Andreev reflection (AR), not shown in the figure, also take place in  $S_aN_aSN_bS_b$ . With suitable gate voltages, the *n*- and *p*-doped semiconductors on panel (b) have a vanishingly small density of states at negative and positive energies, respectively. They filter CAR and dCAR, and eliminate EC, dEC, and AR.

junction a doublet at opposite energies  $(E_{-}, E_{+})$ , and carrying Josephson currents in opposite directions. In the  $S_a N_a S N_b S_b$  setup, the phases of  $S_a$ ,  $S_b$ , and S being  $\phi_a$ ,  $\phi_b$ , and  $\phi_s$ , and taking the bias voltages  $V_a$ ,  $V_b$  to be zero, the total currents  $I_a$ ,  $I_b$  are given at zero temperature T = 0by  $I_a(\phi_a, \phi_b) \approx (2e/\hbar) \sum_{n=1,2} (\partial E_n / \partial \phi_a), \ I_b(\phi_a, \phi_b) \approx$  $(2e/\hbar)\sum_{n=1,2}(\partial E_n/\partial \phi_b)$ . The  $E_n$  are the energies of the ABSs formed by hybridizing the ABSs of both junctions through dCAR and dEC. They depend on both phase differences  $\delta \phi_a = \phi_a - \phi_s$  and  $\delta \phi_b = \phi_b - \phi_s$ . If  $d_S \leq \xi_S$ , this leads (at lowest order in the dCAR and dEC processes) to  $I_a = I_a^0(\delta\phi_a) + I^{\text{dEC}}(\delta\phi_a - \delta\phi_b) + I^{\text{dEC}}(\delta\phi_a - \delta\phi_b)$  $I^{\text{dCAR}}(\delta\phi_a + \delta\phi_b) \quad \text{and} \quad I_b = I_b^0(\delta\phi_b) - I^{\text{dEC}}(\delta\phi_a - \delta\phi_b) + I^{\text{dCAR}}(\delta\phi_a + \delta\phi_b). \text{ Production of nonlocal}$ quartets (dCAR) and pair transmission (dEC) couple the coherent dc Josephson currents in  $S_a$  and  $S_b$  by the inverse crossed inductances  $(L^{-1})_{a,b} = \partial I_a(\phi_a, \phi_b)/\partial \phi_b$  and  $(L^{-1})_{b,a} = \partial I_b(\phi_a, \phi_b) / \partial \phi_a$ , which is an extension of the concept of crossed *conductances* in a  $N_a S N_b$  structure.

Voltages  $V_a$ ,  $V_b$  are applied now on the electrodes  $S_a$ ,  $S_b$ ( $V_S = 0$  is the reference voltage). Yet a dc Josephson current can flow from S to  $S_a$  and  $S_b$  if  $d_S \sim \xi_S$  [29], in addition to the standard ac Josephson currents. The intensity of this current can be seen as a synchronization of the phases of the ac oscillations. Indeed, considering for simplicity low transparency contacts, the double Josephson junction is described by the Hamiltonian  $\mathcal{H} = \mathcal{H}_{loc} + \mathcal{H}_{dCAR} + \mathcal{H}_{dEC} + 2e(\hat{n}_S V_S + \hat{n}_a V_a + \hat{n}_b V_b)$ , with  $\mathcal{H}_{loc} = -E_J \{\cos[\delta \phi_a(t)] + \cos[\delta \phi_b(t)]\}$ , and  $\mathcal{H}_{dCAR} = -E_J^{dCAR} \cos[\delta \phi_a(t) + \delta \phi_b(t)]$ ,  $\mathcal{H}_{dEC} = -E_J^{dEC} \cos[\delta \phi_a(t) - \delta \phi_b(t)]$ . The currents are obtained from Hamilton equations, for instance:

$$\left(\frac{\hbar}{2e}\right)I_{a}(t) = E_{J}\sin[\delta\phi_{a}(t)] + E_{J}^{\text{dCAR}}\sin[\delta\phi_{a}(t) + \delta\phi_{b}(t)] \\
+ E_{J}^{\text{dEC}}\sin[\delta\phi_{a}(t) - \delta\phi_{b}(t)].$$
(1)

Applying opposite voltages  $V_a = -V_b \equiv V$  leads to a dc Josephson effect for nonlocal quartets because the phase combination  $\delta \phi_a(t) + \delta \phi_b(t) = e(V_a + V_b)t/\hbar + \delta \phi_a + \delta \phi_b$  is then time independent. The Josephson effect for nonlocal quartets becomes ac only if the energy  $eV_a + eV_b$  acquired by the quartet when separately transmitted into  $S_a$  and  $S_b$  is finite. This result holds for any transparency.

The dc Josephson effect for nonlocal quartets is further considered for a  $S_a n S p S_b$  junction biased at opposite voltages, where the previous  $N_a$  and  $N_b$  metals have been replaced by n- and p-doped semiconductors. The conduction band edge on one side (n type) and the valence band edge on the other (p type) are at zero energy. The gaps in the density of states of the *n*- and *p*-doped semiconductors filter the processes with positive energies in *n*, and with negative energies in p [18] [Fig. 1(c)]. This excludes both the local Josephson effect and the nonlocal dEC, thus leaving at T = 0 only the nonlocal dCAR as a coherent coupling between the condensates [30]. In this ideal situation, one obtains a perfect superconducting beam splitter operating at the scale of the coherence length, and producing correlated pairs of Cooper pairs flowing in the leads  $S_a$  and  $S_b$ . Notice again that the biases  $V_a$  and  $V_b$  should be *opposite* in the coherent Josephson regime, while they are equal in a normal beam splitter  $N_a N N_b$  or a Cooper pair splitter  $N_a S N_b$ , where quasiparticles are emitted instead of pairs.

Let us now discuss the noise cross-correlations in a  $S_a N_a S N_b S_b$  structure. Zero-frequency thermal noise is present in the absence of applied voltage for sufficiently transparent single junctions [31,32]. Finite values are obtained for all the components of the correlators  $S_{i,i}(t) =$  $\langle \delta I_i(t+t') \delta I_i(t) \rangle$ , where  $\delta I_k$  is the current fluctuation in  $S_k$  (k = a, b). This equilibrium noise, due to thermally activated fluctuations between ABSs carrying opposite currents, is phase sensitive because the population of quasiparticles in thermal equilibrium exchanges charge with the condensate. The thermal noise can be very large for a long inelastic lifetime of the ABSs [31,32]. CAR and EC lead to components  $S_{a,b}^{CAR}$  and  $S_{a,b}^{EC}$  of  $S_{a,b}$ , which are independent of  $\delta \phi_a$  and  $\delta \phi_b$ . They correspond to CAR and EC assisted by thermal activation over the gap  $\Delta$  (Fig. 1). On the contrary, dCAR and dEC result in thermal fluctuations between hybridized ABSs. dCAR corresponds to random emission and absorption of nonlocal quartets between S and  $S_a$ ,  $S_b$ . dEC corresponds to random transmission of pairs between



FIG. 2 (color online). The plots show  $-(L^{-1})_{a,b}$  [panels (a), (c), (e)] and  $S_{a,b}$  [panels (b), (d), (f)] as a function of  $(\delta \phi_a, \delta \phi_b)$  for  $T/\Delta = 0.1$ ,  $T_N = 4 \times 10^{-4}$  [panels (a),(b)],  $T_N = 0.64$  [panels (c), (d)] and  $T_N = 1$  [panels (e), (f)]. The ratio  $\Delta/\Delta_S = 0.1$  is used.

 $S_a$  and  $S_b$ . The contributions  $S_{a,b}^{\text{dCAR}}$  and  $S_{a,b}^{\text{dEC}}$  of dCAR and dEC to  $S_{a,b}$  depend, respectively, on the phase combinations  $\delta\phi_a + \delta\phi_b$  and  $\delta\phi_a - \delta\phi_b$ . Generalizing Refs. [31–33], the zero-frequency noise cross-correlations  $S_{a,b}$  in the absence of applied voltage is written as

$$S_{a,b} = \frac{e^2}{\eta \hbar^2} \sum_{n} \frac{1}{\cosh(E_n/2k_B T)} \frac{\partial E_n}{\partial \phi_a} \frac{\partial E_n}{\partial \phi_b}, \qquad (2)$$

where the lifetime of the Andreev states  $1/\eta$  shows the relevance of pair-breaking effects in the superconductor [21]. Such noise correlations could be large at temperatures  $T^*$  comparable to the ABS energy level difference. The crossover temperature  $T^*$  is strongly reduced as the interface transparency increases.

Calculations of the equilibrium nonlocal inverse inductance  $(L^{-1})_{a,b}(\delta\phi_a, \delta\phi_b)$  and cross-correlations  $S_{a,b}$  $(\delta \phi_a, \delta \phi_b)$  are based on microscopic Nambu-Keldysh Green's functions [29] in which the interfacial hopping amplitude is accounted for by a self-energy. Arbitrary values of the temperature and of the normal-state transmission coefficient  $T_N$  can be treated in equilibrium because the time convolutions in the Dyson equations then simplify into products of Green's function depending only on energy. The microscopic calculations carried out for a three-dimensional ballistic superconductor apply to a voltage range in which the proximity effect is negligible.  $S_{a,b}$  goes to zero at T = 0because it is thermally activated over the ABS gap.  $(L^{-1})_{a,b}$ saturates at low T to the zero-temperature response of the condensate. As seen from perturbation theory in the tunnel amplitudes, both dCAR and dEC contribute with a positive value to  $S_{a,b}(\delta \phi_a = 0, \delta \phi_b = 0)$ . Thus, for tunnel contacts,  $S_{a,b}(\delta \phi_a = 0, \delta \phi_b = 0)$  is positive at the small  $T/\Delta = 0.1$  [see Fig. 2(b)]. The oscillations in  $-(L^{-1})_{a,b}(\delta\phi_a, \delta\phi_b)$  [Fig. 2(a)] match those of  $S_{a,b}(\delta\phi_a, \delta\phi_b)$  [see Fig. 2(b)]. They reflect the distinguishing phase dependences of dCAR and dEC. Increasing  $T_N$  has the effect of favoring the transmission of pairs by dEC from  $S_a$  to  $S_b$  (or from  $S_b$  to  $S_a$ ), and disfavoring their transmission as a pair of holelike quasiparticles by dCAR. The cross-shaped variations of  $(L^{-1})_{a,b}(\delta\phi_a, \delta\phi_b)$  and  $S_{a,b}(\delta\phi_a, \delta\phi_b)$  for intermediate  $T_N$  can be understood from the bound state energy level minima in the  $(\delta\phi_a, \delta\phi_b)$  plane for  $\delta\phi_a, \delta\phi_b \in \{0, \pi, 2\pi\}$ .

This superconducting beam splitter also generates entanglement between pair numbers in the two branches, as pairs are emitted two by two. Indeed a split Josephson current due to nonlocal quartets connects coherently pair number states  $|N_a\rangle|N_s\rangle|N_b\rangle$  with states  $|N_a + 1\rangle|N_s - 2\rangle|N_b + 1\rangle$ , entangling the added pairs in  $S_a$  and  $S_b$  [29]. To illustrate this, let us replace the N junctions by quantum dots  $D_a$ ,  $D_b$ . Indeed phase-sensitive entanglement is obtained in a  $S_a D_a S D_b S_b$ structure biased at voltages  $V_a = -V_b \equiv V$  larger than  $\Delta$ , with  $\Delta_S \gg \Delta$ , eV. In a first step,  $D_a$  and  $D_b$  are supposed to carry spin-degenerate orbitals. In addition, the gate voltages are such that  $D_a(D_b)$  has a level at  $eV_a(eV_b)$  but no level at  $-eV_a$  ( $-eV_b$ ). Quasiparticles are transmitted from  $S_a$  to  $D_a$  (from  $S_b$  to  $D_b$ ) but local Andreev processes between  $D_a$ and  $S(D_h \text{ and } S)$  are not possible. In the limit of large gaps, three terms contribute to the effective Hamiltonian of the two coherently coupled levels at energies  $eV_a$  (in  $D_a$ ) and  $eV_b$  (in  $D_b$ ): (i) The exchange of pairs between  $S_a$  and  $D_a$  $(S_b \text{ and } D_b): \mathcal{H}_{AR,a(b)} = -\alpha[\exp(i\phi_{a(b)})c^{\dagger}_{a(b),\uparrow}c^{\dagger}_{a(b),\downarrow} + H.c.]. (ii) Cooper pair splitting: \mathcal{H}_{CAR} = -\beta[c^{\dagger}_{a,\uparrow}c^{\dagger}_{b,\downarrow} + H.c.]$  $c_{h1}^{\dagger}c_{a1}^{\dagger}$  + H.c.]. (iii) Production of nonlocal quartets:  $\mathcal{H}_{dCAR}^{\text{tr},\text{tr}} = -\gamma [c_{a,\uparrow}^{\dagger} c_{b,\uparrow}^{\dagger} c_{b,\downarrow}^{\dagger} c_{b,\downarrow}^{\dagger} + \text{H.c.}]. \text{ Exact diagonalizations of } \mathcal{H}_{AR,a} + \mathcal{H}_{AR,b} + \mathcal{H}_{CAR} + \mathcal{H}_{dCAR} \text{ result in}$ an entangled ground state characterized by a positive concurrence, which depends on the values of  $\delta \phi_a$  and  $\delta \phi_b$  via the combination  $\delta \phi_a + \delta \phi_b$  typical of dCAR [29]. The Coulomb interaction Hamiltonian is  $\mathcal{H}_U = U\hat{n}_{a(b),\uparrow}\hat{n}_{a(b),\downarrow}$ , with  $\hat{n}_{a(b),\sigma}$  the number of spin- $\sigma$  electrons in dot a (in dot b). As U increases, the zero-temperature concurrence of the ground state remains finite because of virtual excitations coupling to dCAR.

Entanglement can also be more directly assessed by showing that no classical correlation can account for the crossed noise of nonlocal quartets. Let us consider  $S_a D_a S D_b S_b$  or  $S_a p S n S_b$  setups, biased at opposite  $V_a$ and  $V_b$  larger than  $\Delta$ , and with  $\Delta_S \gg \Delta$ , eV[assumption (A1)]. The notation  $\langle N_a(t,\tau) \rangle_\rho$  stands for the average number of electrons transmitted into electrode  $S_a$  in the time interval  $[t, t + \tau]$ . The average over the hidden-variable density matrix  $\rho = \int d\lambda \rho_a(\lambda) \otimes \rho_b(\lambda)$  is noted  $\langle \cdots \rangle_\rho$  [11]. We make the assumption (A2) that each subsystem a and b is separately described by quantum solid-state physics: only communication through S corresponds to a hidden variable. The assumption (A2) leads to  $\langle \delta N_a(t,\tau) \delta N_b(t,\tau) \rangle_{qu} = \langle \delta N_a(t,\tau) \delta N_b(t,\tau) \rangle_\rho$ , with

$$\langle \delta N_a(t,\tau) \delta N_b(t,\tau) \rangle_{\rho} = \int d\lambda f(\lambda) \langle \delta N_a(t,\tau) \rangle_{\lambda} \langle \delta N_b(t,\tau) \rangle_{\lambda},$$
(3)

where  $f(\lambda)$  is the probability density of the hidden variable  $\lambda$ . The value of  $\delta N_a(t, \tau)$  for the specific value  $\lambda$  of the hidden variable is denoted by  $\langle \delta N_a(t, \tau) \rangle_{\lambda}$ .

An additional assumption (A3) concerning the setup is made in order to simplify the discussion: the gap  $\Delta_S$  is larger than the bandwidth W of the superconductors  $S_a$  and  $S_b$ , and the linewidth broadening in S is  $\eta_S = 0$ . The three assumptions (A1), (A2) and (A3) imply that charge transport is blocked at any temperature because the  $D_a$  and  $D_b$ , or the *n* and *p* energy filters [assumption (A1)], suppress all Josephson processes taking place locally within each subsystem a or b for any realization of the hidden variable  $\lambda$  [assumption (A2)]. No quasiparticle is transmitted from S to  $S_a$  (from S to  $S_b$ ) within subsystem a (b) for any value of  $\lambda$  [assumption (A3)]. The equality  $\langle \delta N_a(t, \tau) \delta N_b(t, \tau) \rangle_{qu} = 0$  is then obtained because  $\langle \delta N_a(t,\tau) \rangle_{\lambda} = \langle \delta N_b(t,\tau) \rangle_{\lambda} = 0$ . The following two statements are in conflict: (i) The cross-correlations are finite; and (ii) The nonlocal processes is a classical communication (related to some hidden variables) rather than the quantum mechanical CAR, dCAR.

For the  $S_a n S p S_b$  structure considered above, the crosscorrelations are vanishingly small at zero temperature and the Bell-like argument does not imply entanglement in this case. Indeed, the ground state of a Josephson junction is entangled only if the total number of pairs is fixed [29]. However, the denomination *phase-sensitive entanglement* is appropriate at zero temperature for the  $S_a D_a S D_b S_b$ structure because the cross-correlations are finite at T = 0, and the Bell-like argument is in agreement with the direct calculation of the concurrence, being also finite.

To conclude, we propose to generate nonlocal quartets in the solid state, by producing correlated pairs in a superconducting beam splitter involving three superconductors. The nonlocal inductance and the phase-sensitive thermal cross-correlations may be probed in future experiments. Of particular interest is the possibility to obtain entanglement at zero temperature from current-current cross-correlations in a  $S_a D_a S D_b S_b$  structure. Nonlocal Shapiro steplike experiments are also promising for investigating dCAR and dEC, in the sense that resonances due to dCAR or dEC could be obtained if the ac voltage oscillations are synchronized in two incoming channels  $S_a$  and  $S_b$ .

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