Quantum master equations for the superconductor–quantum dot entangler

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The operation of a source of entangled electron spins, based on a superconductor and two quantum dots in parallel, is described in detail with the help of quantum master equations. These are derived including the main parasitic processes in a fully consistent and nonperturbative way, starting from a microscopic Hamiltonian. The average current is calculated, including the contribution of entangled and nonentangled pairs. The constraints on the operation of the device are illustrated by a calculation of the various charge state probabilities.

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I. INTRODUCTION

Entanglement is a basic resource in quantum computation and quantum communication.[1] Recently, various experiments for quantum information processing schemes have been successfully implemented with photons such as Bell inequality violation[2] or teleportation.[3,4]

Any system with a two-level quantum degree of freedom is a possible candidate to carry a quantum bit. One of such is the electron and its spin. In principle, individual electrons can be manipulated in a quantum circuit and have the advantage of promising high-level integration in electronic devices.[5] Notice that the electron flow can be in principle much larger than the photon flow in equivalent optical devices where attenuation is necessary to produce individual photons. Moreover, photons essentially do not interact except during their generation process, whereas Coulomb correlations between electrons in a quantum circuit open the possibility for new operations between quantum bits.[6,7]

Nonlocality in quantum mechanics can be probed by letting two quantum degrees of freedom interact, and subsequently separating these two systems. Here, electronic entanglement can be created using a superconductor,[8,9] where two electrons forming a Cooper pair are in a singlet state. The superconductor is coupled to two arms, each of them collecting one electron from each Cooper pair. The emission of one electron in each lead from the same Cooper pair corresponds to the so-called Crossed Andreev process,[10–12] which can be understood as a nonlocal Andreev reflection: the emission of one of the electrons can be seen as the absorption of a hole with opposite spin and opposite momentum. The two electrons forming the singlet are then spatially separated. It is then necessary to avoid the "ordinary" Andreev reflection where the two electrons go into the same lead. This selection can be enforced, either with the help of spin filters, leading to energy entanglement.[9] Or alternatively, one can use energy filters, leading to spin entanglement.[8,9] Quantum dots with Coulomb blockade, inserted in each branch, can efficiently select the crossed Andreev process. As another possibility, the superconductor can be replaced by a normal quantum dot.[6,13] In this paper, the studied device consisting of a superconductor connected to two quantum dots in parallel will be called the entangler (see Fig. 1). Branching currents in the right and left leads were calculated for this entangler in Ref. 8 using a *T*-matrix approach. Entanglement can be probed by sending the electrons from a splitted pair into a beam splitter[14] and by measuring noise correlations.[15]

In the present paper a microscopic derivation of quantum master equations[16] for the entangler is presented. It provides a simple, intuitive approach to probe entanglement and to monitor the effect of parasitic processes. Compared to a *T*-matrix derivation,[8] this approach has the advantage of describing the whole charge dynamics in a nonperturbative way (this statement will be qualified below). This allows one to derive not only the average current but also the higher moments of the current distribution. Another point is that quantum master equations can be applied to any arbitrary quantum system containing superconducting elements, or to another kind of entangler.

Over the past years a great interest has been devoted to the description of the transport properties through devices containing coupled nanostructures, where quantum interference has a strong influence. A rather accessible method, generalizing the classical master equations,[17] has been developed in Ref. 18 where Bloch-type quantum rate equations have been derived using the Schrödinger equation. When the system is an isolated quantum dot in the Coulomb blockade, only the diagonal elements of the density matrix (the occu-

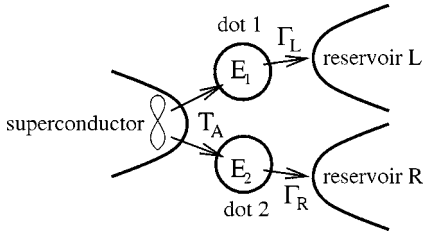

FIG. 1. The Entangler setup: a superconductor injects electrons in quantum dots D_1 and D_2, whose energies in state $|1\rangle$ (i.e., one excess electron) are, respectively, E_1 and E_2. Electrons in the dots can subsequently tunnel into the normal reservoirs L, R.

pation probabilities) enter the rate equations. On the other hand, when the transfer of electrons through a quantum device goes through a superposition of states in the different parts of this device, nondiagonal matrix elements will appear in the equations of motion. The master equations then take into account coherent processes and are a generalization of the Bloch equations.[19]

The microscopic derivation of these equations provides a good understanding of the correspondence between quantum and classical descriptions of transport in mesoscopic systems. The crucial point is the decoupling between the time scales which specify, first, the dynamics inside the reservoirs and, secondly, the inverse rates for coupling the quantum states and the leads. This decoupling procedure is justified as long as the time scales characterizing transfer within the quantum system and injection (emission) from (to) the reservoirs are both large compared to the time scale for fluctuations within the reservoirs. This is equivalent to a markoffian hypothesis.[19]

Quantum master equations have been derived in the case of sequential tunneling within quantum dots coupled to normal reservoirs, using a microscopic Hubbard-type Hamiltonian.[18,20] In the present work, it is generalized to electron transfer which is mediated by high lying virtual states. Consider now the case of a superconductor coupled to quantum dots: single-electron tunneling does not conserve energy and is forbidden as the electron transfer is accompanied by the emission of a Bogolubov quasiparticle. However two-electron events such as Andreev processes (transfer of a pair of electrons out of the superconductor) and superconducting cotunneling (S cotunneling) processes[21] (transfer of an electron from one dot to another via the superconductor) connect low-energy states, and thus enter the lowest-order contribution to the tunneling current from the superconductor. One simplification would be to assume that the two-electron tunneling processes occur simultaneously, and are described by a pair Hamiltonian: rate equations have been written recently in this manner for the transport processes in a teleportation cell which employs an array of normal and superconducting quantum dots.[7] However, in presence of transport channels mixing different processes, it is safer to derive quantum master equations starting directly from the microscopic Hamiltonian. This is achieved in the present work, taking into account the main parasitic processes. The sequence of relevant steps will clearly require virtual states which contribute to Andreev and cotunneling events. After having established the equations including coherent quantum-mechanical effects and Coulomb blockade, we will determine their range of validity and show the relevance of the lifetime of virtual states. The derivation of quantum master equation is nonperturbative with regard to transitions within the entangler, while the coupling to the leads is treated within the Fermi golden rule as in the orthodox theory of Coulomb blockade.[17]

The paper is organized as follows. In Sec. II, we present the system and its energy scales, together with the crossed Andreev process—the main process—and the important parasitic processes that can occur during its evolution. This allows one to write the many-excitation wave function which is the starting point of each microscopic derivation. This derivation is first described in Sec. III for the crossed Andreev process, without any parasitic process. Parasitic processes are presented next, and compared in Sec. IV before giving the complete description of the system by quantum master equations in Sec. V and Appendix B. Section VI provides the physical discussion of the operation of the device as a function of its parameters.

II. THE ENTANGLER DEVICE AND ITS PARAMETERS

A. The model

Let us first provide a qualitative description of the entangler. The setup involves a superconductor (S) coupled by tunneling barriers to two quantum dots (D_1 and D_2) which are themselves coupled to normal leads L and R (see Fig. 1). Only one level is retained in each dot, assuming the level separation in each dot to be large enough.[8] The energy levels of the dots can be tuned by external gate voltages. The microscopic Hamiltonian of the entire system is the following:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{tunnel}}, \tag{1}$$

where

$$\mathcal{H}_0 = \sum_{k,\sigma} E_k \gamma_{k\sigma}^\dagger \gamma_{k\sigma} + E_1 d_{1\sigma}^\dagger d_{1\sigma} + E_2 d_{2\sigma}^\dagger d_{2\sigma} + U_1 n_{1\sigma} n_{1-\sigma} + U_2 n_{2\sigma} n_{2-\sigma} + \sum_{l\sigma} E_l a_{l\sigma}^\dagger a_{l\sigma} + \sum_{r\sigma} E_r a_{r\sigma}^\dagger a_{r\sigma}, \tag{2}$$

where $\gamma_{k\sigma}$, $d_{i\sigma}$, $a_{l\sigma}$, $a_{r\sigma}$ are destruction operators for Bogolubov quasiparticles, dot electrons, and reservoir electrons. $n_{i\sigma}=d_{i\sigma}^\dagger d_{i\sigma}$ is the occupation number in the dots, which enters the Hubbard repulsion term with coupling constants U_1 and U_2. A possible interdot repulsion is omitted here for sake of simplicity, but it could easily be incorporated in the energies of various charge states of the two dots system.

The tunnel Hamiltonian which connects these elements by a one-electron transition reads

$$\mathcal{H}_{\text{tunnel}} = \sum_{k,\sigma} \Omega_{k1} d_{1\sigma}^\dagger c_{k\sigma} + \sum_{k,\sigma} \Omega_{-k2} d_{2-\sigma}^\dagger c_{-k-\sigma} + \sum_{l,\sigma} \Omega_l a_{l\sigma}^\dagger d_{1\sigma} + \sum_{r,\sigma} \Omega_r a_{r\sigma}^\dagger d_{2\sigma} + \text{H.c.} \tag{3}$$

with a single electron tunneling amplitude Ω_1 (Ω_2) between S and D_1 (S and D_2), and Ω_l (Ω_r) between D_1 and L (between D_2 and R). $\sigma=\{\frac{1}{2},-\frac{1}{2}\}$ is the spin variable. Note that $\mathcal{H}_{\text{tunnel}}$ is written in the Fourier space. Point contacts are assumed between S and dots 1 and 2 (in $\vec{r}_1$ and $\vec{r}_2$) thus the tunneling term is $\Omega_i d_{i\sigma}^\dagger c_{r\sigma}$,[8] which can be written in the Fourier space $\Sigma_k \Omega_i e^{i\vec{k}\cdot\vec{r}} d_{i\sigma}^\dagger c_{k\sigma} = \Sigma_k \Omega_{ki} d_{i\sigma}^\dagger c_{k\sigma}$. The effective momentum dependence of the tunneling amplitude Ω_{ki} introduces a geometrical factor, which can strongly influence the transition amplitude for processes involving the two quantum dots. During the injection process, Cooper pairs are initially separated into one electron in a dot and one quasiparticle in S. We introduce the Bogulubov transformation

$$c_{k\sigma}^{\dagger} = u_k \gamma_{k\sigma}^{\dagger} + \sigma v_k \gamma_{-k-\sigma} S^{\dagger},$$

$$c_{k\sigma} = u_k^{*} \gamma_{k\sigma} + \sigma v_k^{*} \gamma_{-k-\sigma}^{\dagger} S \tag{4}$$

with

$$u_k = \frac{1}{\sqrt{2}} \left(1 + \frac{\xi_k}{E_k} \right)^{1/2}, \tag{5}$$

$$v_k = \frac{1}{\sqrt{2}} \left(1 - \frac{\xi_k}{E_k} \right)^{1/2} e^{i\phi_S}, \tag{6}$$

$$E_k = \sqrt{\xi_k^2 + \Delta^2} = \sqrt{\frac{\hbar^2 k^2}{2m} - \mu_S + \Delta^2}. \tag{7}$$

Here S stands for the annihilation of a Cooper pair[22] and ϕ_S is the superconductor's phase. The two electrons from a Cooper pair become an entangled pair of electrons (only the singlet state is involved) when going into different leads. Current flow is imposed by a voltage bias $\Delta\mu$ between the superconductor and the leads. The basic mechanism for entanglement is based on a crossed Andreev process between the superconductor and the two quantum dots, forced by the Coulomb blockade in the dots. First, two entangled electrons are created in D_1 and D_2 via a virtual state which contains a quasiparticle in S whose energy is larger than Δ_S, the superconducting gap. This process is coherent, and couples the superconducting chemical potential μ_S and the final energy of the pair in the dots E_1+E_2. This Andreev process probability is optimized at $E_1+E_2=\mu_S$, and behaves similar to a narrow two-particle Breit-Wigner resonance. Then the two electrons tunnel independently to each lead. This whole sequence of events forms the Crossed Andreev channel.

B. Working conditions

Next, the relevant parameters describing the device are discussed, following Ref. 8. First, the charge states of the quantum system have to be well separated to avoid transitions due to thermal excitations. But the thermal energy must be large enough in comparison to the transition probability to allow the markoffian hypothesis. Therefore $\Gamma_{L,R} \ll k_B\Theta \ll E_i - \mu_{L,R}$. In order to conserve spin and thus the singlet state during the electron transfer, spin flip must be excluded. Thus each dot cannot carry a magnetic moment which could interfere with an electron coming from S, i.e., it must carry an even number of electrons.[8] Moreover, when an electron is deposited on a dot, another electron of this dot with opposite spin could in principle escape to the normal leads thus spoiling the entanglement. This spin-flip process is suppressed when the dot level spacing $\delta\epsilon$ is larger than the imposed bias $\Delta\mu$ and the temperature $k_B\Theta$. Entanglement loss can also occur because of electron-hole excitations out of the Fermi sea of the leads during the tunneling sequence. Such many-particle contributions are suppressed if the resonance width $\Gamma_{L,R}=2\pi\rho_{L,R}(E_{1,2})|\Omega_{L,R}(E_{1,2})|^2$ is smaller than $E_{1,2}-\mu_{L,R}$. This justifies the microscopic Hamiltonian of Eq. (1).

Next, given this Hamiltonian, one needs to justify the derivation of the quantum master equation. Single-electron tunneling from the superconductor to the leads via the dots is avoided because it implies the creation of a quasiparticle in S. This process costs at least Δ_S which is assumed to be much larger than $\Delta\mu$ and $k_B\Theta$.

C. Parasitic processes

The main purpose of this device is to force the two electrons from a pair to propagate in the two different leads. In a clean three-dimensional superconductor, this process is decreased by a geometrical factor $\gamma_A=e^{-r/\pi\xi_0}[\sin(k_F r)/k_F r]$ (ξ_0 is the superconductor coherence length and $r=|\vec{r}_1-\vec{r}_2|$ is the distance of the two contacts between dots and S). The crossed Andreev amplitude is then $\gamma_A T$, with $T=(\pi/2)N(0)\Omega_1\Omega_2$. In addition to the decay on ξ_0, the algebraic factor can be improved by reducing the dimensionality[23] or using a dirty superconductor.[24] Incidentally, the finite width of the contacts may introduce diffraction corrections to the geometrical factor. Note that when taking into account the momentum-dependent tunneling, the geometrical factor can be modified.[25]

There are three main parasitic processes which could decrease the entangler efficiency. Two of them create different channels of emission of two electrons coming from a Cooper pair, for which the two electrons can tunnel to the same lead.[8] Although they involve higher energy intermediate states, those do not suffer from the geometrical factor of the crossed Andreev channel. In addition, an elastic cotunneling—this process will be called S cotunneling in what follows—connects every channel to other processes by transferring an electron between the two dots via S.

The two electrons of a Cooper pair can tunnel through the same dot by an Andreev process (see Fig. 3). Because of double occupancy, the pair would get an energy U due to Coulomb repulsion. This is a coherent process between two energy levels with a large energy difference U. Because this energy cost is much larger than the Andreev process probability amplitude $T_i \sim N(0)\Omega_i^2$ involving a single lead, this process is strongly suppressed.[8] Alternatively, a pair could propagate to the same lead if the first electron injected on a given dot leaves it before the second electron is deposited on either dots. It goes to the corresponding lead while its twin electron "has been staying in S" as part of a quasiparticle (see Fig. 4). The latter can then choose toward which dot it will tunnel. It will prefer the same dot in order to avoid paying the geometrical factor. This latter process costs Δ_S and thus can be suppressed with $\Delta_S \gg \gamma_A T$. Let us notice that this process requires three transitions, including one transition to a reservoir, thus it is not coherent.

By a S-cotunneling process via S, an electron can tunnel from D_1 (D_2) to D_2 (D_1) (see Fig. 5). This is a coherent process between two discrete energy levels E_1 and E_2 for a single electron in the two dots or U_1 and E_1+E_2 for a doubly occupied dot (U_2 and E_1+E_2 for the opposite configuration). Cotunneling is characterized by an amplitude $\gamma_C T$, with its own geometrical factor γ_C. If the energy difference between the two coupled levels is much larger than the process amplitude T_C, this process will be weak.

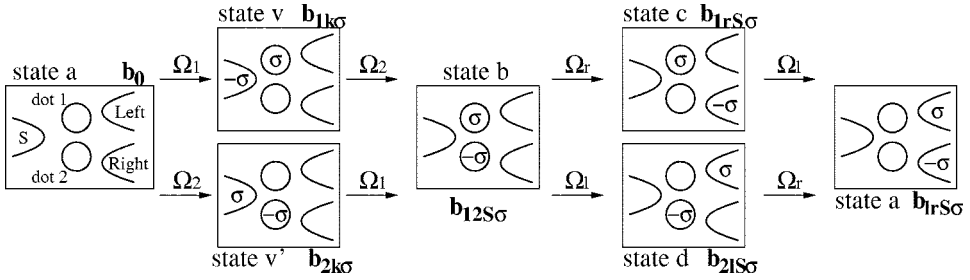

FIG. 2. Sequence of states for the crossed Andreev channel of the entangler. For instance, $b_{ik,\sigma}$ denotes the amplitude to have an electron in dot i while a quasiparticle is created in the superconductor. First an electron is deposited in either dot, next the second electron tunnels and forms a singlet state in the pair of dots, next either electron is absorbed in the reservoir, and finally the two dots are empty.

To summarize, the working regime of the device is the following:

$$\Delta_S, U, |E_1 - E_2| > \delta\epsilon > \Delta\mu, k_B\Theta > \Gamma_{L,R}, T_A, T_C. \quad (8)$$

This working regime contains the justifications for the approximations made in the derivation of the master equation: the markoffian approximation and the relevant processes involving at most two successive virtual states with only one quasiparticle in S. In what follows we also assume that $\Delta\mu > k_B\Theta$, in order to ensure the irreversibility of the pair production.

III. MASTER EQUATIONS FOR THE CROSSED ANDREEV CHANNEL

The transport channels which are described above can be characterized by the charge configuration of the isolated quantum system for each step of the entangler operation. The quantum system is composed of the dots and the superconductor, but its dynamics can be directly probed by integrating out excitations in the reservoirs and superconductor. Using the Schrödinger equation and generalizing the procedure of Ref. 18, it is shown here how to derive quantum master equations which describe the evolution of the reduce density matrix of the system. As a starting point, we consider the dynamics in the situation where only the crossed Andreev process and one-electron relaxation processes are effective—the ideal regime. The wave function is thus chosen to include charge states involved in this particular channel. A reduced Hilbert space containing the lowest energy states and the required virtual intermediate states is chosen (containing a single quasiparticle in S).

The many-excitation wave function for this problem is written as

$$\begin{aligned}|\Psi(t)\rangle = \Bigg[& b_0(t) + \sum_{k,\sigma} b_{1k,\sigma}(t) d^\dagger_{1\sigma}\gamma^\dagger_{-k-\sigma} S + \sum_{k,\sigma} b_{2k,\sigma}(t) d^\dagger_{2-\sigma}\gamma^\dagger_{k\sigma} S + \sum_{\sigma} b_{12S,\sigma}(t) d^\dagger_{1\sigma} d^\dagger_{2-\sigma} S + \sum_{l,\sigma} b_{2lS,\sigma}(t) a^\dagger_{l\sigma} d^\dagger_{2-\sigma} S \\ & + \sum_{r,\sigma} b_{1rS,\sigma}(t) d^\dagger_{1\sigma} a^\dagger_{r-\sigma} S + \sum_{lr,\sigma} b_{lrS,\sigma}(t) a^\dagger_{l\sigma} a^\dagger_{r-\sigma} S + \sum_{lr,\sigma,k',\sigma'} b_{lrS\sigma,1k'\sigma'}(t) d^\dagger_{1\sigma'} \gamma^\dagger_{-k'-\sigma'} a^\dagger_{l\sigma} a^\dagger_{r-\sigma} SS' \\ & + \sum_{lr,\sigma,k',\sigma'} b_{lrS\sigma,2k'\sigma'}(t) d^\dagger_{2-\sigma'} \gamma^\dagger_{k'\sigma'} a^\dagger_{l\sigma} a^\dagger_{r-\sigma} SS' + \cdots + \cdots + \cdots \Bigg] |0\rangle, \end{aligned} \quad (9)$$

where $b\cdots(t)$ are the time-dependent amplitudes for finding the system in the corresponding states with the initial conditions $b_0(0)=1$ and all other $b(0)$ are zero. The indices indicate the electron occupation in the dots and reservoirs, as depicted in Fig. 2. The use of Schrödinger equation and the form of $|\Psi(t)\rangle$ call for some comments. In fact, as said above, the temperature is not zero thus one should in principle rely on a density matrix description from the beginning. Yet, under the condition $\Gamma_{L,R} < k_B\Theta < \Delta\mu$, one can simply use the Schrödinger equation in a reduced subspace of states.[18] Those states for instance do not include electron-hole excitations in the same reservoir: these are supposed to relax on a very short time, due to inelastic processes occurring in L and R. On the contrary, all possible charge and spin states on the dots, together with all excitations including holes in L and electrons in R, are considered. Summing on these reservoir states eventually lead to the equations for the two reduced density matrix.[18]

After substituting Eq. (9) into the Schrödinger equation $i|\dot{\psi}(t)\rangle = \mathcal{H}|\psi(t)\rangle$, an infinite set of coupled linear differential equations is obtained for $b(t)$ by projecting $i\langle\psi_i|\dot{\psi}(t)\rangle = \langle\psi_i|\mathcal{H}|\psi(t)\rangle$ for each state $|\psi_i\rangle$. $|\psi_i\rangle$ characterizes the quantum state of the total system including the environment. Applying the Laplace transform

$$\tilde{b}(E) = \int_0^\infty e^{i(E+i\eta)t} b(t) dt \tag{10}$$

and taking into account the initial conditions, an infinite set of algebraic equations is obtained for the amplitudes $\tilde{b}(E)$ (see Fig. 2):

$$(E + i\eta)\tilde{b}_0 - \sum_{k\sigma} \sigma v_k \Omega_{k1}^* \tilde{b}_{1k\sigma} + \sum_{k\sigma} \sigma v_k \Omega_{-k2}^* \tilde{b}_{2k\sigma} = i, \tag{11a}$$

$$(E + i\eta - E_1 - E_k)\tilde{b}_{1k\sigma} = \sigma v_k^* \Omega_{k1} \tilde{b}_0 + u_k \Omega_{-k2}^* \tilde{b}_{12S\sigma}, \tag{11b}$$

$$(E + i\eta - E_2 - E_k)\tilde{b}_{2k\sigma} = -\sigma v_k^* \Omega_{-k2} \tilde{b}_0 - u_k \Omega_{k1}^* \tilde{b}_{12S\sigma}, \tag{11c}$$

$$\begin{aligned}(E + i\eta - E_1 - E_2)\tilde{b}_{12S\sigma} &= \sum_k u_k^* \Omega_{-k2} \tilde{b}_{1k\sigma} - \sum_k u_k^* \Omega_{k1} \tilde{b}_{2k\sigma} \\ &+ \sum_r \Omega_r \tilde{b}_{1rS\sigma} + \sum_l \Omega_l \tilde{b}_{2lS\sigma},\end{aligned} \tag{11d}$$

$$(E + i\eta - E_1 - E_r)\tilde{b}_{1rS\sigma} = \Omega_r \tilde{b}_{12S\sigma} + \sum_l \Omega_l \tilde{b}_{lrS\sigma}, \tag{11e}$$

$$(E + i\eta - E_2 - E_l)\tilde{b}_{2lS\sigma} = \Omega_l \tilde{b}_{12S\sigma} + \sum_r \Omega_r \tilde{b}_{lrS\sigma}, \tag{11f}$$

$$\begin{aligned}(E + i\eta - E_l - E_r)\tilde{b}_{lrS\sigma} &- \sum_{k'\sigma'} \sigma' v_k \Omega_{k1}^* \tilde{b}_{lrS\sigma,1k'\sigma'} \\ &+ \sum_{k'\sigma'} \sigma' v_k \Omega_{-k2}^* \tilde{b}_{lrS\sigma,2k'\sigma'} = \Omega_l \tilde{b}_{1r\sigma} + \Omega_r \tilde{b}_{2l\sigma}, \dots .\end{aligned} \tag{11g}$$

Each term corresponds to the transition between two successive states. Each transition leads to the creation or annihilation of a quasiparticle either in S or in a reservoir. There is a fundamental difference between the two types of transitions. The first one involves an excited state whose lifetime is so small ($\tau_{qp} \sim 1/\Delta_S \ll 1/T$) that coherence is kept until the quasiparticle is destroyed. On the other hand, in the reservoirs, quasiparticles instantaneously decay ($\tau_{relax} \sim 1/E_F \ll 1/\Gamma$) so coherence is lost (Markoff process). To simplify the system of equations, the expression for $\tilde{b}$ is substituted in terms of the type $\Sigma \Omega \tilde{b}$ from equations containing sums. Every sum over the continuum states (k,l,r) is replaced by integrals (see Appendix A). Crossed terms [such as $\Sigma_l \tilde{b}_l \Omega_l \Omega_r/(E - E_l)$] vanish,[18] and the following set of equations is obtained:

$$[E + i\eta - 2c(T_1 + T_2)]\tilde{b}_0 = 2\gamma_A T e^{i\phi_S}(\tilde{b}_{12S,\sigma} - \tilde{b}_{12S,-\sigma}), \tag{12a}$$

$$\left(E + i\eta - E_1 - E_2 - c'(T_1 + T_2) + i\frac{\Gamma_L}{2} + i\frac{\Gamma_R}{2}\right)\tilde{b}_{12S,\sigma} = 2\sigma\gamma_A T e^{-i\phi_S}\tilde{b}_0, \tag{12b}$$

$$\left(E + i\eta - E_1 - E_r + i\frac{\Gamma_L}{2}\right)\tilde{b}_{1rS,\sigma} = \Omega_r \tilde{b}_{12S,\sigma}, \tag{12c}$$

$$\left(E + i\eta - E_2 - E_l + i\frac{\Gamma_R}{2}\right)\tilde{b}_{2lS,\sigma} = \Omega_l \tilde{b}_{12S,\sigma}, \tag{12d}$$

$$\begin{aligned}[E + i\eta - E_l - E_r - 2c(T_1 + T_2)]\tilde{b}_{lrS,\sigma} &= 2\gamma_A T e^{i\phi_S}(\tilde{b}_{lrS\sigma,12S'\sigma'} - \tilde{b}_{lrS\sigma,12S'-\sigma'}) \\ &+ \Omega_l \tilde{b}_{1rS,\sigma} + \Omega_r \tilde{b}_{2lS,\sigma}, \dots ,\end{aligned} \tag{12e}$$

with $T_i = \frac{\pi}{2} N(0)\Omega_i^2$ and c, c' are numerical constants (see Appendix A), involved in self-energy corrections. Here the coefficients for virtual states (states $|v\rangle$ and $|v'\rangle$ in Fig. 2) have disappeared from the equations. This is the consequence of the succession of quasiparticle creation and annihilation transitions forced by the assumption that two quasiparticles cannot coexist in S.

The singlet/triplet basis is now chosen. For instance, in the global wave function $\Sigma_\sigma \tilde{b}_{12S,\sigma} d_{1\sigma}^\dagger d_{2-\sigma}^\dagger$ is replaced by $\tilde{b}_{12S}^{\text{singlet}}(d_{1\sigma}^\dagger d_{2-\sigma}^\dagger - d_{1-\sigma}^\dagger d_{2\sigma}^\dagger)/\sqrt{2} + \tilde{b}_{12S}^{\text{triplet}}(d_{1\sigma}^\dagger d_{2-\sigma}^\dagger + d_{1-\sigma}^\dagger d_{2\sigma}^\dagger)/\sqrt{2}$. From Eq. (12b) one can say that coefficients $\tilde{b}_{12S,\sigma}$ and $\tilde{b}_{12S,-\sigma}$ for a given spin are opposite. This is the same for $\tilde{b}_{1rS,\sigma}$ and $\tilde{b}_{1rS,-\sigma}$, $\tilde{b}_{2lS,\sigma}$ and $\tilde{b}_{2lS,-\sigma}$. The tunnel Hamiltonian conserves spin, therefore there is no coupling towards triplet spin states. Thus $\tilde{b}_{ijS}^{\text{singlet}} = \sqrt{2} b_{ijS,\sigma} = -\sqrt{2} b_{ijS,-\sigma}$ and $\tilde{b}_{ijS}^{\text{triplet}} = 0$.

The density matrix elements of the set-up are now introduced. The Fock space of the quantum dots consists of four possible charge states: $|a\rangle$ levels E_1 and E_2 are empty, $|b\rangle$ levels E_1 and E_2 are occupied, $|c\rangle$ level E_1 is occupied, $|d\rangle$ level E_2 is occupied. Reservoirs states are identified by n, the number of pairs of electrons out from S to the reservoirs. To obtain the reduced density matrix, elements are summed over n:

$$\sigma_{\alpha\beta} = \sum_{n=0}^{\infty} \sigma_{\alpha\beta}^{(n)}. \tag{13}$$

In every state, electrons are paired in a singlet state. The matrix elements are defined as

$$\sigma_{aa} = |\tilde{b}_0(t)|^2 + \sum_{l,r} |\tilde{b}_{lrS}^{\text{singlet}}|^2 + \sum_{l<l',r<r'} |\tilde{b}_{lrS,l'r'S'}^{\text{singlet}}|^2 + \cdots ,$$

$$\sigma_{bb} = |\tilde{b}_{12S}^{\text{singlet}}|^2 + \sum_{l',r'} |\tilde{b}_{l'r'S',12S}^{\text{singlet}}|^2 + \cdots ,$$

$$\sigma_{cc} = \sum_r |\tilde{b}_{1rS}^{\text{singlet}}|^2 + \sum_{l',r'<r} |\tilde{b}_{l'r'S',1rS}^{\text{singlet}}|^2 + \cdots ,$$

$$\sigma_{dd} = \sum_{l} |\tilde{b}^{\text{singlet}}_{2lS}|^2 + \sum_{l'<r,r'} |\tilde{b}^{\text{singlet}}_{l'r'S',2lS}|^2 + \cdots,$$

$$\sigma_{ab} = \tilde{b}_0 \tilde{b}^{\text{singlet}*}_{12S} + \sum_{l,r} \tilde{b}^{\text{singlet}}_{lrS} \tilde{b}^{\text{singlet}*}_{lrS,12S'} + \cdots,$$

$$\sigma_{ba} = \sigma^*_{ab}.$$

The matrix density elements are directly related to the coefficients $\tilde{b}(E)$ by a Laplace transform

$$\sigma^{(n)}_{\alpha\beta} = \sum_{l,\ldots,r,\ldots} \int \frac{dE dE'}{4\pi^2} \tilde{b}_{l,\ldots,r,\ldots}(E) \tilde{b}^*_{l,\ldots,r,\ldots}(E'), \quad (14)$$

where α/β specify the charging states associated with the amplitudes (b's). The equations for $n=0$ can be obtained straightforwardly. For instance, to get $\sigma^{(0)}_{aa}$, Eq. (12a) is multiplied by $\tilde{b}^*_0(E')$ and the conjugate equation written for E' is subtracted:

$$\dot{\sigma}^{(0)}_{aa} = 2\sqrt{2} i \gamma_A T (e^{-i\phi_S} \sigma^{(0)}_{ab} - e^{i\phi_S} \sigma^{(0)}_{ba}) \quad (15a)$$

$$\dot{\sigma}^{(0)}_{bb} = -(\Gamma_L + \Gamma_R)\sigma^{(0)}_{bb} - 2\sqrt{2} i \gamma_A T (e^{-i\phi_S} \sigma^{(0)}_{ab} - e^{i\phi_S} \sigma^{(0)}_{ba}) \quad (15b)$$

$$\dot{\sigma}^{(0)}_{cc} = -\Gamma_L \sigma^{(0)}_{cc} + \Gamma_R \sigma^{(0)}_{bb} \quad (15c)$$

$$\dot{\sigma}^{(0)}_{dd} = -\Gamma_R \sigma^{(0)}_{dd} + \Gamma_L \sigma^{(0)}_{bb} \quad (15d)$$

$$\dot{\sigma}^{(1)}_{aa} = 2\sqrt{2} i \gamma_A T (e^{-i\phi_S} \sigma^{(1)}_{ab} - e^{i\phi_S} \sigma^{(1)}_{ba}) + \Gamma_L \sigma^{(0)}_{dd} + \Gamma_R \sigma^{(0)}_{cc}, \ldots . \quad (15e)$$

Note that the diagonal matrix elements (the "populations") are coupled with the off-diagonal density-matrix elements ("coherences"), which is symptomatic of a coherent, reversible transition.

To obtain the equations for the coherence one subtracts Eq. (12a) for E multiplied by $\tilde{b}^*_{12s,\text{singlet}}(E')$ and Eq. (19) for E' multiplied by $\tilde{b}^*_0(E)$:

$$\dot{\sigma}^{(0)}_{ab} = -\frac{1}{2}(\Gamma_L + \Gamma_R)\sigma^{(0)}_{ab} + i[E_1 + E_2 + K(T_1 + T_2)]\sigma^{(0)}_{ab} + 2\sqrt{2} i \gamma_A T e^{i\phi_S} (\sigma^{(0)}_{aa} - \sigma^{(0)}_{bb}), \quad (16)$$

where $K = c' - 2c$.

These equations describe the sequential evolution of the system and involve consequently only processes between real states. Coherent processes (not involving reservoirs) couple nondiagonal elements to diagonal elements while relaxation processes couple only diagonal elements. From the set of Eq. (15), one can see that these processes do not interfere because of the loss of phase coherence introduced by the markoffian approximation, i.e., the sum over reservoir states. A density matrix element for one particular state is then only coupled to the elements for adjacent states in the sequence. Thus the processes can be added easily, which will be crucial when considering the full operation including all channels.

Here, because only one current channel is implied in the ideal operation, we can easily verify that the equations are the same for each n. Therefore the sum over n is obvious and one obtains the master equations for the evolution of the density matrix describing the system:

$$\dot{\sigma}_{aa} = 2\sqrt{2} i \gamma_A T (e^{-i\phi_S} \sigma_{ab} - e^{i\phi_S} \sigma_{ba}) + \Gamma_L \sigma_{cc} + \Gamma_R \sigma_{dd}, \quad (17a)$$

$$\dot{\sigma}_{bb} = -2\sqrt{2} i \gamma_A T (e^{-i\phi_S} \sigma_{ab} - e^{i\phi_S} \sigma_{ba}) - (\Gamma_L + \Gamma_R)\sigma_{bb}, \quad (17b)$$

$$\dot{\sigma}_{cc} = -\Gamma_L \sigma_{cc} + \Gamma_R \sigma_{bb}, \quad (17c)$$

$$\dot{\sigma}_{dd} = -\Gamma_R \sigma_{dd} + \Gamma_L \sigma_{bb}, \quad (17d)$$

$$\dot{\sigma}_{ab} = -\frac{1}{2}(\Gamma_L + \Gamma_R)\sigma_{ab} + i(E'_1 + E'_2)\sigma_{ab} + 2\sqrt{2} i \gamma_A T e^{i\phi_S} (\sigma_{aa} - \sigma_{bb}) \quad (17e)$$

with $E'_i = E_i + KT_i$.

This is the main result of this section. First, let us remark that the transition rates $\Gamma_{L,R}$ appear only from the dots to the reservoirs, and not in the opposite direction. This is consistent with the assumption that $k_B\Theta$ is small compared to the transition energies between dots and reservoirs. This limitation of Gurvitz's method is not a problem here since the entangler actually needs to be strongly biased to avoid decoherence effects. The second term of Eq. (17e) expresses that two discrete energy levels are coupled by a coherent process involving two transitions. Note that the probability of transmission between these two states is maximum in the resonant case, e.g., $\varepsilon = E'_1 + E'_2$ is zero.

The ideal operation of the system involves only one channel for transferring a Cooper pair to the reservoirs: the two electrons tunnel towards different leads. Actually, using the normalization condition for the populations $\sigma_{aa} + \sigma_{bb} + \sigma_{cc} + \sigma_{dd} = 1$, equations (17) are easily solved for the stationary current, $I = I(t \to \infty)$ ($\dot{\sigma}_{\alpha\beta} = 0$):

$$I^{\text{ent}}_L/e = \Gamma_L \sigma_{bb} + \Gamma_L \sigma_{cc} = \frac{\Gamma_L \Gamma_R}{\Gamma_L + \Gamma_R} \frac{8\gamma_A^2 T^2}{8\gamma_A^2 T^2 + \frac{\Gamma_L \Gamma_R}{4} + \varepsilon^2 \frac{\Gamma_L \Gamma_R}{(\Gamma_L + \Gamma_R)^2}}, \quad (18)$$

$$I^{\text{ent}}_R = e\Gamma_R \sigma_{bb} + e\Gamma_R \sigma_{dd} = I^{\text{ent}}_L. \quad (19)$$

This current is made of entangled singlet pairs. This result was obtained earlier in Ref. 8 in the limit $\gamma_A T \ll \Gamma$ and $\Gamma_L = \Gamma_R$. Here the presence of the term $8\gamma_A^2 T^2$ in the denomination comes from a complete (nonperturbative) treatment of both Andreev and decay processes.

The equality of the currents in the two branches of the device is a direct consequence of the crossed Andreev process. Every electron pair crosses and goes out of the

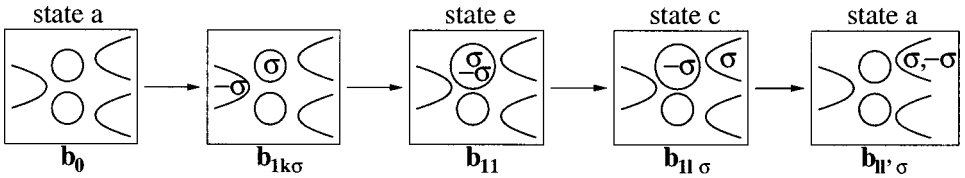

FIG. 3. A current channel sending a pair of electrons to the same reservoir. Andreev process towards one quantum dot can happen against strong Coulomb repulsion U.

system—each electron on its own side—before the next pair is injected in the dots. Those "cycles" never overlap in this ideal working regime.

In the case $\Gamma \gg \gamma_A T$, one obtains

$$I_L^{\mathrm{ent}}/e = \frac{8\gamma_A^2 T^2(\Gamma_L+\Gamma_R)}{\varepsilon^2+(\Gamma_L+\Gamma_R)^2/4} \tag{20}$$

while in the case $\Gamma \ll \gamma_A T$,

$$I_L^{\mathrm{ent}}/e = \frac{\Gamma_L\Gamma_R}{\Gamma_L+\Gamma_R} \tag{21}$$

similar to a single quantum dot between two leads.[17] In the latter situation, the dots are almost always occupied, so that the resistance is dominated by the rate associated with the two barriers—in parallel—between dots and leads.

IV. PARASITIC CHANNELS

The ideal working regime is affected by parasitic processes: Andreev tunneling via a single dot, one-by-one tunneling or S cotunneling. The two first ones have been separately computed by the T matrix in Ref. 8. Their effect is to create different channels of pair current which decrease the efficiency of entanglement. As said before, the terms for each process can be added in the equations and combined before including them together in a whole system of quantum master equations collecting every possible processes (see Sec. V). To start with, the different processes will be separately considered.

A. Direct Andreev effect process against Coulomb blockade

Let us imagine that a Cooper pair tunnels to the same quantum dot by an Andreev process, while generating a doubly occupied state. Because of Coulomb repulsion, an energy U [Eq. (2)] is required for having two electrons in a same quantum dot (see Fig. 3). If U is "large enough," such a process will have a low probability. With conventional dot technology, the interaction energy $U \sim 1$ K in the quantum dots can be controlled so that it is smaller than the superconducting gap $\Delta_S > 2$ K. Therefore the doubly occupied energy level has no coupling to the continuum of S quasiparticles, which would effectively introduce a broadening.

Similarly to the case of the ideal working regime (Sec. III), the set of differential equations associated with this direct Andreev channel are established for the reduced density matrix elements. Here, only one branch—say L—is considered for simplicity. The Fock space of the quantum dots consists here of three possible charge states: $|a\rangle$ both dots are empty, $|e\rangle$ dot 1 is doubly occupied, $|c\rangle$ dot 1 is singly occupied. The wave function takes the following form:

$$|\Psi(t)\rangle = \Bigg[b_0(t) + \sum_{k,\sigma} b_{1k,\sigma}(t) d^\dagger_{1\sigma}\gamma^\dagger_{-k-\sigma}S + \sum_{\sigma} b_{11}(t) d^\dagger_{1\sigma}d^\dagger_{1-\sigma}S + \sum_{l,\sigma} b_{1lS,\sigma}(t) a^\dagger_{l\sigma}d^\dagger_{1-\sigma}S + \sum_{l<l',\sigma} b_{ll'S,\sigma}(t) a^\dagger_{l\sigma}a^\dagger_{l'-\sigma}S$$
$$+ \sum_{l<l',\sigma,k',\sigma'} b_{ll'S\sigma,1k'\sigma'}(t) d^\dagger_{1\sigma'}\gamma^\dagger_{-k'-\sigma'}a^\dagger_{l\sigma}a^\dagger_{l'-\sigma}SS' + \cdots + \cdots + \cdots \Bigg]|0\rangle. \tag{22}$$

From the Schrödinger equation, and performing steps similar to Sec. III, the set of equations for the density matrix elements is

$$\dot\sigma_{aa} = 2iT_1(e^{-i\phi_S}\sigma_{ae} - e^{i\phi_S}\sigma_{ea}) + \Gamma_L\sigma_{cc}, \tag{23a}$$

$$\dot\sigma_{ee} = -2iT_1(e^{-i\phi_S}\sigma_{ae} - e^{i\phi_S}\sigma_{ea}) - 2\Gamma'_L\sigma_{ee}, \tag{23b}$$

$$\dot\sigma_{cc} = -\Gamma_L\sigma_{cc} + 2\Gamma'_L\sigma_{ee}, \tag{23c}$$

$$\dot\sigma_{ae} = i[U_1 + K'T_1]\sigma_{ae} + 2iT_1e^{i\phi_S}(\sigma_{aa} - \sigma_{ee}) - \Gamma'_L\sigma_{ae} \tag{23d}$$

with K' a numerical constant, and $\Gamma'_L = 2\pi\rho_L(U_1+E_1)|\Omega_L(U_1+E_1)|^2$ the level broadening introduced by coupling of the two-electrons level with lead L. These equations are similar to Eqs. (17). Nevertheless, the sequence passes through a high energy-level (U) via an Andreev process which implies an oscillation between two discrete energy levels, μ_S and $U'_1 = U_1 + K'T_1$. On the contrary, in the ideal regime, this energy difference can be as small as desired.

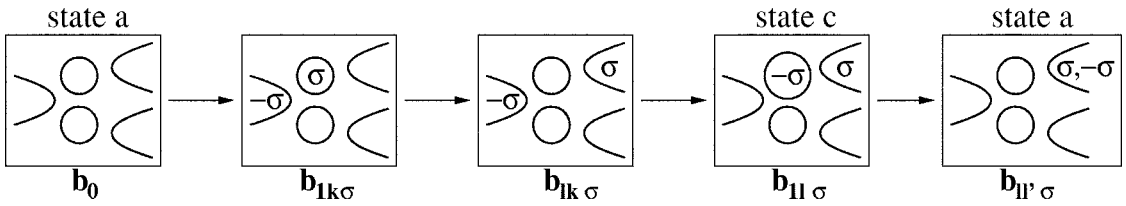

FIG. 4. Sequence corresponding to the tunneling of a singlet pair through one branch of the device. States $|a\rangle$ and $|c\rangle$ are coupled through two successive virtual states.

B. One-by-one electron tunneling to the reservoir

This channel is another way to send a pair into one single lead. Before the second electron of a broken Cooper pair can tunnel to a dot, the first one already leaves the dot to the corresponding lead. The second electron will tunnel through the same dot as its twin electron with a much higher probability (Fig. 4) than through the other dot, because of the geometrical factor. The latter process will be simply neglected.

There are only two processes involved in this channel. The first one, between states $|a\rangle$ and $|c\rangle$, requires two consecutive virtual states, both containing a quasiparticle in S. Because of the coupling with a continuum of states in the lead, phase coherence is lost thus off-diagonal matrix elements—or coherences—are not coupled to populations. Therefore this channel is peculiar in the sense that it is incoherent even though it involves transitions with S. The equations describing the evolution of the density matrix are obtained as before. The Schrödinger equation gives

$$(E + i\eta)\tilde{b}_0 = i + \sum_{k,\sigma} \sigma v_k \Omega_{k1}^* \tilde{b}_{1k\sigma}, \tag{24a}$$

$$(E + i\eta - E_1 - E_k)\tilde{b}_{1k\sigma} = \sigma v_k^* \Omega_{k1} \tilde{b}_0 + \sum_l \Omega_l^* \tilde{b}_{lk\sigma}, \tag{24b}$$

$$(E + i\eta - E_l - E_k)\tilde{b}_{lk\sigma} = \Omega_l \tilde{b}_{1k\sigma} + u_k \Omega_{k1}^* \tilde{b}_{1l\sigma}, \tag{24c}$$

$$(E + i\eta - E_1 - E_l)\tilde{b}_{1l\sigma} = \sum_k u_k^* \Omega_{k1} \tilde{b}_{lk\sigma} + \sum_{l'} \sigma \Omega_{l'} \tilde{b}_{ll'\sigma}, \tag{24d}$$

$$(E + i\eta - E_l - E_{l'})\tilde{b}_{ll'\sigma} = \Omega_{l'} \tilde{b}_{1l\sigma} - \Omega_l \tilde{b}_{1l'-\sigma} + \sum_{k,\sigma'} \sigma' v_k \Omega_{k1}^* \tilde{b}_{ll'\sigma,1k\sigma'}. \tag{24e}$$

Let us eliminate $\tilde{b}_{lk\sigma}$. To simplify, the notation $\Delta_{ij} = E + i\eta - E_i - E_j$ is introduced:

$$(E + i\eta - 2c\Omega_1^2)\tilde{b}_0 = i + \sum_{k,l,\sigma} \frac{\sigma u_k v_k |\Omega_{k1}|^2 \Omega_l}{\Delta_{lk}(\Delta_{1k} + i\Gamma_L/2)} \tilde{b}_{1l\sigma}, \tag{25}$$

$$\left(\Delta_{1l} - c''\Omega_l^2 + i\frac{\Gamma_L}{2}\right)\tilde{b}_{1l\sigma} = \sum_k \frac{\sigma u_k^* v_k^* |\Omega_{k1}|^2 \Omega_l}{\Delta_{lk}\left(\Delta_{1k} + i\dfrac{\Gamma_L(\Delta)}{2}\right)} \tilde{b}_0 + \sum_{kl'} \frac{|u_k|^2 |\Omega_{k1}|^2 \Omega_{l'}}{\Delta_{lk}\Delta_{l'k}\left(\Delta_{1k} + i\dfrac{\Gamma_L(\Delta)}{2}\right)} \tilde{b}_{1l'\sigma}. \tag{26}$$

Finally, using integrals calculated in Appendix A 5, Eq. (A14), the following set of equations is obtained:

$$\left[E + i\eta - 2c\Omega_1^2 + i\left(\frac{2T_1}{\pi\Delta_S}\right)^2 \Gamma_L\right]\tilde{b}_0 = i, \tag{27}$$

$$\left(\Delta_{1l} - c''\Omega_1^2 + i\frac{\Gamma_L}{2}\right)\tilde{b}_{1l\sigma} = \sigma\frac{2T_1}{\pi\Delta_S} e^{-i\phi_S} \Omega_l \tilde{b}_0 + \sigma\frac{3T_1}{2\pi\Delta_S}\Omega_1^2 \sum_{l'} \Omega_{l'} \tilde{b}_{1l'\sigma}, \tag{28}$$

$$\left[\Delta_{ll'} - 2c\Omega_1^2 + i\left(\frac{2T_1}{\pi\Delta_S}\right)^2 \Gamma_L\right]\tilde{b}_{ll'\sigma} = \Omega_{l'} \tilde{b}_{1l\sigma} - \Omega_l \tilde{b}_{1l'\sigma}. \tag{29}$$

Virtual states have disappeared from the equations. The remaining term in $T_1\Omega_l$ corresponds to the three-step process coupling $|a\rangle - |c\rangle$. Introducing the elements of the density matrix one gets

$$\dot{\sigma}_{aa} = -K''\frac{T_1^2}{\Delta_S^2}\Gamma_L \sigma_{aa} + \Gamma_L \sigma_{cc}, \tag{30a}$$

$$\dot{\sigma}_{cc} = -\Gamma_L \sigma_{cc} + K''\frac{T_1^2}{\Delta_S^2}\Gamma_L \sigma_{aa}, \tag{30b}$$

where $K'' = 4/\pi^2$. This process behaves as the transport through a single dot where the first barrier between the left lead and the dot is a three-step process via two virtual states and the second barrier is a classic tunnel barrier.

C. S cotunneling between the two quantum dots

Another process involves intermediate virtual states of the quantum device which are common to the other processes: cotunneling[12,21] between the two quantum dots via S. This

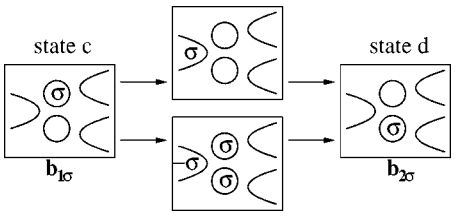

FIG. 5. Cotunneling between the two dots. An electron from dot 1 tunnels towards the dot 2 via a virtual intermediate state containing a quasiparticle. Two contributions participate to the cotunneling depending on when the initial electron is transferred.

process involves oscillations between two position states and connects all of the channels studied until now.

It can occur in different situations: between states containing only one or two electrons in the two dots. As for the crossed Andreev process, the transmission probability depends on the energy difference between the two coupled states. The equation of evolution for the density matrix describing oscillations between two states—$|c\rangle$ (electron on dot 1) and $|d\rangle$ (electron on dot 2) (see Fig. 5)–are established:

$$\dot{\sigma}_{cc} = i\gamma_C T(\sigma_{cd} - \sigma_{dc}), \tag{31a}$$

$$\dot{\sigma}_{dd} = i\gamma_C T(\sigma_{dc} - \sigma_{cd}), \tag{31b}$$

$$\dot{\sigma}_{cd} = i\Delta E\sigma_{cd} + i\gamma_C T(\sigma_{cc} - \sigma_{dd}), \tag{31c}$$

where $\Delta E = E_2 - E_1$, $\gamma_C = e^{-r/\pi\xi_0}[\cos(k_F r)/k_F r]$ is the geometrical factor corresponding to this cotunneling process.[12] Note that when the distance r which separates the two tunneling locations is zero, γ_C diverges. This is expected because this process has no meaning for the same tunneling location: this local process brings back the system in the same state, it only participates to the renormalization of the energy level of the state by coupling with the continuum of quasiparticles in S. Note that the transition amplitude T is the same as for Andreev process.

V. ENTANGLER IN THE PRESENCE OF PARASITIC PROCESSES

One of the advantage of Bloch-type equations is to be able to study all processes together and nonperturbatively. In the previous sections, a specific system of dynamical equations was obtained separately for different channels of pair current. In particular, such channels are repeated cycle after cycle, which allows to systematically group the contributions with different reservoir variables (by recurrence over the number of pairs transmitted to the leads).

In reality, each channel (induced by crossed Andreev, direct Andreev, S cotunneling) mixes into one another, so one needs to gather all transitions in a single set of equations for the density matrix (see Fig. 6). Because of this mixing, it is no more possible to establish a set of equations cycle after cycle.

A starting point for deriving generalized quantum master equations is thus to label the amplitude associated with each process by variables which count how many entangled pair have passed through reservoir R or L or both (while being split). Note that such variables do not appear in the quantum master equation of each channel because they have been summed over. It is straightforward, but tedious, to write a full Schrödinger equation for the most general operation, combining all states, and to derive the density matrix equations. The basic assumption is that not more than one quasiparticle is excited in the superconductor during the processes.

As was said in Sec. III, all the processes can be gathered without appealing to the full derivation of the Schrödinger equation, by adding terms corresponding to each process. We set the equations for a given state of reservoirs were n_L (n_R)

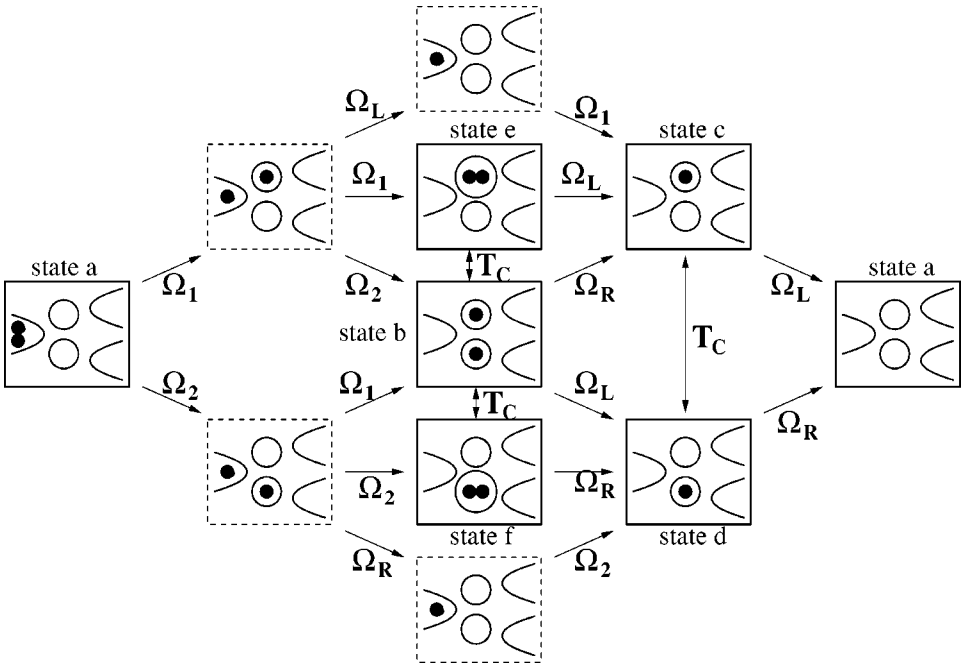

FIG. 6. General operation including the three Andreev channels and S cotunneling. States with three or four electron states are omitted for clarity. Real states are fully squared while virtual states are dashed squared. To make it simpler, spin is not represented. The Ω's correspond to transitions between two quantum states $|S\rangle \otimes |\text{dots}\rangle \otimes |l,r\rangle$, while T_C indicates the resonant cotunneling process. Certain mixing process, such as the direct Andreev effect between states $|c\rangle$ and state with one electron in dot 1 and two electrons in dot 2, are not presented for lack of space. However, such processes are included in the quantum master equations.

singlet pairs of electrons have tunneled to the reservoir L (R) and n_0 pairs whose electrons have tunneled to different leads. States are thus defined by the charge of quantum dots, one electron already in the reservoir while its twin electron is still in the quantum system, and n_L, n_R, and n_0: $|\psi\rangle = |\text{dot1}, \text{dot2}\rangle \otimes |ll'rr'\rangle \otimes |n_L, n_R, n_0\rangle$. To get equations for only the charge states of the dots, they are summed over l, l', r, r' and the recurrence is made over n_L, n_R, n_0. The obtained set does not depend on the number of channels and leads. Thus Gurvitz's method for generating quantum master equations[18] can be generalized to the multiterminal case with many current channels. The full system is given in Appendix B. One can notice that the parasitic processes may generate triplet pairs in the leads L, R.

VI. DISCUSSION

The set of quantum master equations will now be used to describe more quantitatively the transport properties. To assess the constraints on parameters, each channel will first be studied, before using the complete set to obtain a numerical evaluation of the operation in a realistic regime.

By solving quantum master equations one can find the average current for each uncoupled channel. This will be done for the symmetric case ($\Gamma_L = \Gamma_R$, $U_1 = U_2$, and $T_1 = T_2$) and assuming that $\Gamma_{L,R} = \Gamma'_{L,R} = \Gamma$ when the coupling between quantum dots and lead depends weakly on the energy: The direct-Andreev current is computed in the stationary regime with Eqs. (23):

$$I_L^{\text{Andreev}}/e = 2\Gamma'_L \sigma_{bb} + \Gamma_L \sigma_{cc} = \Gamma \frac{16T^2}{16T^2 + \Gamma^2 + U'^2}. \quad (32)$$

With $U' \gg 2T$, Γ we have, as in Ref. 8

$$I_L^{\text{Andreev}} \approx e\Gamma \frac{16T^2}{U'^2} \quad (33)$$

while with $\gamma_A^2 T^2 \gg \varepsilon^2$ Eq. (18) can be written

$$I_L^{C\ \text{Andreev}} \approx e\Gamma \frac{8\gamma_A^2 T^2}{8\gamma_A^2 T^2 + \Gamma^2/4}. \quad (34)$$

The current created by the one-by-one tunneling process is given by

$$I_L^{sb} = e\frac{K'' T^2 \Gamma}{\Delta_S^2 + K'' T^2} \approx 4e\Gamma \frac{T^2}{\pi^2 \Delta_S^2}. \quad (35)$$

Without taking here into account elastic cotunneling, one can see here the relationship between parameters that must be fulfilled to approach the ideal working of the entangler: $U', \Delta_S \gg \max[T, \Gamma/\gamma_A, \varepsilon/\gamma_A]$. This can be understood with a dynamical study of each channel. Actually Andreev processes are coherent processes which create an oscillation between the state where the Cooper pair is in S and states where the pair of electrons is in the dots. Thus it will be a competition between the period and the amplitude of oscillations and the probability of tunneling from a dot to a reservoir. Let us first consider the case where $\gamma_A T \gg \Gamma$. Then for resonant crossed-Andreev process

$$\sigma_{bb}(t) = \frac{1}{2}\left(1 - \frac{\Gamma^2}{2T^2}\right)[1 - \cos(2\gamma_A T t)]e^{-\Gamma t} \quad (36)$$

while for the direct Andreev process

$$\sigma_{ee}(t) = \frac{2T^2}{U'^2 + 4T^2}[1 - \cos(\sqrt{U'^2 + 4T^2}\,t)]e^{-\Gamma t}. \quad (37)$$

Because Γ is small we are here in the regime where the crossed Andreev channel is more probable than the direct Andreev one because many oscillations between coherent states can occur before a transition to a reservoir has happened. In the other limit ($\gamma_A T \ll \Gamma$), one gets

$$\sigma_{bb}(t) = \frac{\gamma_A^2 T^2}{\Gamma^2}(e^{-2\gamma_A^2 T^2/\Gamma t} + e^{-2\Gamma t} - 2e^{-\Gamma t}), \quad (38)$$

$$\sigma_{ee}(t) = \frac{T^2}{U'^2 + \Gamma^2}[e^{-\Gamma T^2/(U'^2+\Gamma^2)t} + e^{-2\Gamma t} - 2\cos(Ut)e^{-\Gamma t}]. \quad (39)$$

As soon as the pair has tunneled to the dots, it goes to the reservoirs. And because the direct Andreev frequency is larger than the crossed Andreev one ($U \gg \gamma_A T$), there is a small time interval in which direct Andreev is favored even though the amplitude of oscillation (and thus tunneling between $|a\rangle$ and $|e\rangle$) is smaller: for a relaxation time $1/\Gamma$ of the order of half a period of oscillation for direct Andreev effect (π/U), after a time $t \sim 1/\Gamma$, the population of state $|e\rangle$ can be much larger than population of state $|b\rangle$.

The same kind of argument can be given to study the effect of S cotunneling. As said before, for U and Δ_S large enough, the only parasitic effect is elastic cotunneling. Using only this process and crossed Andreev process in the master equation, the efficiency of entanglement is calculated depending on $E = |E_1 - E_2|$ which controls S-cotunneling probability. We want to know the proportion of electrons from a same pair tunneling to different reservoirs ($P_{\text{entangled}}$) or to the same reservoir ($P_{\text{parasitic}}$). Cycles of current do not overlap so the probability is the same for each cycle. To calculate them, we can use Bloch equations describing the evolution on only one cycle to get first $|c\rangle$ and $|d\rangle$ populations as a function of time. From state $|b\rangle$ the first electron tunnels for example towards the left reservoir. The chance for the second electron to tunnel towards the right (left) reservoir is $\Gamma_R p_d(t)$ $[\Gamma_L p_c(t)]$ assuming that $p_d(0) = 1$. Thus $P_{\text{entangled}} = \int_0^\infty \Gamma_R p_d(t) dt$. For $\Gamma_L = \Gamma_R$:

$$P_{\text{entangled}} = \frac{\Gamma^2 + E^2 + 2\gamma_c^2 T^2}{\Gamma^2 + E^2 + 4\gamma_c^2 T^2}. \quad (40)$$

From Eq. (40), we can see that the condition to neglect S cotunneling, leading to $P_{\text{entangled}} \sim 1$, is $\gamma_c T \ll \max[E, \Gamma]$.

A more general study using the complete set of equations (see Appendix B) has to be performed. This set of equations can be solved in the stationary regime, but the general solution is typically cumbersome. For the sake of readability, it is presented here taking into account the parasitic processes only to first order. This fixes the different energy scales, previously discussed above, which define the working regime of

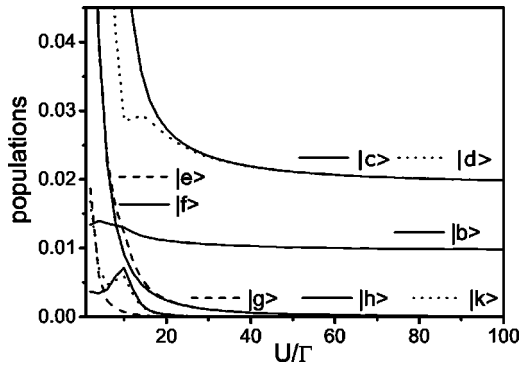

FIG. 7. Charge states populations as a function of U for $\Delta_S = 9.5$ K, $E_1 = -E_2 = 0.5$ K, $\Gamma_{L,R} = \Gamma'_{L,R} = T = 0.1$ K, γ_A, $\gamma_C \sim 0.2$. States $|a\rangle$, $|b\rangle$, $|c\rangle$, $|d\rangle$, $|e\rangle$, $|f\rangle$ refer to Fig. 6. State $|k\rangle$ refers to the triplet state shared between dots, and states $|g\rangle$ and $|h\rangle$ refer to three electrons states (see Appendix B). $|g\rangle$, $|h\rangle$, and $|k\rangle$ populations correspond to the three lowest curves. The population of states containing doubly occupied dots vanishes when U increases. For low values of U ($U \sim |E_1 - E_2|$), the asymmetry is introduced by energy difference between states $|e\rangle$ (two electrons in dot 1) and $|f\rangle$ (two electrons in dot 2).

the entangler. Here the asymmetry $\Gamma_L \neq \Gamma_R$ is kept to show the role of S cotunneling:

$$I_L = e\Gamma_L \sigma_{bb} + e\Gamma_L \sigma_{ee} + 2e\Gamma'_L \sigma_{cc}, \tag{41}$$

$$\begin{aligned} I_L = e\sigma_0 \Bigg\{ & \Gamma_L + \Gamma_R + 4\Gamma^2 \left(\frac{1}{\Gamma_L} - \frac{1}{\Gamma_R} \right) \frac{\gamma_C^2 T^2}{\Delta E^2} \\ & - 2K'' A \sigma_0 \Gamma_L \frac{T^2}{\Delta_S^2} \left(1 - \frac{1}{2\sigma_0} \right) \\ & - 8A\sigma_0 \Gamma_L \frac{T^2}{U^2} \left[2 + \frac{\Gamma'_L}{\Gamma_L} \left(1 - \frac{1}{\sigma_0} \right) + \frac{2\Gamma'_R}{\Gamma_R} \right] \\ & - 2\sigma_0 \Gamma_L \frac{\gamma_C^2 T^2}{U^2} \left[2 + \frac{\Gamma'_L}{\Gamma_L} \left(1 - \frac{1}{\sigma_0} \right) + \frac{2\Gamma'_R}{\Gamma_R} \right. \\ & \left. + \frac{2\Gamma\Gamma'_L}{\Gamma^2 + 8\gamma_A^2 T_A^2 + \varepsilon^2} \right] \Bigg\}, \end{aligned} \tag{42}$$

where $A = 8\gamma_A^2 T_A^2 / (\Gamma^2 + 8\gamma_A^2 T_A^2 + \varepsilon^2)$ and $\sigma_0^{-1} = A + 1 + \Gamma_R/\Gamma_L + \Gamma_L/\Gamma_R$. From Eq. (42) we can exhibit which parameters are controlling each contribution to the total current.

To complete this discussion, the set of equations is used to describe the average populations of each state depending on some relevant parameters. Δ_S is taken to be the largest energy scale. With niobium as superconductor, one takes $\Delta_S \sim 9.25$ K. For a two-dimensional quantum dot, small enough (10 nm^2), one takes $|E_i| \sim 0.5$ K and $U \sim 9$ K, with T, $\Gamma \sim 0.1$ K (Ref. 26) and γ_A, $\gamma_C \sim 0.2$.

On Fig. 7, it can be seen that the population of states containing doubly occupied dots vanishes when U increases. It is important to notice that when $U \sim |E_i|$, the system is asymmetric and the channel with $U \sim |E_1 - E_2|$ is favored because a direct Andreev process becomes resonant. At the working point ($U/T = 90$) $p_e/p_b = 0.012$. Two channels can be compared in calculating the ratio between two populations: on Fig. 8 the ratio between the population of state $|b\rangle$ and the one of state $|e\rangle$ indicates which of the crossed Andreev and direct Andreev processes is the most likely depending on U. Thus increasing U increases the efficiency of entanglement. For small $U \sim \Gamma$, the two channels become comparable because decays to reservoirs are much faster than crossed-Andreev oscillations.

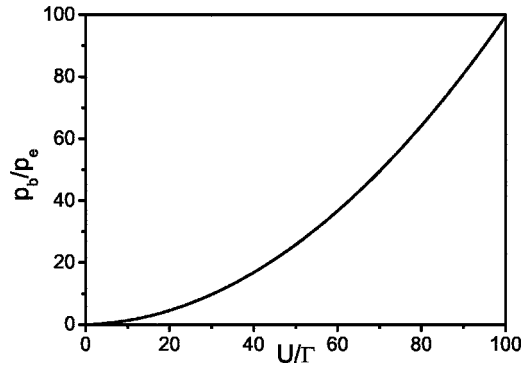

FIG. 8. Ratio between populations of state $|b\rangle$ (singlet state shared between dots 1 and 2) and of state $|e\rangle$ population (two electrons in dot 1) for $\Delta_S = 9.5$ K, $E_1 = -E_2 = 0.5$ K, $\Gamma_{L,R} = \Gamma'_{L,R} = T = 0.1$ K, γ_A, $\gamma_C \sim 0.2$. It indicates the ratio between direct Andreev channel and crossed Andreev channel. The latter is strongly favored when U increases [$p_b/p_e(U/\Gamma = 90) = 83.3$].

A large Γ will allow a fast transition between dots and reservoirs. That is why increasing Γ/T will favor the most likely process which connects the superconductor to the dots.[8] On Fig. 9 we can see that increasing Γ favors the decay of a single charge state before another pair tunnels towards the free quantum dot. Actually, because direct Andreev oscillations are faster (frequency $\sim \sqrt{E^2 + 4T^2}$) than crossed Andreev oscillations (frequency $\sim \gamma_a T$), even if their probability is smaller, the decay towards reservoirs can happen before one crossed Andreev process has been achieved.

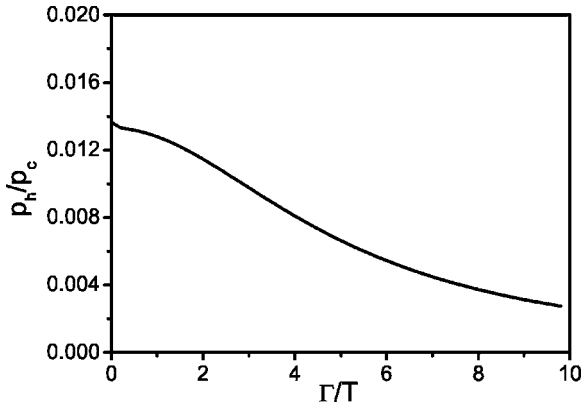

FIG. 9. Ratio between populations of state $|h\rangle$ (one electron in dot 1, two electrons in dot 2) and of state $|c\rangle$ (one electron in dot 1) for $\Delta_S = 9.5$ K, $E_1 = -E_2 = 0.05$ K, $\Gamma_{L,R} = \Gamma'_{L,R}$, $T = 0.1$ K, $U = 1$ K, γ_A, $\gamma_C \sim 0.2$. Increasing Γ compared to the transition rate of direct Andreev and crossed Andreev processes allows us to favor the decay of single charge states before another Cooper pair tunnels to the free quantum dot. For $U = 1$ K $= 10$ T, $p_h/p_e < 1.5\%$.

Thus increasing Γ/T at fixed U, direct Andreev process increase to the detriment of the crossed Andreev one.

VII. CONCLUSION

In this article, quantum master equations have been derived, starting from a microscopic Hamiltonian for the superconducting-dot entangler. Using the Schrödinger equation technique developed in Ref. 18, the full equations describing the evolution of the reduced density matrix are obtained, retaining as virtual states only single particle excitations in the superconductor. Considering only one level by dot, all possible processes are taken into account in a fully consistent and nonperturbative way: crossed Andreev process, responsible for entanglement, as well as direct Andreev and one-by-one tunneling processes, and cotunneling through the superconductor. The latter connects all the other processes, yet the quantum master equations written in Appendix B take into account all processes in a coherent way. From them, the average current has been calculated. The conditions on the entangler parameters, needed for an optimal operation of the device, have been derived, and extend the result of Ref. 8.

The power of master equations is to give access, not only to the first moment, but to all moments of the current distribution.[27] In a forthcoming paper,[28] shot noise correlations are computed in order to give a clear diagnosis of entanglement.[9,29] Another extension of Bloch equations is to include explicitly spin/charge relaxation or coupling to external degrees of freedom, in order to quantitatively study decoherence effects.

Such a derivation of quantum master equations, including higher order process, can obviously be generalized to a wide class of quantum systems involving discrete charge states and coherent/incoherent transitions. It is therefore a valuable tool for investigating nanostructures in view of controlling quantum information based on spin/charge degrees of freedom.

ACKNOWLEDGMENTS

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APPENDIX A: CALCULATION OF INTEGRALS

To obtain the evolution equation of the density matrix, it is necessary to compute some integrals arising from the coupling between S and the two dots.

1. Crossed-Andreev effect

The tunneling of the two electrons of a same Cooper pair to two different dots gives a contribution [see Eqs. (11a)–(11g)]:

$$I_A = \sum_k \frac{u_k v_k \Omega_{k1} \Omega_{-k2}}{E - E_i - E_k}. \tag{A1}$$

The two energy levels of the dots are assumed to be close to μ_S. The transitions amplitudes Ω depend weakly on the energy so they can be considered as constant with a phase factor $e^{i\vec{k}.\vec{r}}$. Neglecting $E-E_i \ll E_k \sim \Delta_S$ one obtains

$$\begin{aligned} I_A &= -\Omega_1\Omega_2 \frac{V}{(2\pi)^3}\int d^3\vec{k}\frac{\Delta}{2E_k^2}e^{i\vec{k}\cdot\vec{r}} \\ &= -\frac{\Omega_1\Omega_2}{2}\frac{V}{(2\pi)^3}\int_0^{2\pi} d\phi \int_0^{\pi} d\theta \int_0^{\infty} dk k^2 \sin\theta e^{ikr\cos\theta} \\ &\quad \times \frac{\Delta}{\Delta^2 + \left(\frac{\hbar^2k^2}{2m} - \mu\right)^2}. \end{aligned} \tag{A2}$$

with $\mu = \hbar^2 k_F^2/2m$ and V the volume.

Because of parity the integral can be extended from $-\infty$ to ∞.

$$I_A = \frac{\pi\Omega_1\Omega_2}{2ir}\frac{V}{(2\pi)^3}\int_{-\infty}^{\infty} dk k (e^{ikr} - e^{-ikr}) \frac{\Delta}{\Delta^2 + \left[\frac{\hbar^2k^2}{2m} - \frac{\hbar^2k_F^2}{2m}\right]^2}. \tag{A3}$$

The four poles are

$$k = \pm k_F \sqrt[4]{1 + \left(\frac{2m\Delta}{\hbar^2 k_F^2}\right)^2} e^{\pm i/2 \arctan(2m\Delta/\hbar^2 k_F^2)} \equiv \pm a k_F e^{\pm i\theta},$$

$k_1 = a k_F e^{i\theta}$, $k_2 = -a k_F e^{-i\theta}$, $k_3 = -a k_F e^{i\theta}$, $k_4 = a k_F e^{-i\theta}$. The contour is the positive half circle for e^{ikr} and the negative one for e^{-ikr}:

$$\begin{aligned} I_A = \frac{\pi\Omega_1\Omega_2}{2ir}\frac{V}{(2\pi)^3} 2i\pi\Delta\left(\frac{2m}{\hbar^2}\right)^2 \Bigg[& \frac{k_1 e^{ik_1 r}}{(k_1-k_2)(k_1-k_3)(k_1-k_4)} \\ & + \frac{k_2 e^{ik_2 r}}{(k_2-k_1)(k_2-k_3)(k_2-k_4)} - \frac{-k_3 e^{-ik_3 r}}{(k_3-k_1)(k_3-k_2)(k_3-k_4)} \\ & - \frac{-k_4 e^{-ik_4 r}}{(k_4-k_1)(k_4-k_2)(k_4-k_3)} \Bigg], \end{aligned}$$

$$\begin{aligned} I_A = \frac{\pi\Omega_1\Omega_2}{2ir}\frac{V}{(2\pi)^3}\Delta\left(\frac{2m}{\hbar^2}\right)^2 \frac{\pi e^{-ak_F r\sin\theta}}{2(ak_F)^2\sin(2\theta)} \\ \times [e^{iak_F r\cos\theta} - e^{-iak_F r\cos\theta}] \end{aligned} \tag{A4}$$

with $\sin 2\theta = 2m\Delta/(a^2\hbar^2 k_F^2)$, $\sin\theta = \Delta/2E_F$. Given that a and $\cos\theta \sim 1$ ($\Delta_S \ll E_F$), one obtains

$$I_A = \frac{\pi}{2} N(0)\Omega_1\Omega_2 e^{-(r/\pi\xi_0)}\frac{\sin(k_F r)}{k_F r}. \tag{A5}$$

In what follows, I_A is noted: $I_A = \gamma_A T$ with $T = (\pi/2)N(0)\Omega_1\Omega_2$ and $\gamma_A = e^{-r/\pi\xi_0}[\sin(k_F r)/k_F r]$, the geometrical factor for the crossed-Andreev effect.

2. Direct-Andreev effect

The tunneling of the two electron of a same Cooper pair to the same dot, i, gives a contribution

$$T_i = \sum_k \frac{u_k v_k \Omega_{ki} \Omega_{-ki}}{E - E_i - E_k}. \tag{A6}$$

From the previous calculation, one must take the limit $r \to 0$ in Eq. (A5). The same result is found when making the calculation without taking into account the phase factor e^{ikx} which generates the geometrical factor. The amplitude of this effect towards the i side is then $T_i = (\pi/2)N(0)\Omega_i^2$.

3. Self-energy

The self-energy terms are due to the coupling between a discrete state (state with zero or one electron in a dot) and a continuum of states (quasiparticle states in S). They correspond to the renormalization of these energy levels. They involve $|v_k|^2$ when the annihilation of an electron in S corresponds to the creation of quasiparticle, and $|u_k|^2$ when the creation of an electron corresponds to the creation of quasiparticle. In Eqs. (11a)–(11g), this term corresponds to

$$I_R = \sum_k \frac{|v_k|^2 |\Omega_{ki}|^2}{E - E_i - E_k}, \tag{A7}$$

$$J_R = \sum_k \frac{|u_k|^2 |\Omega_{ki}|^2}{E - E_i - E_k} \tag{A8}$$

for a given i side.

The sum are transformed into integrals over quasiparticle energies E_k with a density of states given by $N(E) = N(0)E/\sqrt{E^2 - \Delta_S^2}$. For the calculation, E_i/Δ_S is noted e_i and E/Δ_S is noted x.

a. Terms in $|v_k|^2$.

$$I_R = \frac{T_i}{2}\int_1^\infty \left(\frac{1}{\sqrt{x^2-1}} - \frac{1}{x}\right)\frac{x}{e_i - x}dx = -\frac{T_i}{2}\ln[2(1-e_i)] - T_i \frac{e_i}{\sqrt{e_i^2-1}}\left(\frac{\pi}{2} + \arcsin(e_i)\right) \approx -\frac{T_i}{2}\ln 2,$$

$$I_R = cT_i, \tag{A9}$$

where c is a numerical constant.

b. Terms in $|u_k|^2$. This term never appears alone, so we just have to calculate terms with $|u_k|^2 - |v_k|^2$:

$$J_R - I_R = \frac{T_i}{2}\int_1^\infty \frac{dx}{e_i - x} = -\frac{T_i}{2}[\ln|e_i - x|]_1^\infty,$$

$$J_R = c' T_i. \tag{A10}$$

To avoid the logarithmic divergence we introduce a physical cutoff—the electron band width—to get a finite result. This does not yield a large contribution because of the logarithm: if the band width is 1000 times higher than the gap it only gives a factor $8\Omega^2$ where $\Omega \ll E_i$. Self-energy terms remain small. Let us define $K = c' - 2c$ for the following.

4. *S* cotunneling

Local S cotunneling has no meaning (tunneling of an electron between two places) so keeping the geometrical contribution of the integrand in this process, one gets

$$I_C = \sum_k \frac{(|u_k|^2 - |v_k|^2)\Omega_{k1}\Omega_{-k2}}{E - E_i - E_k}. \tag{A11}$$

With $|u_k|^2 - |v_k|^2 = \xi_k/E_k$:

$$I_C = \frac{\pi \Omega_1 \Omega_2}{2ir}\frac{V}{(2\pi)^3}\int_{-\infty}^{\infty} dk\, k(e^{ikr} - e^{-ikr})\frac{\frac{\hbar^2 k^2}{2m} - \mu}{\Delta^2 + \left(\frac{\hbar^2 k^2}{2m} - \mu\right)^2}.$$

Using once again the residue theorem one gets

$$I_C = \frac{\pi \Omega_1 \Omega_2}{2ir}\frac{V}{(2\pi)^3}\left(\frac{2m}{\hbar^2}\right)^2 \frac{\pi k_F^2 e^{-ak_F r \sin\theta}}{(ak_F)^2 \sin(2\theta)} \times [e^{i(ak_F r\cos\theta + \theta)}(a^2 e^{i\theta} - e^{-i\theta}) - e^{-i(ak_F r\cos\theta + \theta)}(a^2 e^{-i\theta} - e^{i\theta})]. \tag{A12}$$

With $a^2 \sim 1 + 1/2(2m\Delta/\hbar^2 k_F^2)^2$:

$$I_C = \frac{\pi}{2}N(0)\Omega_1\Omega_2 e^{-r/\pi\xi_0}\left[\frac{\cos(k_F r)}{k_F r} + \frac{1}{2}\left(\frac{2m\Delta}{\hbar^2 k_F^2}\right)^2\left(\frac{\sin(k_F r)}{k_F r} - \frac{\cos(k_F r)}{k_F r}\right)\right]. \tag{A13}$$

The second term is much smaller than the first one ($\Delta_S \ll E_F$). The only difference with the Andreev amplitude is the $\cos(k_F r)/k_F r$ instead of $\sin(k_F r)/k_F r$. S cotunneling diverges for $r \to 0$.

5. One-by-one electron tunneling to the reservoir

Here the calculation is not complicated by a phase factor. The sum over k is simply replaced by an integral over energy:

$$\begin{aligned} I_P &= \sum_k \frac{u_k v_k |\Omega_{k1}|^2}{\Delta_{lk}(\Delta_{1k+i\Gamma_L/2})} \\ &\simeq \sum_k \frac{u_k v_k}{E_k^2}\Omega_1^2\Omega_l \\ &= N(0)\Omega_1^2\Omega_l \int_\Delta^\infty \frac{E}{\sqrt{E^2-\Delta^2}}\frac{\Delta}{E^3}dE \\ &= N(0)\frac{\Omega_1^2\Omega_l}{\Delta}\int_1^\infty \frac{dx}{x^2\sqrt{x^2-1}} \\ &= N(0)\frac{\Omega_1^2\Omega_l}{\Delta}. \end{aligned} \tag{A14}$$

APPENDIX B: QUANTUM MASTER EQUATIONS FOR THE ENTANGLER

The set of fully consistent and nonperturbative quantum master equations can be derived (see main part of the paper). For simplicity, the space of charge states has been restricted here to 0, 1, 2, or 3 electrons in the two dots. Numerical calculations have been made with this set of equations including states $|g\rangle$ (one electron in dot 2, two electrons in dot 1), $|h\rangle$ (one electron in dot 1, two electrons in dot 2), and $|k\rangle$ (triplet state shared between dots 1 and 2):

$$\dot{\sigma}_{aa} = +2iT_1(\sigma_{ae}-\sigma_{ea}) + 2iT_2(\sigma_{af}-\sigma_{fa}) \times 2\sqrt{2}i\gamma_A T(\sigma_{ab}-\sigma_{ba}) + \Gamma_L\sigma_{cc} + \Gamma_R\sigma_{dd} - 2(\tilde{\Gamma}_L+\tilde{\Gamma}_R)\sigma_{aa}, \tag{B1}$$

$$\dot{\sigma}_{bb} = +i\sqrt{2}\gamma_C T(\sigma_{be}-\sigma_{eb}) + i\sqrt{2}\gamma_C T(\sigma_{bf}-\sigma_{fb}) - 2\sqrt{2}i\gamma_A T(\sigma_{ab}-\sigma_{ba}) + \frac{1}{2}\tilde{\Gamma}_R\sigma_{cc} + \frac{1}{2}\tilde{\Gamma}_L\sigma_{dd} + \frac{1}{2}\Gamma'_L\sigma_{gg} + \frac{1}{2}\Gamma'_R\sigma_{hh} - 2(\Gamma_L+\Gamma_R)\sigma_{bb}, \tag{B2}$$

$$\dot{\sigma}_{cc} = i\gamma_C T(\sigma_{cd}-\sigma_{dc}) + 2iT_2(\sigma_{ch}-\sigma_{hc}) + 2\tilde{\Gamma}_L\sigma_{aa} + 2\Gamma_R\sigma_{bb} + \Gamma_R\sigma_{kk} + 2\Gamma'_L\sigma_{ee} - (\Gamma_L+2\tilde{\Gamma}_R)\sigma_{cc}, \tag{B3}$$

$$\dot{\sigma}_{dd} = -i\gamma_C T(\sigma_{cd}-\sigma_{dc}) + 2iT_1(\sigma_{dg}-\sigma_{gd}) + 2\tilde{\Gamma}_R\sigma_{aa} + 2\Gamma_L\sigma_{bb} + \Gamma_L\sigma_{kk} + 2\Gamma'_R\sigma_{ff} - (\Gamma_R+2\tilde{\Gamma}_L)\sigma_{dd}, \tag{B4}$$

$$\dot{\sigma}_{ee} = -2iT_1(\sigma_{ae}-\sigma_{ea}) - i\sqrt{2}\gamma_C T(\sigma_{be}-\sigma_{eb}) + \Gamma_R\sigma_{gg} - 2\Gamma'_L\sigma_{ee}, \tag{B5}$$

$$\dot{\sigma}_{ff} = -2iT_2(\sigma_{af}-\sigma_{fa}) - i\sqrt{2}\gamma_C T(\sigma_{bf}-\sigma_{fb}) + \Gamma_L\sigma_{hh} - 2\Gamma'_R\sigma_{ff}, \tag{B6}$$

$$\dot{\sigma}_{gg} = -2iT_1(\sigma_{dg}-\sigma_{gd}) - (2\Gamma'_L+\Gamma_R)\sigma_{gg}, \tag{B7}$$

$$\dot{\sigma}_{hh} = -2iT_2(\sigma_{ch}-\sigma_{hc}) - (2\Gamma'_R+\Gamma_L)\sigma_{hh}, \tag{B8}$$

$$\dot{\sigma}_{kk} = \frac{3}{2}(\tilde{\Gamma}_R\sigma_{cc} + \tilde{\Gamma}_L\sigma_{dd} + \Gamma'_L\sigma_{gg} + \Gamma'_R\sigma_{hh}) - (\Gamma_L+\Gamma_R)\sigma_{kk}, \tag{B9}$$

$$\dot{\sigma}_{ab} = i(E'_1+E'_2)\sigma_{ab} + 2\sqrt{2}i\gamma_A T(\sigma_{aa}-\sigma_{bb}) + i\sqrt{2}\gamma_C T(\sigma_{ae}+\sigma_{af}) - (iT_1\sigma_{eb}+iT_2\sigma_{fb}) - \frac{1}{2}(2\tilde{\Gamma}_L+2\tilde{\Gamma}_R+\Gamma_L+\Gamma_R)\sigma_{ab}, \tag{B10}$$

$$\dot{\sigma}_{ae} = i(E'_1+U'_{11})\sigma_{ae} + 2iT_1(\sigma_{aa}-\sigma_{ee}) + i(\sqrt{2}\gamma_C T\sigma_{ab} - 2\sqrt{2}\gamma_A T\sigma_{be} - 2T_2\sigma_{fe}) - (\tilde{\Gamma}_L+\tilde{\Gamma}_R+\Gamma'_L)\sigma_{ae}, \tag{B11}$$

$$\dot{\sigma}_{af} = i(E'_2+U'_{22})\sigma_{af} + 2iT_2(\sigma_{aa}-\sigma_{ff}) + i(\sqrt{2}\gamma_C T\sigma_{ab} - 2\sqrt{2}\gamma_A T\sigma_{bf} - 2T_1\sigma_{ef}) - (\tilde{\Gamma}_L+\tilde{\Gamma}_R+\Gamma'_R)\sigma_{af}, \tag{B12}$$

$$\dot{\sigma}_{be} = i(U'_{11}-E'_2)\sigma_{be} + i\sqrt{2}\gamma_C T(\sigma_{bb}-\sigma_{ee}) + i(2T_1\sigma_{ba} - 2\sqrt{2}\gamma_A T\sigma_{ae} - \sqrt{2}\gamma_C T\sigma_{fe}) - (\Gamma_L+\Gamma_R+\Gamma'_L)\sigma_{be}, \tag{B13}$$

$$\dot{\sigma}_{bf} = i(U'_{22}-E'_1)\sigma_{bf} + i\sqrt{2}\gamma_C T(\sigma_{bb}-\sigma_{ff}) + i(2T_2\sigma_{ba} - 2\sqrt{2}\gamma_A T\sigma_{af} - \sqrt{2}\gamma_C T\sigma_{ef}) - (\Gamma_L+\Gamma_R+\Gamma'_R)\sigma_{bf}, \tag{B14}$$

$$\dot{\sigma}_{cd} = i(E'_2-E'_1)\sigma_{cd} + i\gamma_C T(\sigma_{cc}-\sigma_{dd}) + 2i(T_1\sigma_{cg}-T_2\sigma_{hd}) - \frac{1}{2}(\Gamma_L+\Gamma_R+2\tilde{\Gamma}_L+2\tilde{\Gamma}_R)\sigma_{cd}, \tag{B15}$$

$$\dot{\sigma}_{ch} = i(U'_{22}+E'_2)\sigma_{ch} + 2iT_2(\sigma_{cc}-\sigma_{hh}) - i\gamma_C T\sigma_{dh} - (\Gamma_L+\Gamma'_R+\tilde{\Gamma}_R)\sigma_{ch}, \tag{B16}$$

$$\dot{\sigma}_{dg} = i(U'_{11}+E'_1)\sigma_{dg} + 2iT_1(\sigma_{dd}-\sigma_{gg}) - i\gamma_C T\sigma_{cg} - (\Gamma_R+\Gamma'_L+\tilde{\Gamma}_L)\sigma_{ch}, \tag{B17}$$

$$\dot{\sigma}_{ef} = i(U'_{22}-U'_{11})\sigma_{ef} + 2i(T_2\sigma_{ea}-T_1\sigma_{af}) + i\sqrt{2}\gamma_C T(\sigma_{eb}-\sigma_{bf}) - (\Gamma'_L+\Gamma'_R)\sigma_{ef}, \tag{B18}$$

$$\dot{\sigma}_{cg} = i(U'_{11}+E'_2)\sigma_{cg} + iT_1\sigma_{cd} - i\gamma_C T\sigma_{dg} - \frac{1}{2}(\Gamma_L+\Gamma_R+2\Gamma'_L+2\tilde{\Gamma}_R)\sigma_{cg}, \tag{B19}$$

$$\dot{\sigma}_{dh} = i(U'_{22}+E'_1)\sigma_{dh} + iT_2\sigma_{dc} - i\gamma_C T\sigma_{ch} - \frac{1}{2}(\Gamma_L+\Gamma_R+2\Gamma'_R+2\tilde{\Gamma}_L)\sigma_{dg}, \tag{B20}$$

where E' and U' are the renormalized energy levels. $\tilde{\Gamma}_{L,R} = K''^2\Gamma_{L,R}T_{1,2}/\Delta_S$ corresponds to the one-by-one process. The superconducting phase, which do not change any result, is omitted here.

APPENDIX C: DERIVATION OF QUANTUM MASTER EQUATIONS FOR THE ENTANGLER USING A PAIR-HAMILTONIAN

The same set of quantum master equations could be obtained from an effective Hamiltonian applied to the method developed in Ref. 18. In considering all processes, this Hamiltonian can be derived from the microscopic Hamiltonian (1) using a projective transformation which eliminates states with quasiparticles in the superconductor to the lowest order:

$$\begin{aligned}\mathcal{H}_{\text{eff}} = P\mathcal{H}_0 P &+ 2\sqrt{2}\gamma_A T\frac{1}{\sqrt{2}}(d_{1\sigma}^{\dagger}d_{2-\sigma}^{\dagger} - d_{1-\sigma}^{\dagger}d_{2\sigma}^{\dagger})S \\ &+ 2T_1(d_{1\sigma}^{\dagger}d_{1-\sigma}^{\dagger})S + 2T_2(d_{2\sigma}^{\dagger}d_{2-\sigma}^{\dagger})S + \sqrt{2}\gamma_C T\sum_{\sigma} d_{1\sigma}^{\dagger}d_{2\sigma} \\ &+ \sum_{l,\sigma}\Omega_l a_{l\sigma}^{\dagger}d_{1\sigma} + \sum_{r,\sigma}\Omega_r a_{r\sigma}^{\dagger}d_{2\sigma} + \sum_{l,\sigma}\hat{\Omega}_l d_{1\sigma}^{\dagger}a_{l\sigma}^{\dagger}S \\ &+ \sum_{r,\sigma}\hat{\Omega}_r d_{2\sigma}^{\dagger}a_{r\sigma}^{\dagger}S + \text{H.c.} \qquad \text{(C1)}\end{aligned}$$

with $\hat{\Omega}_{l,r}=\Omega_{l,r}T_{1,2}/\Delta_S$ corresponds to the one-by-one process. The method only requires the amplitude for probability of processes coupling different states of the quantum system, and leads to a following general system:

$$\dot{\sigma}_{\alpha\alpha} = i\sum_{\gamma\neq\alpha}\Omega_{\alpha\gamma}(\sigma_{\alpha\gamma} - \sigma_{\gamma\alpha}) - \sum_{\gamma\neq\alpha}\Gamma_{\alpha\to\gamma}\sigma_{\alpha\alpha} + \sum_{\gamma\neq\alpha}\Gamma_{\gamma\to\alpha}\sigma_{\gamma\gamma}, \qquad \text{(C2)}$$

$$\begin{aligned}\dot{\sigma}_{\alpha\beta} = i(E_\beta - E_\alpha)\sigma_{\alpha\beta} &+ i\sum_{\gamma\neq\beta}\left(\sigma_{\alpha\gamma}\Omega_{\gamma\beta} - i\sum_{\gamma\neq\alpha}\Omega_{\alpha\gamma}\sigma_{\gamma\beta}\right) \\ &- \frac{1}{2}\left(\sum_{\gamma\neq\alpha}\Gamma_{\alpha\to\gamma} - \sum_{\gamma\neq\beta}\Gamma_{\beta\to\gamma}\right)\sigma_{\alpha\beta} \\ &+ \frac{1}{2}\sum_{\gamma\delta\neq\alpha\beta}(\Gamma_{\gamma\to\alpha} + \Gamma_{\delta\to\beta})\sigma_{\gamma\delta}, \qquad \text{(C3)}\end{aligned}$$

where the Ω's are the coherent transition matrix elements and the Γ's the relaxation rates.

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