## **Dynamical Coulomb Blockade and Spin-Entangled Electrons**

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We consider the creation of mobile and nonlocal spin-entangled electrons from tunneling of a BCSsuperconductor (SC) to two normal leads of finite resistivity. The resulting dynamical Coulomb blockade effect, which we describe phenomenologically in terms of an electromagnetic environment, is shown to be enhanced for tunneling of two electrons from a Cooper pair into the same lead compared to the desired pair-split process where each electron enters a different lead. Conversely, this latter process is suppressed by a finite separation between the tunneling points on the SC.

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Introduction.-The controlled creation of nonlocal entanglement is crucial in quantum communication as well as in quantum computation tasks [1]. Several solid state entanglers, a device that creates mobile and nonlocal pairwise entangled electrons, were proposed recently [2-6]. A particularly interesting quantity is the spin of the electron which was shown to be a promising realization of a quantum bit [7]. A natural source of spin entanglement is provided by Cooper pairs in an s-wave superconductor (SC), since the Cooper pairs are in a spin-singlet state. Weakly coupling the SC to a normal region allows for (pair-)tunneling of Cooper pairs from the SC to normal leads and single-particle tunneling is suppressed at low energies below the SC gap. Coulomb interaction between the two electrons of a pair can then be used to separate them spatially leading to nonlocality. To mediate the necessary interaction entangler setups containing quantum dots [3] or that exhibit Luttinger liquid correlations [4,5] (e.g., nanotubes in the metallic regime) were proposed recently.

In this Letter we show that a considerably simpler experimental realization can be used to generate the necessary Coulomb interaction between the electrons of a pair. Indeed, if the normal leads are resistive a dynamical Coulomb blockade (CB) effect is generated with the consequence that in a pair tunneling process into the same lead the second electron still experiences the Coulomb repulsion of the first one, which has not yet diffused away. Natural existing candidates with long spin decoherence lengths (~100  $\mu$ m [8]) for such a setup are, e.g., semiconductor systems tunnel coupled to a SC, as experimentally implemented in InAs [9], InGaAs [10], or GaAs/AlGaAs [11]. Recently, a two-dimensional electron gas (2DEG) with a resistance per square approaching the quantum resistance  $R_Q = h/e^2 \sim 25.8 \text{ k}\Omega$  could be achieved by depleting the 2DEG with a voltage applied between a back gate and the 2DEG [12]. In metallic normal NiCr leads of width ~100 nm and length ~10  $\mu$ m, resistances of R = 22-24 k $\Omega$ , have been produced at low temperatures. Even larger resistances R =200–250 k $\Omega$  have been measured in Cr leads [13].

We use a phenomenological approach to describe charge dynamics in the electromagnetic circuit which is described in terms of normal-lead impedances and junction capacitances; see Fig. 1. The subgap transport of a single SN junction under the influence of an electromagnetic environment has been studied in detail [14,15]. In order to create nonlocal entangled states in the leads we have to go beyond previous work to investigate the physics of two tunnel junctions in parallel with two distinct transport channels for singlets. A Cooper pair can tunnel as a whole into one lead, or the pair can split and the two electrons enter separate leads, leading to a nonlocal spin singlet in the leads. Subsequently, the degree of spin entanglement can be detected via an enhanced shot noise in a beam splitter setup [16] or, alternatively, by measuring Bell's inequality [17]. By using spin filters, e.g., quantum dots in the CB regime [18], one can then use current-current correlation measurements for testing Bell's inequality. In the case where the pair splits we



FIG. 1. Entangler setup: A BCS bulk superconductor (SC) with chemical potential  $\mu_S$  is tunnel coupled (amplitude  $t_0$ ) via points  $\mathbf{r}_1$  and  $\mathbf{r}_2$  of the SC to two Fermi liquid leads 1, 2 with resistance  $R_{1,2}$ . The leads are held at the same chemical potential  $\mu_I$  such that a bias voltage  $\mu = \mu_S - \mu_I$  is applied between the SC and the two leads via the voltage source V. The tunnel junctions 1, 2 have capacitances  $C_{1,2}$ .

find that the dynamical CB effect is uncorrelated for the two electron charges. In contrast, if the two electrons tunnel into the same lead we find a dynamical CB consistent with a charge q = 2e, where e is the elementary charge. Thus the CB effect is twice as large for the unsplit process which enhances the probability for a nonlocal (pair-split) process. On the other hand, we show that the spatial correlations of a Cooper pair results in a suppression factor for tunneling via different junctions which is weaker for lower dimensional SCs.

Setup and formalism.—The setup is sketched in Fig. 1. The SC is held at the (electro-)chemical potential  $\mu_S$  by a voltage source V. The two electrons of a Cooper pair can tunnel via two junctions placed at points  $r_1$  and  $r_2$  on the SC to two separate normal leads 1 and 2 which have resistances  $R_1$  and  $R_2$ , respectively. They are kept at the same chemical potential  $\mu_l$  so that a bias voltage  $\mu \equiv$  $\mu_S - \mu_l$  is applied between SC and leads [19]. The system Hamiltonian decomposes into three parts H = $H^e + H_{env} + H_T$ . Here  $H^e = H_S + \sum_{n=1,2} H_{ln}$  describes the electronic parts of the isolated subsystems consisting of the SC and Fermi liquid leads n = 1, 2, with  $H_{ln} =$  $\sum_{\mathbf{p}\sigma} \varepsilon_p c_{n\mathbf{p}\sigma}^{\dagger} c_{n\mathbf{p}\sigma}$ , where  $\sigma = (\uparrow, \downarrow)$ . The s-wave bulk SC is described by the BCS Hamiltonian  $H_S - \mu_S N_S =$  $\sum_{\mathbf{k}\sigma} E_k \gamma_{\mathbf{k}\sigma}^{\mathsf{T}} \gamma_{\mathbf{k}\sigma}$  with the quasiparticle spectrum  $E_k =$  $\overline{(\xi_k^2 + \Delta^2)^{1/2}}$  where  $\xi_k = k^2/2m - \mu_s$ . The electron creation  $(c_{\mathbf{k}\sigma}^{\dagger})$  and annihilation  $(c_{\mathbf{k}\sigma})$  operators are related to the quasiparticle operators by the Bogoliubov transformation  $c_{\mathbf{k}(\uparrow\downarrow)} = u_k \gamma_{\mathbf{k}(\uparrow\downarrow)} \pm v_k \gamma_{-\mathbf{k}(\downarrow\uparrow\uparrow)}^{\dagger}$ , where  $u_k$  and  $v_k$ are the usual BCS coherence factors. To describe resistance and dissipation in the normal leads we use a phenomenological approach [20], where the electromagnetic fluctuations in the circuit (being bosonic excitations) due to electron-electron interaction and the lead resistances are modeled by a bath of harmonic oscillators which is linearly coupled to the charge fluctuation  $Q_n$  of the junction capacitor *n* (induced by the tunneling electron). This physics is described by [20,21]

$$H_{\text{env},n} = \frac{Q_n^2}{2C_n} + \sum_{j=1}^N \left[ \frac{q_{nj}^2}{2C_{nj}} + \frac{(\phi_n - \varphi_{nj})^2}{2e^2 L_{nj}} \right].$$
(1)

The phase  $\phi_n$  of junction *n* is the conjugate variable to the charge satisfying  $[\phi_n, Q_m] = ie\delta_{n,m}$ . As a consequence  $e^{-i\phi_n}$  reduces  $Q_n$  by one elementary charge *e*. We remark that the SC is held at constant chemical potential  $\mu_S$  by the voltage source; see Fig. 1. Therefore the charge relaxation of a nonequilibrium charge on one of the capacitors described by (1) does not influence the charge dynamics of the other junction and, as a consequence,  $H_{\text{env}} = \sum_{n=1,2} H_{\text{env},n}$  [22]. Electron tunneling through junctions 1, 2 located at points  $\mathbf{r}_1$ ,  $\mathbf{r}_2$  of the SC nearest to the leads 1, 2 is described by the tunneling Hamiltonian  $H_T = \sum_{n=1,2} H_{Tn} + \text{H.c.}$ , where

$$H_{Tn} = t_0 \sum_{\sigma} \psi_{n\sigma}^{\dagger} \Psi_{\sigma}(\mathbf{r}_n) e^{-i\phi_n}.$$
 (2)

Here  $t_0$  is the bare electron tunneling amplitude which we assume to be spin independent and the same for both leads. Since  $H_T$  conserves spin we have  $[H, \mathbf{S}_{tot}^2] = 0$ , and thus the two electrons from a given Cooper pair singlet which have tunneled to the lead(s) remain in the singlet state.

Current of two electrons tunneling into different leads.—We use a T-matrix approach [23] to calculate tunneling currents. At zero temperature the current  $I_1$ for tunneling of two electrons coming from the same Cooper pair into different leads is given to lowest order in  $t_0$  by [4]

$$I_{1} = 2e \sum_{\substack{n \neq n' \\ m \neq m'}} \int_{-\infty}^{\infty} dt \int_{0}^{\infty} dt' \int_{0}^{\infty} dt'' e^{-\eta(t'+t'')+i(2t-t'-t'')\mu} \langle H_{Tm}^{\dagger}(t-t'')H_{Tm'}^{\dagger}(t)H_{Tn}(t')H_{Tn'}(0) \rangle,$$
(3)

where  $\eta \to 0^+$ , and the expectation value is to be taken in the ground state of the unperturbed system. The physical interpretation of Eq. (3) is a hopping process of two electrons with opposite spins from two spatial points  $\mathbf{r}_1$  and  $\mathbf{r}_2$  of the SC to the two leads 1, 2, thereby removing a Cooper pair in the SC, and back again. The delay times between the two tunneling processes of the electrons within a pair are t' and t'', respectively, whereas the time between destroying and creating a Cooper pair is given by t. This process is contained in the correlation function

$$\sum_{n\neq n'\atop m\neq m'} \langle H_{Tm}^{\dagger}(t-t'') H_{Tm'}^{\dagger}(t) H_{Tn}(t') H_{Tn'}(0) \rangle = |t_0|^4 \sum_{\sigma, n\neq m} \{ G_{n\sigma}(t-t'') G_{m,-\sigma}(t-t') \mathcal{F}_{nm\sigma}(t') \mathcal{F}_{nm\sigma}^{*}(t'') \langle e^{i\phi_n(t-t'')} e^{-i\phi_n(0)} \rangle \\ \times \langle e^{i\phi_m(t-t')} e^{-i\phi_m(0)} \rangle - G_{m,-\sigma}(t-t'-t'') G_{n\sigma}(t) \mathcal{F}_{nm\sigma}(t') \\ \times \mathcal{F}_{mn}^{*} - \sigma(t'') \langle e^{i\phi_m(t-t'-t'')} e^{-i\phi_m(0)} \rangle \langle e^{i\phi_n(t)} e^{-i\phi_n(0)} \rangle \}.$$
(4)

The lead Green's functions are  $G_{n\sigma}(t) \equiv \langle \psi_{n\sigma}(t)\psi_{n\sigma}^{\dagger}(0) \rangle \simeq (\nu_l/2)/it$ , with  $\nu_l$  being the density of states (DOS) per volume at the Fermi level  $\mu_l$  of the leads. The anomalous Green's function of the SC is  $\mathcal{F}_{nm\sigma}(t) \equiv \langle \Psi_{-\sigma}(\mathbf{r}_n, t)\Psi_{\sigma}(\mathbf{r}_n, 0) \rangle = [\text{sgn}(\sigma)/V_S] \sum_{\mathbf{k}} u_k v_k \exp(-iE_k t + i\mathbf{k} \cdot \delta \mathbf{r})$  with  $\delta \mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ , and  $V_S$  is the volume of the SC. The bath correlator can be expressed as  $\langle \exp(i\phi_n(t)) \exp(-i\phi_n(0)) \rangle = \exp[J(t)]$  with  $J(t) = 2 \int_0^\infty (d\omega/\omega) [\text{Re}Z_T(\omega)/R_Q] [\exp(-i\omega t) - 1]$ . Here we introduced the total impedance  $Z_T = (i\omega C + R^{-1})^{-1}$ , with a purely Ohmic lead impedance  $Z_n(\omega) = R$ , which we assume to be the same for both tunnel junctions and leads. For 267003-2

(7)

small times,  $\omega_R |t| \ll 1$ , we can approximate  $J(t) \sim -iE_c t$  where  $E_c = e^2/2C$  is the charging energy and  $\omega_R = 1/RC$  is the bath frequency cutoff which is the inverse classical charge relaxation time  $\tau_{cl}$  of an RC circuit. For the long-time behavior,  $\omega_R |t| \gg 1$ , we get  $J(t) \sim -(2/g)[\ln(i\omega_R t) + \gamma]$  with  $\gamma = 0.5772$  the Euler number and  $g = R_Q/R$  is the dimensionless lead conductance.

We first consider the low bias regime  $\mu \ll \Delta$ ,  $\omega_R$ . In this limit the delay times t' and  $t'' \leq 1/\Delta$  can be neglected compared to  $t \leq 1/\mu$  in all correlators in (4) and the bath correlators are dominated by the long-time behavior of J(t). We then obtain for the current

$$I_1 = e\pi\mu\Gamma^2 F_d^2(\delta r) \frac{e^{-4\gamma/g}}{\Gamma(4/g+2)} \left(\frac{2\mu}{\omega_R}\right)^{4/g}.$$
 (5)

The spatial correlation of a Cooper pair results in the suppression factor  $F_{d=3}(\delta r) = [\sin(k_F \delta r)/k_F \delta r] \times$  $\exp(-\delta r/\pi\xi)$  with  $\delta r = |\delta \mathbf{r}|$ . The exponential decay sets in on the length scale of the coherence length  $\xi$ . It is on the order of micrometers for usual s-wave materials and therefore  $\delta r \ll \xi$  for  $\delta r$  in the range of nanometers. More severe is the power-law decay  $\propto 1/(k_F \delta r)^2$  with  $k_F$ the Fermi wave number in the SC. This power law is sensitive to the effective dimensionality d of the SC with weaker decay in lower dimensions. Indeed, in two dimensions [24] and for  $k_F \delta r \gg 1$ , but still  $\delta r < \xi$ , we get  $F_{d=2}^2 \propto 1/(k_F \delta r)$ , and in one dimension there is no power-law decay as a function of  $k_F \delta r$ . In (5) we introduced the gamma function  $\Gamma(x)$  and the dimensionless tunnel conductance  $\Gamma = \pi \nu_S \nu_l |t_0|^2$  with  $\nu_S$  being the DOS per volume of the SC at the Fermi level  $\mu_s$ . The exponent 4/g in (5) is twice the value for single electron tunneling [20] via one junction since the tunneling events into different leads are uncorrelated.

We consider now the large bias regime  $\Delta$ ,  $\mu \gg \omega_R$ . For  $\Delta \gg |\mu - E_c| \gg \omega_R$  we can use the short time expansion for J(t) in (4) and can again neglect the delay times t' and t'' compared to t in all correlators in (4). We then obtain for  $I_1$  in the large bias limit and up to small contributions  $\sim e \pi \Gamma^2 F_d^2(\delta r) \omega_R [\mathcal{O}(\omega_R/\mu) + \mathcal{O}(\omega_R/|\mu - E_c|)]$ 

$$I_1 = e\pi\Gamma^2 F_d^2(\delta r)\Theta(\mu - E_c)(\mu - E_c).$$
(6)

This shows a gap in  $I_1$  for  $\mu < E_c$  and  $R \rightarrow \infty$  which is the hallmark of dynamical CB.

Current of two electrons tunneling into the same lead.—The current  $I_2$  for tunneling of two electrons into the same lead, 1 or 2, is given by (3) but with n = n' and m = m' = n. We assume that the two electrons tunnel off the SC from the same point and therefore  $\delta r = 0$  here. Since both electrons tunnel into the same lead the bath correlation functions do not separate anymore as was the case in (4). Instead we have to look at the full 4-point correlator

and the  $e^{J(t-t'-t'')+J(t-t')+J(t-t'')+J(t)-J(t')-J(t')-J(-t'')}.$ 

 $\langle e^{i\phi_n(t-t'')}e^{i\phi_n(t)}e^{-i\phi_n(t')}e^{-i\phi_n(0)}\rangle$ 

The lead correlators again factorize into a product of two single-particle Green's functions since they are assumed to be Fermi liquids and in addition there appear no spin correlations due to tunneling of two electrons with opposite spins.

We first consider the low bias regime  $\mu \ll \omega_R, \Delta$ . Here again we can assume that |t| is large compared to the delay times t' and t'', but it turns out to be crucial to distinguish carefully between  $\Delta \gg \omega_R$  and  $\Delta \ll \omega_R$ . We first treat the case  $\Delta \gg \omega_R$  and approximate  $\exp[-(J(t') + J(-t''))] \simeq \exp[-iE_c(t'' - t')]$  in (7). In this limit and for  $\Delta > E_c$  the current  $I_2$  becomes

$$I_{2} = e \pi \mu \Gamma^{2} \frac{(4\Delta/\pi)^{2}}{\Delta^{2} - E_{c}^{2}}$$

$$\times \arctan^{2} \left\{ \sqrt{\frac{\Delta + E_{c}}{\Delta - E_{c}}} \right\} \frac{e^{-8\gamma/g}}{\Gamma(8/g + 2)} \left(\frac{2\mu}{\omega_{R}}\right)^{8/g}.$$
 (8)

The tunneling of a charge q = 2e clearly shows up in the exponent 8/g in (8). In addition to this double charging effect we see from (8) that increasing  $E_c$  has two opposite effects. On the one hand the factor  $(2\mu/\omega_R)^{8/g}$ decreases, while the  $\Delta$ -dependent terms increase. The latter terms result from a charge relaxation in the virtual state with one electron of the entangled pair still being on the SC. The formula (8) is valid if  $\sqrt{(\Delta - E_c)/(\Delta + E_c)} \gg \sqrt{\omega_R/\Delta}$ .

In the other limit where  $\Delta \ll \omega_R$ , e.g., for small R, we can assume that  $\omega_R t'$  and  $\omega_R t'' \gg 1$  and therefore approximate  $\exp[-(J(t') + J(-t''))] \simeq \exp(4\gamma/g) \times \omega_R^{4/g} (t't'')^{2/g}$ . In this limit we obtain

$$I_2 = e \pi \mu \Gamma^2 A(g) \left(\frac{2\mu}{\omega_R}\right)^{4/g} \left(\frac{2\mu}{\Delta}\right)^{4/g}, \tag{9}$$

with  $A(g) = (2e^{-\gamma})^{4/g}\Gamma^4(1/g + 1/2)/\pi^2\Gamma(8/g + 2)$ . Here the relative suppression of the current  $I_2$  compared to  $I_1$  is proportional to  $(2\mu/\Delta)^{4/g}$  and not to  $(2\mu/\omega_R)^{4/g}$ as in the case of an infinite  $\Delta$ . This is because the virtual state with a quasiparticle in the SC can last much longer than  $\tau_{cl}$ , and, as a consequence, the power-law suppression of the current is weakened since  $\Delta \ll \omega_R$  here. To our knowledge, the result (9) was not discussed in the literature so far [27], but similar results are obtained when SCs are coupled to Luttinger liquids [4]. A large gap  $\Delta$  is therefore crucial to suppress  $I_2$ .

In the large voltage regime  $\Delta$ ,  $\mu \gg \omega_R$  we expect a Coulomb gap due to a charge q = 2e. Indeed, in the parameter range  $|\mu - 2E_c| \gg \omega_R$  and  $\Delta \gg |\mu - E_c|$  we obtain for  $I_2$  again up to small contributions  $\sim e\pi\Gamma^2\omega_R[\mathcal{O}(\omega_R/\mu) + \mathcal{O}(\omega_R/|\mu - 2E_c|)]$ ,

$$I_2 = e \pi \Gamma^2 \Theta(\mu - 2E_c)(\mu - 2E_c). \tag{10}$$

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FIG. 2. Current ratio  $I_2/I_1$  (entangler efficiency) and  $I_1$  in the regime  $\mu \ll \Delta$ ,  $\omega_R$  and  $\Delta \gg E_c$ ,  $\omega_R$  as a function of  $4/g = 4R/R_Q$ . Chosen parameters:  $E_c = 0.1 \text{ meV}$ ,  $k_F \delta r = 10$ ,  $\Gamma = 0.1$ , and  $\mu = 5 \mu \text{eV}$  (left plot),  $\mu = 15 \mu \text{eV}$  (right plot). For a 2D SC,  $I_1$  and  $I_1/I_2$  can be multiplied by 10.

This shows that  $I_2$  is small  $(\propto \omega_R^2/|\mu - 2E_c|)$  in the regime  $E_c < \mu < 2E_c$ , whereas  $I_1$  is finite  $[\propto F_d^2(\delta r)(\mu - E_c)]$ .

Discussion and conclusions.—We now give numerical values for the current magnitudes and efficiencies of our entangler. We first discuss the low bias regime  $\mu \ll \Delta$ ,  $\omega_R$ . In Fig. 2 we show the ratio  $I_2/I_1$  (efficiency of entangler) and  $I_1$  for  $\Delta \gg E_c$ ,  $\omega_R$  as a function of 4/g for realistic system parameters (see caption of Fig. 2). The plots show that a very efficient entangler can be expected for lead resistances  $R \leq R_0$ . The total current is then on the order of  $I_1 \gtrsim 10$  fA. In the large bias regime  $\mu \gg \omega_R$  and for  $E_c < \mu < 2E_c$  we obtain  $I_2/I_1 \propto (k_F \delta r)^{d-1} \omega_R^2/(2E_c - \mu)(\mu - E_c)$ , where we assume that  $2E_c - \mu$  and  $\mu - E_c \gg \omega_R$ . For  $\mu \simeq 1.5E_c$ and using  $\omega_R = gE_c/\pi$  we obtain approximately  $I_2/I_1 \propto (k_F \delta r)^{d-1}g^2$ . To have  $I_2/I_1 < 1$  we require  $g^2 < 0.01$  for d = 3, and  $g^2 < 0.1$  for d = 2. Such small values of g have been produced approximately in Cr leads [13]. For  $I_1$  we obtain  $I_1 \simeq e(k_F \delta r)^{1-d} (\mu - E_c) \Gamma^2 \simeq e(k_F \delta r)^{1-d} E_c \Gamma^2 \simeq 2.5$  pA for d = 3 and for the same parameters as used in Fig. 2. This shows that  $I_1$  is much larger than for low bias voltages, but an efficient entangler requires high lead resistances  $R \gtrsim 10R_O$ . Our discussion shows that the proposed device should be realizable within state-of-the-art techniques. Finally, we note that the delay time  $\sim 1/\Delta$  within a pair is much shorter than the time separation between subsequent pairs  $\sim 2e/I_1$ , so that different pairs do not overlap in time. This is crucial for detection of entanglement via correlation measurements described in the Introduction.

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