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Attractive correlation and charge skipping in a metallic grain capacitively coupled to a superconducting island

C. Holmqvist ^{a)b)}, D. Feinberg ^{a)}, and A. Zazunov ^{c)d)}

^{a)} Institut NEEL, Centre National de la Recherche Scientifique and Université Joseph Fourier, BP 166, 38042 Grenoble, France

^{b)} Chalmers University of Technology, S-412 96 Göteborg, Sweden

^{c)} Laboratoire de Physique et Modélisation des Milieux Condensés,

Université Joseph Fourier and CNRS, BP 166, 38042 Grenoble, France and

^{d)} Centre de Physique Théorique, Case 907, Luminy, 13288 Marseille Cedex 9, France

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Capacitively coupling a normal metallic grain to a superconducting island can lead to attractive correlation (negative- U center) in the grain. The considered setup geometry is similar to that of a Cooper pair box coupled to a single-electron transistor, but operates in the regime of strong capacitive coupling. In the Coulomb staircase for the grain, positive steps ($+2e$) skipping odd charge numbers are found to be followed by negative ones ($-e$) signaling the occurrence of a negative differential capacitance. The condition for charge skipping is analyzed as a function of the ratio between the Josephson (E_J) and charging energies. The non-monotonous charging curves are calculated in the limits of large and small E_J .

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Single electron charging effects are characteristic of small metallic structures. When a small-capacitance grain is weakly coupled to a metallic reservoir at sufficiently low temperatures, the average number of charges, n_N , in the grain increases one by one as the gate voltage V_{gN} is continuously varied, leading to Coulomb blockade oscillations in the conductance [1]. Plateaus in the charging curve indicate an insulating-like regime, with zero differential capacitance $C_{diff} = e \frac{dn_N}{dV_{gN}}$, while steps signal a metallic-like regime where C_{diff} is very large. Such oscillations have also been studied in superconducting islands where charging steps involve only electron pairs if the superconducting gap in the island is larger than the charging energy [2], and single charges in the opposite case [3]. The study of charging patterns has been extended to double islands, coupled by a capacitance as well as by electron tunneling. The islands can be both normal metals [4, 5], or superconducting [6]. The latter embodies a particular case of coupled superconducting qubits and displays challenging coherent electronic transport.

The present Letter addresses the “mixed” case where a superconducting island (S) is coupled to a normal grain (N) by a large capacitance (Fig. 1). Moreover, we disregard any electron tunneling between S and N. This situation might be achieved for instance at the interface between a superconductor (Nb) and a two-dimensional electron gas (InAs). The S island is connected to a superconducting reservoir by a Josephson junction (JJ), and the N grain is connected to a normal reservoir. Notice that if N is instead coupled to two reservoirs and a current flows through N, the set-up is similar to that of a Cooper pair box (CB) coupled to a single-electron transistor (SET). It has been studied in great detail as a read-out device for a superconducting (charge) qubit embodied in the S island [7]. In this case, the coupling between N and S is assumed to be very small, in order to minimize the decoherence due to backaction of the normal part of the device onto the superconducting one. Here we instead consider the case of a large capacitive coupling, which strongly correlates the charge fluctuations in the two islands. We focus on the charging properties

of the N grain, under the influence of the S island. Notice that no proximity effect occurs in the present case due to the absence of tunneling between the two islands. As shown below, this situation may lead to an overscreening of the Coulomb repulsion in N and to skipping of certain charge states as the N gate voltage is varied. This signals a correlated motion of electrons in and out the N grain. Such an attractive correlation is reminiscent of the so-called “negative- U ” center in solids [8]. Here it is due to screening by the neighboring pair fluctuations in S. A related effect has been proposed by Averin and Bruder for providing a controlled coupling between two superconducting charge qubits [9].

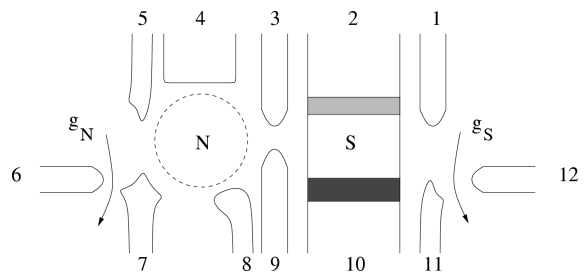


FIG. 1: Schematic view of a normal grain (2DEG) coupled to a Cooper pair box composed of a Josephson junction connecting superconducting reservoir 2 and island S gated by 10. For strong capacitive coupling (controlled by 3, 9), S imposes an attractive correlation amongst electrons tunneling between the normal grain (N) and its reservoir (defined by 7, 8). Detection is made by sweeping the gate voltage (4) and measuring the island voltages using quantum point contacts for both N (5,6,7) and S (1,11,12).

The S island is connected to the reservoir by a JJ, with Josephson energy E_J and capacitance C_J , and coupled to a gate imposing a charge offset $Q_S = 2eN_S = C_{gS}V_{gS}$, with $C_{gS} \ll C_J$. Symmetrically, the N island is connected to a normal reservoir by a tunnel junction, with one-electron tunneling rate Γ and capacitance C_N , and experiences a gate offset $Q_N = eN_N = C_{gN}V_{gN}$, with $C_{gN} \ll C_N$. Most

importantly, the islands N and S are coupled by a large capacitance $C_0 > C_N, C_J$. We take the gap in S to be larger than the charging energy, so that only even charge number states $2\bar{n}_S$ occur in S, while all charge states n_N are a priori possible in N (\bar{n}_S is the number of Cooper pairs in S). The temperature is supposed to be small in order to neglect quasiparticle tunneling in S. Defining $C_{\Sigma S} = C_J + C_0 + C_{gS}$ and $C_{\Sigma N} = C_N + C_0 + C_{gN}$, $b = \frac{C_{\Sigma N}}{C_{\Sigma S}}$ and $r = \frac{C_0}{\sqrt{C_{\Sigma N} C_{\Sigma S}}}$, the total charging energy of the NS system can be written as

$$E_C = E_{CN}[(n_N - N_N)^2 + 4b(\bar{n}_S - N_S)^2 + 4r\sqrt{b}(n_N - N_N)(\bar{n}_S - N_S)] \quad (1)$$

with $E_{CN} = \frac{e^2}{2C_{\Sigma N}(1-r^2)}$. The numbers n_N and \bar{n}_S are integers while N_N, N_S are continuous control parameters. Notice that the asymmetry parameter b and the coupling parameter $r < 1$ are not independent, as $r < \min(b, \frac{1}{\sqrt{b}})$. From Eq. (1), one can plot the charge stability diagram of the isolated NS system in the (N_N, N_S) plane. First, for a value N_S imposing an integer number of pairs in S, say $N_S = 1$, the charging number n_N increases monotonously with N_N . Next, consider a case where \bar{n}_S fluctuates, for instance $N_S = 0.5$. For small r , as shown in Fig. 2(a), one sees that n_N is again a monotonous function of N_N : the sequence of charge states (n_N, \bar{n}_S) as N_N increases reads $(0, \bar{0}), (0, \bar{1}), (1, \bar{0}), (1, \bar{1}), (2, \bar{0}), (2, \bar{1})$, etc. (note the oscillation of \bar{n}_S). The result is very different if r is large. In Fig. 2(b), for $N_S = 0.5$, n_N increases with N_N but in a non-monotonous way, the charge state sequence being $(1, \bar{0}), (0, \bar{1}), (2, \bar{0}), (1, \bar{1}), (3, \bar{0}), (2, \bar{1})$, etc. The corresponding Coulomb staircases are plotted in inset.

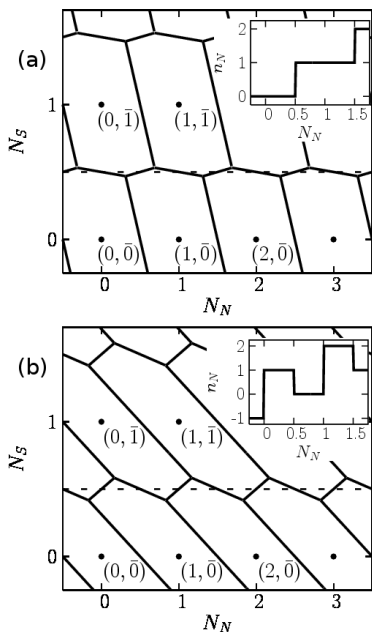


FIG. 2: (a) Charge stability (or honeycomb) diagram for $r = 0.2$, $b = 1$. The inset shows the charging curve for N. (b) Same for $r = 0.8$. The Coulomb staircase (inset) exhibits charge skipping effects.

Let us comment this result. The transition from $(n_N, \bar{1})$ to $(n_N + 2, \bar{0})$ at $N_N = n_N + 1$ happens to “skip” the charge state $n_N + 1$ in the grain. This indicates a strong charge correlation between the island charges in N and S, fluctuating by two units in opposite directions. In other terms, the attractive potential present in the S island leads to an attractive potential (“negative-U”[10]) in N which overcomes the Coulomb repulsion between charges in N. After increasing by two units, n_N must necessarily decrease by one unit, such that n_N as a function of N_N has an average slope equal to one. Therefore, together with charge skipping at integer values of N_N , a negative differential capacitance (NDCA) $C_{diff} = C_{gN} \frac{dn_N}{dN_N}$ occurs at half-integer values of N_N . In addition to the already known “insulating” and “metallic” behaviors, this phenomenon signals an overscreening of the charge repulsion in N due to pair fluctuations in S. Strikingly, the total number of steps, both positive and negative, is doubled with respect to the usual case. Both “charge skipping” and NDCA effects occur above the dotted line indicated in the inset in Fig. 3 displaying a (b, r) diagram. This line can be determined by a simple magnetic analogy: defining charge pseudospins $\sigma_S^z = 2(\bar{n}_S - N_S)$ and $\sigma_N^z = n_N - N_N$ for $N_S = 0.5$ and N_N integer, E_C can be rewritten as $E_C = E_{CN}[(\sigma_N^z)^2 + 2r\sqrt{b}\sigma_N^z\sigma_S^z + b]$, thus with an anisotropy and an antiferromagnetic coupling between charge pseudospins. If $r\sqrt{b} > \frac{1}{2}$, the “antiferromagnetic” solution $\sigma_N^z = \pm 1$, $\sigma_S^z = \mp 1$ is favored, e.g. skipping the state $n_N = N_N$ ($\sigma_N^z = 0$). Notice that in the limit $r = b = 1$, $E_C = E_{CN}(n - N)^2$, where $N = N_N + 2N_S$ and $n = n_N + 2\bar{n}_S$, is a conserved quantity. This case (C_0 infinite) where the two islands can be considered as a single capacitor is infinitely degenerate. As a result, for $r \rightarrow 1$ – provided such a regime can be reached – a large number of charge configurations are very close in energy, leading to huge charge fluctuations in the system.

To go beyond a purely electrostatic consideration, let us now write the full Hamiltonian of the open NS system:

$$H = E_C + \sum_{k\sigma} \varepsilon_k c_{kR,\sigma}^\dagger c_{kR,\sigma} + \sum_{q\sigma} \varepsilon_q c_{qN,\sigma}^\dagger c_{qN,\sigma} + \sum_{kq\sigma} T_{k,q} c_{kR,\sigma}^\dagger c_{qN,\sigma} + H.c. - \frac{E_J}{2} (|\bar{n}_S + 1\rangle \langle \bar{n}_S| + H.c.), \quad (2)$$

where k (q) denotes electron states in the normal reservoir R (grain N), and the Coulomb interaction E_C is given by Eq. (1). The total charge in N is $n_N = \sum_{q\sigma} c_{qN,\sigma}^\dagger c_{qN,\sigma}$. Assuming constant densities of states in N and R, the single-electron transition rate from R to N is given in the golden rule approximation by $\Gamma^{(+1)} = \frac{\delta E_C^{(+1)}}{e^2 R_N} [\exp(\delta E_C^{(+1)}/k_B T) - 1]^{-1}$.

Considering first the case of small E_J , we perform a T-matrix calculation of the transition rates from $(0, \bar{1})$ to $(2, \bar{0})$ (close to $N_N = 1$) and from $(2, \bar{0})$ to $(1, \bar{1})$ (close to $N_N = 1.5$). For the first transition, we take into account three configuration paths involving higher-energy states: $(0, \bar{1}) \rightarrow (1, \bar{1}) \rightarrow (2, \bar{1}) \rightarrow (2, \bar{0})$, $(0, \bar{1}) \rightarrow (1, \bar{1}) \rightarrow (1, \bar{0}) \rightarrow (2, \bar{0})$, and $(0, \bar{1}) \rightarrow (0, \bar{0}) \rightarrow (1, \bar{0}) \rightarrow (2, \bar{0})$. For the second transition, only one excited state is involved: $(2, \bar{0}) \rightarrow (1, \bar{0}) \rightarrow (1, \bar{1})$ and $(2, \bar{0}) \rightarrow (2, \bar{1}) \rightarrow (1, \bar{1})$.

The shape of each step is calculated at finite temperature by solving the master equation governing the dynamics of the probabilities $p(0, \bar{1})$, $p(2, \bar{0})$ for the positive step and $p(2, \bar{0})$, $p(1, \bar{1})$ for the negative one. The master equation reads as usual $\dot{p}(a) = \Gamma^{b \rightarrow a} p(b) - \Gamma^{a \rightarrow b} p(a)$ with $p(b) = 1 - p(a)$ for the main states a, b involved in the transition. Here, the probabilities of other states are neglected, e.g., close to $N_N = 1$ or $N_N = 1.5$. This is a valid assumption if the steps are sufficiently narrow. The calculated steps are shown in Fig. 3.

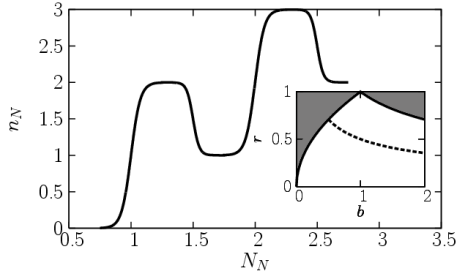


FIG. 3: Coulomb staircase in the small E_J regime for $r = 0.8$, $b = 1$, $E_J/E_{CN} = 0.5$, $k_B T/E_{CN} = 3 \cdot 10^{-2}$, $R_N/R_K = 10$. Charge skipping occurs for $r > \frac{1}{2\sqrt{b}}$ (dotted line in the inset).

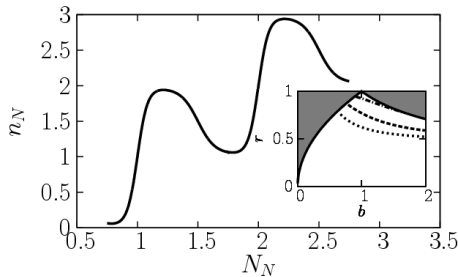


FIG. 4: Coulomb staircase in the large E_J regime for $E_J/E_{CN} = 2$ (the other parameters are the same as in Fig. 3). The constraint for charge skipping depends on E_J . The inset shows the minimum r values for $E_J/E_{CN} = 1$ (dotted), $E_J/E_{CN} = 2$ (dashed), and $E_J/E_{CN} = 4$ (dash-dot).

For the parameters indicated in the caption of Fig. 3, a positive step (where the charge number $n_N = 1$ is skipped) and a consecutive negative step are stabilized. Notice that contrary to the usual staircase, where all real transitions between n and $n \pm 1$ can be treated by the same master equation [11], here the rates are of higher order and the virtual states involved in one transition (positive step) become real states for the next (negative) one, preventing from such a simple treatment. A full treatment is at least similar in complexity to a master equation treatment of cotunneling in single dots [12] and is beyond the scope of this Letter.

Let us now turn to the case of a large Josephson energy, $E_J > E_{CS} = e^2/2C_{\Sigma S}(1-r^2)$. Then one can rely on an adiabatic assumption [9], and setting the phase difference to be ϕ across the JJ, one can solve the Hamiltonian (2) neglecting the normal electron tunneling term. The adiabatic Hamiltonian $H_{ad} = E_C - E_J \cos \phi$ describes a Cooper pair box with an effective gate voltage, which is an adiabatic function of

n_N . In the tight-binding limit $E_J/E_{CN} \gg b$, assuming that the junction dynamics is confined to the lowest Bloch-band, one obtains:

$$H_{ad} = E_{CN}(1-r^2)(n_N - N_N)^2 - \Delta_0 \cos\left[2\pi\left(N_S - \frac{r}{2\sqrt{b}}(n_N - N_N)\right)\right], \quad (3)$$

where the bandwidth is given by

$$\Delta_0 = 16\sqrt{\frac{2}{\pi}} b E_{CN} \left(\frac{E_J}{2bE_{CN}}\right)^{3/4} e^{-\sqrt{8E_J/bE_{CN}}}. \quad (4)$$

The second term in H_{ad} represents an effective screening potential acting on the charge in N. Choosing N_S which controls the phase of the cosine term, one can achieve a negative curvature of H_{ad} , seen as an effective charging energy E_{CN}^{eff} for the gauged charge in N, $n_N - N_N$. A necessary condition for this is $\frac{\pi^2}{2} \frac{r^2}{b(1-r^2)} \Delta_0 > 1$, yielding the frontier lines in the inset in Fig. 4.

One notices that a large E_J puts a strong constraint on the coupling capacitance C_0 , requiring values of r closer to one than in the case of small E_J . If this is satisfied, one can calculate the shape of the charge skipping and negative steps using a master equation based on transition rates between charge states $n_N = 0, 2$ or $n_N = 2, 1$, respectively. The adiabatic transition rates are given by $\Gamma_{ad} = \frac{\delta E_{CN}^{eff}}{e^2 R_N} [\exp(\delta E_{CN}^{eff}/k_B T) - 1]^{-1}$. The corresponding steps are shown in Fig. 4, and are flatter than in the small E_J case.

To operate in the Coulomb blockade regime, the temperature must be sufficiently low to suppress thermal excitations. The energy difference between two charge states depends on r - a larger r facilitates charge skipping. However, a too strong coupling spoils it since the system virtually becomes one island and the energy no longer depends on the location of the charge. For these reasons, r should be close to 0.75 (for $b = 1$) for small Josephson energies. In this case, the requirement for Coulomb blockade is $k_B T < E_{CN}/4$. The factor 1/4 is due to the doubling of the number of steps and the charging energies' quadratic dependence on charges and gate charges. In the step calculations, the value $r = 0.8$ was used to accommodate for both the small and large Josephson energy cases. Furthermore, a temperature of $T \sim 30$ mK and a typical charging energy of $E_{CN} \sim 1 \cdot 10^{-4}$ eV were used. For the symmetric case where $b = 1$, this charging energy gives $C_N = C_S \sim 2$ fF. Furthermore, if we assume, e.g., $C_{gN} = C_{gS} = 0.02$ fF, then the gate charges $N_S = 0.5$ and $N_N = 0.75 - 2.75$ correspond to $V_{gS} = 4$ mV and $V_{gN} = 6 - 22$ mV, respectively. The value of r chosen for the calculations corresponds to $C_0 = 4C_N = 8$ fF. The second requirement for Coulomb blockade is that the tunnel resistance R_N must be larger than the resistance quantum $R_K = h/e^2 \approx 25.8$ k Ω . In the calculations of the transition rates, $R_N/R_K = 10$ was used. This gives a first order tunneling rate of $\Gamma \sim 10^9$ s $^{-1}$. The second and third order tunneling rates are 10^7 s $^{-1}$ and $5 \cdot 10^3$ s $^{-1}$, respectively.

Let us briefly discuss the issue of phase coherence in such a strongly coupled NS set-up. As shown above, charge skip-

ping only requires that pair tunneling occurs between the superconducting reservoir and the S island in order to screen the repulsive interaction in the normal grain. No phase coherence is needed, as shown by the first calculation where the Josephson tunneling is treated perturbatively. Here, charge skipping and NDCA are a result of pair fluctuations in S. As a backaction effect, charge fluctuations in N should strongly act upon S and reduce the phase coherence. A treatment of this goes beyond the adiabatic approximation made in the large E_J case. One can anticipate that corrections to the adiabatic behavior can cause substantial fluctuations in the phase ϕ , renormalizing E_J to a smaller value. In this sense, the small- E_J case is generic. The study of such feedback effects between strongly coupled N and S islands is worth interest, but is beyond the scope of this work.

Let us now comment on the relationship between charge skipping and proximity effect. The latter manifests the onset of pairing correlations in a metal, despite the absence of a pairing potential, due to Cooper pair diffusion. Here, in the absence of any tunneling of electrons between N and S, no phase coherence can be established whatsoever in N. Charge skipping indicates instead a local attractive (negative-U) potential capacitively induced in N. Then, adding a small tunneling term T_{NS} between N and S opens the possibility of establishing a true phase coherence between states $n_N, n_N + 2$. Such a proximity effect could be studied here in a quite unusual regime, where $T_{NS} < |U|$. More generally, the occurrence of an attractive correlation in a metallic dot has interesting consequences, some of them having been theoretically explored in the context of molecules with polaronic behavior, like pair tunneling [10, 13], or the possibility of a charge Kondo effect [14] in the coherent regime of tunneling between N and R. Another application of the mechanism proposed in this Letter consists in inducing an attractive correlation between excess

charges in two or more neighboring normal dots capacitively coupled to the same S island. Such a device could be useful in quantum information processing based on the charge [15] or spin [16] degree of freedom of individual electrons in normal quantum dots.

To finish, we propose a scheme for detecting a capacitively induced attractive correlation in a normal metallic grain. The goal is to detect the non-monotonous charging of the N grain. SETs or point contacts [17] provide very sensitive detection of the local change in the electrostatic potential (rather than the charge). In double-dot setups with weak mutual coupling, the potential fluctuations in each dot can be measured by a different neighboring point contact [18]. In the present case, placing a point contact close to N does not measure δn_N , but instead $\delta V_N = e(C^{-1})_{NN}(e\delta n_N) + 2e(C^{-1})_{NS}(\delta \bar{n}_S) = \frac{e}{C_{\Sigma N}(1-r^2)}[\delta n_N + 2r\sqrt{b}\delta \bar{n}_S]$. If $r\sqrt{b} > \frac{1}{2}$, doubling of the number of steps can be detected, but not the non-monotonous charging curve. To access the latter, it is necessary to measure $\delta V_S = \frac{e}{C_{\Sigma N}(1-r^2)}[r\sqrt{b}\delta n_N + 2b\delta \bar{n}_S]$ as well, with a second point contact close to S, and reconstruct $\delta n_N = \frac{C_{\Sigma N}}{e}[\delta V_N - \frac{r}{\sqrt{b}}\delta V_S]$. The parameters $C_{\Sigma N}, r, b$ can be easily measured from a honeycomb diagram obtained in the normal (non-superconducting) state in the presence of very weak tunneling between N and S. A setup adapting that of Ref. 18 is proposed in Fig. 1, involving a superconducting strip with a Cooper pair box, coupled laterally to a 2DEG. Notice that the direction of charge transfer also can be measured [19], and that other experimental access to the correlation between charge fluctuation in N and S could be obtained from shot noise measurements, as in Ref. 20.

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