Emergence of a negative charging energy in a metallic dot capacitively coupled to a superconducting island

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We consider the hybrid setup formed by a metallic dot, capacitively coupled to a superconducting island S connected to a bulk superconductor by a Josephson junction. Charge fluctuations in S act as a dynamical gate and overscreen the electronic repulsion in the metallic dot, producing an attractive interaction between two additional electrons. As the offset charge of the metallic dot is increased, the dot charging curve shows positive steps ($+2e$) followed by negative ones ($-e$) signaling the occurrence of a negative differential capacitance. A proposal for experimental detection is given, and potential applications in nanoelectronics are mentioned.

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At low temperatures, the electronic transport through point-like metallic nanostructures (quantum dots) is dominated by the electronic Coulomb *repulsion* between additional electrons. When a small-capacitance dot is weakly coupled to a normal metallic reservoir, the average number of charges in the dot, n_N, increases one by one with the gate voltage V_{gN}, leading to conductance peaks [1]. This Coulomb blockade phenomenon has recently enabled an individual control of charge or spin, for instance in view of quantum information protocols [2, 3] . We address here the possibility of inverting the sign of the charging energy. Indeed, creating a *negative* effective charging energy in a normal (non-superconducting) metallic dot would induce attractive correlations, triggering for instance pair tunneling from/to a normal reservoir [4] or a charge Kondo effect [5, 6], or giving rise to super-Poissonian shot noise [6, 7]. Going beyond the single dot case, attractive correlations between electrons in spatially *separated* dots could help implementing quantum information protocols involving two-qubit gates, when the qubits are carried by the charge (spin) of the last added electron. Historically, attractive interactions in the solid state are known as valence-skipping states [8], and negative-U centers [9]. Another possible mechanism for electronic attraction is mediated by optical phonons, binding two electrons as a bipolaron in confined geometries for strong electron-phonon coupling and lattice polarizability [10]. Due to the low polarizability and small effective carrier mass, bipolarons are unlikely to form in a clean GaAs/AlGaAs two-dimensional electron gas (2DEG), although they might do so in presence of a few donor impurities [11], or with more polarizable multilayer materials [12]. Molecular junctions are also promising for achieving a negative charging energy [4, 13].

In the present Letter, we propose an alternative mechanism, pointing out that the repulsive charging energy in a metallic dot (N) connected to a normal reservoir (Fig. 1) can be turned into an attractive one when N is *capacitively* coupled to a superconducting island (S). The latter is connected to a superconducting reservoir by a Josephson junction (JJ) and operates in the Cooper pair box regime, e.g. it fluctuates between two pair number states [14, 15]. Here we assume that electron tunneling between S and N is negligible; therefore no proximity effect occurs in the N dot. We instead focus on the charging properties of the N grain as its gate voltage is varied. The S island acts as an effective dynamical gate, whose effects turn out to be nonlinear. As the main result of this Letter, the Coulomb charging energy in N can be overscreened by the neighboring pair fluctuations in S, and an effective *local attraction* appears between electrons added into N. As a corollary, certain charge states are "skipped" as the N gate voltage is varied. The resulting charging curve becomes non-monotonous, displaying positive steps ($+2e$) followed by *negative* ones ($-e$). A related screening effect was proposed by Averin and Bruder for controlling the coupling between two superconducting charge qubits [16]. Notice that if N were coupled to both drain and source reservoirs, our set-up would be similar to a Cooper pair box coupled to a single-electron transistor (SET). The latter has been studied in great detail as a read-out device for a superconducting (charge) qubit embodied in the S island [17]. In this case, the capacitive coupling between N and S must be small, in order to minimize the decoherence due to backaction of the normal part of the device onto the superconducting one, whereas in our proposal, the coupling is very strong.

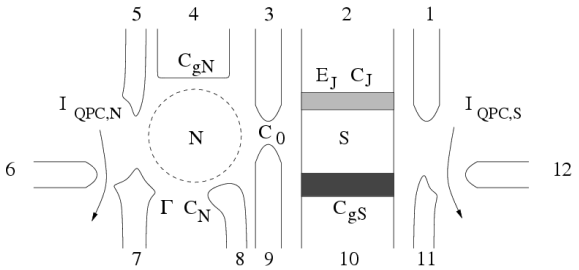

FIG. 1: Schematic view of a normal grain (N) coupled with a strong capacitive coupling (controlled by 3, 9) to a Cooper pair box composed of a Josephson junction connecting superconducting reservoir 2 and island, and gate 10. Electrons tunnel between N and its reservoir (defined by 7, 8). Detection is 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 JJ connecting the S island to the reservoir has a Josephson energy E_J and capacitance C_J, and a gate imposes an offset $\nu_S = C_{gS}V_{gS}/e$, with $C_{gS} \ll C_J$. Similarly, the N island is connected to a normal reservoir by a tunnel junction, with single-electron tunneling rate Γ and capacitance C_N, and experiences a gate offset $\nu_N = C_{gN}V_{gN}/e$, 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 assume the superconducting gap in S to be larger than the charging energy, such that only even charge number states n_S occur in S. At low temperatures, quasiparticle tunneling in S can be neglected. Defining $C_{\Sigma S} = C_J + C_0 + C_{gS}$ and $C_{\Sigma N} = C_N + C_0 + C_{gN}$, and introducing the parameters $b = C_{\Sigma N}/C_{\Sigma S}$ and $r = C_0/\sqrt{C_{\Sigma N}C_{\Sigma S}}$, the total charging energy of the NS system can be written as [18]

$$E_C = E_{CN}[(n_N - \nu_N)^2 + b(n_S - \nu_S)^2 + 2r\sqrt{b}(n_N - \nu_N)(n_S - \nu_S)] \qquad (1)$$

with $E_{CN} = e^2/[2C_{\Sigma N}(1 - r^2)]$. The asymmetry parameter b and the coupling parameter $r < 1$ are not independent, and $r < \min(b, 1/\sqrt{b})$. Eq. (1) determines the charge stability diagram of the isolated NS system in the (ν_N, ν_S) plane. First, for a value ν_S imposing an integer number of pairs in S, say $\nu_S = 2$, the charging number n_N increases monotonously with ν_N. Next, consider a case where n_S fluctuates, for instance $\nu_S = 1$. For small r, as shown in Fig. 2a, n_N is again a monotonous function of ν_N: the sequence of charge states (n_N, n_S) as ν_N increases reads $(0,0), (0,2), (1,0), (1,2), (2,0), (2,2), \ldots$ (notice the oscillation of n_S). The result is very different if r is large. In fig. 2(b), for $\nu_S = 1$, n_N increases with ν_N but in a non-monotonous way, the charge state sequence being $(1,0), (0,2), (2,0), (1,2), (3,0), (2,2)$, etc. The corresponding charging staircases are plotted in the insets.

One sees that the transition from $(n_N, 2)$ to $(n_N + 2, 0)$ at $\nu_N = n_N + 1$ "skips" the charge state $n_N + 1$ in the grain. This signals a negative effective charging energy in N which overcomes the Coulomb repulsion. After increasing by two units, n_N decreases by one unit, yielding a negative differential capacitance (NDCA) $C_{diff} = C_{gN}(dn_N/d\nu_N)$ at half-integer values of ν_N. Strikingly, the total number of steps, positive or negative, is doubled with respect to the usual case. Both charge skipping and NDCA occur above the dotted line indicated in Fig.4 displaying a (b, r) diagram. From the above charging energy, an effective attractive potential $U < 0$ can be estimated for $\nu_S = 1$ as $U = E_C(0,2) + E_C(2,0) - 2E_C(1,2) = e^2(1 - 2r\sqrt{b})/[C_{\Sigma N}(1 - r^2)]$. The necessary condition for the occurrence of a negative charging energy is thus $2r\sqrt{b} > 1$.

To further analyze this possibility in an open NS system, let us consider the full Hamiltonian:

$$H = E_C + \sum_{k\sigma} \varepsilon_k \, c^\dagger_{kR,\sigma} \, c_{kR,\sigma} + \sum_{q\sigma} \varepsilon_q \, c^\dagger_{qN,\sigma} \, c_{qN,\sigma} + [\sum_{kq\sigma} T_{k,q} \, c^\dagger_{kR,\sigma} \, c_{qN,\sigma} - \frac{E_J}{2} \, |n_S + 2\rangle\langle n_S| + \text{H.c.}] \, , \qquad (2)$$

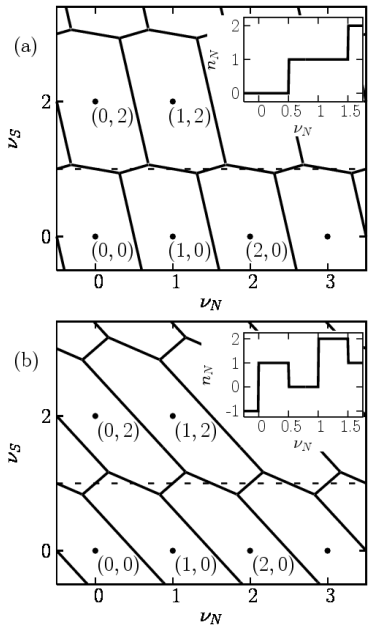

FIG. 2: Charge stability (or honeycomb) diagram for $b = 1$, $r = 0.2$ (a) and $r = 0.8$ (b). The insets show the charging curves for N, taken along the dotted line. In case (b) the charging staircase (inset) exhibits charge skipping effects.

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 expressed as $n_N = \sum_{q\sigma} c^\dagger_{qN,\sigma} \, c_{qN,\sigma}$. Assuming constant densities of states in N and R, the single-electron transition rate from R to N is given by $\Gamma^{(+1)} = [\delta E_C^{(+1)}/e^2 R_N] \, [\exp(\delta E_C^{(+1)}/k_B T) - 1]^{-1}$ within the golden rule approximation, where R_N is the tunnel resistance.

Considering first the case of small $E_J \ll E_{CS} = e^2/[2C_{\Sigma S}(1 - r^2)]$, we perform a T-matrix calculation of the transition rates from $(0,2)$ to $(2,0)$ (close to $\nu_N = 1$) and from $(2,0)$ to $(1,2)$ (close to $\nu_N = 1.5$). For the first transition, we take into account three possible configuration paths involving higher-energy states: $(0,2) \to (1,2) \to (2,2) \to (2,0)$, $(0,2) \to (1,2) \to (1,0) \to (2,0)$, and $(0,2) \to (0,0) \to (1,0) \to (2,0)$. For the second transition, only one excited state is involved: $(2,0) \to (1,0) \to (1,2)$ and $(2,0) \to (2,2) \to (1,2)$. The shape of each step is calculated at finite temperature by solving the master equation governing the dynamics of the probabilities $p(0,2)$, $p(2,0)$ for the positive step and $p(2,0)$, $p(1,2)$ for the negative one. The master equation reads $\dot{p}(a) = \Gamma^{b\to a} p(b) - \Gamma^{a\to b} p(a)$ with $p(b) = 1 - p(a)$ for the states a, b involved in the transition. Here the probabilities of other states are neglected, which is justified close to $\nu_N = 1$ or $\nu_N = 1.5$ and if the steps are sufficiently narrow. The resulting steps are shown in Fig. 3.

As a result, a positive step $+2e$ (where the charge number $n_N = 1$ is skipped) and a consecutive negative step $-e$ are stabilized. Notice that contrary to the usual case where all transitions between n and $n \pm 1$ are real and obey the same master equation [19], here the rates are of higher order and

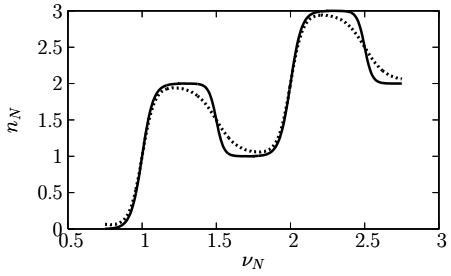

FIG. 3: Charging staircase : $r = 0.8$, $b = 1$, $E_{CS} = E_{CN}$, $k_BT/E_{CN} = 3 \cdot 10^{-2}$, $R_N/R_K = 10$; (full line) $E_J/E_{CN} = 0.5$; (dotted line) $E_J/E_{CN} = 2$.

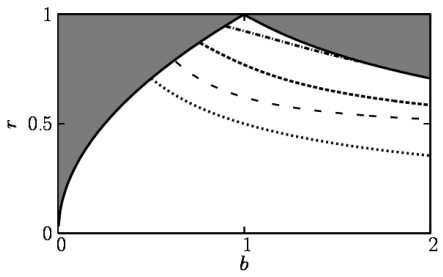

FIG. 4: Phase diagram in the b, r plane. The unphysical grey region is excluded. Charge skipping and NDCA occur above the dotted line, from bottom to top : $E_J/E_{CN} = 0$, and $1, 2, 4$ (adiabatic calculation). All other parameters are the same as in fig. 3.

the virtual states involved in one transition become real states (with first order rate) for the next one. A full treatment of all processes is beyond the scope of this Letter.

Let us now turn to the case of a large Josephson energy, $E_J > E_{CS}$. The adiabatic assumption [16], setting the phase difference to ϕ across the JJ, allows to solve the Hamiltonian (2), neglecting the normal electron tunneling term. The resulting 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 the sum of the N dot charging energy and the adiabatic Bloch band energy :

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

where the bandwidth is given by [14]

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

The second term in H_{ad} represents a nonlinear screening potential acting on the charge in N. The offset ν_S controls the phase of the cosine term, and an appropriate choice (for instance $\nu_S \approx 1$) achieves a negative curvature of H_{ad}, viewed as an effective charging energy E_{CN}^{eff} for the gauged charge in N, $n_N - \nu_N$. The required condition reads $(\pi^2/2)r^2\Delta_0/[b(1-r^2)] > 1$, yielding the lines in Fig. 4. Clearly, a large E_J puts a stronger constraint on the coupling capacitance C_0, and requires larger values of r than for small E_J. The shape of the charge skipping and negative steps is then calculated, using a master equation based on charge states $n_N = 0, 2$ or $n_N = 2, 1$, respectively. The adiabatic transition rates are given by $\Gamma_{ad} = [\delta E_{CN}^{eff}/e^2R_N]\,[\exp(\delta E_{CN}^{eff}/k_BT) - 1]^{-1}$. The corresponding steps (Fig. 3) are less pronounced than in the small E_J case.

Searching the optimum regime must account for the fact that for too large r values, the system behaves like one single island and its energy no longer depends on the location of the charge. The Coulomb blockade requires temperatures much smaller than the energy difference between two charge states. An optimal r is close to 0.75 (with $b = 1$) for small Josephson energies, and the requirement for Coulomb blockade reads $k_BT < E_{CN}/4$. The value $r = 0.8$ was used in the steps calculations. A temperature of $T \sim 30$ mK and a typical charging energy of $E_{CN} \sim 0.1$ meV were used, yielding $C_N = C_J \sim 2$ fF and $C_0 = 4C_N = 8$ fF. The tunnel resistance R_N is chosen as $R_N/R_K = 10$ with $R_K = h/e^2 \approx 25.8$ kΩ. This yields a bare single-electron tunneling rate of $\Gamma \sim 10^9$ s^{-1}. In presence of the S island, the effective tunneling rates are 10^7 s^{-1} (n_N decreasing from 2 to 1) and $5 \cdot 10^3$ s^{-1} (n_N increasing from 0 to 2), respectively. With the above parameters, the maximum attraction $|U|$ is of the order of 50μeV.

Notice that such a negative charging energy cannot render the dot superconducting, because it concerns the energy required to add one or two electrons on the dot, rather than a true attractive potential felt by all electrons near the Fermi level. The effective "negative-U" potential manifests itself only when the dot is weakly coupled to a reservoir such that its charge can fluctuate. This is similar to a single "$-U$" center weakly hybridized with a normal bulk metal [9]. Here, pair fluctuations with the reservoir are assumed to be incoherent. On the contrary, a very small tunneling term T_{NS} between N and S would allow for a true phase coherence between states $n_N, n_N + 2$, thus a proximity effect in a quite unusual regime, where $T_{NS} < |U|$.

Let us briefly discuss the issue of phase coherence in the Cooper pair box. Charge skipping only requires pair tunneling 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 our calculation in the small E_J case. Moreover, even in the large E_J case, charge fluctuations in N should strongly react back upon S and reduce the phase coherence. A full treatment goes beyond the adiabatic approximation [16]. One can anticipate that large fluctuations in the phase ϕ renormalize E_J downwards, making the small-E_J case generic.

We now propose a scheme for detecting the non-monotonous charging of the N grain. SETs, or point contacts [20] provide very sensitive detection of the local change in the electrostatic potential. In double-dot setups with weak mutual coupling, the potential variations in each dot can be measured by a different neighboring point contact [21]. In the present case, placing a point contact close to N does not measure δn_N, but instead $\delta V_N = (C^{-1})_{NN}(e\delta n_N) + (C^{-1})_{NS}(e\delta n_S) =$

$e\,[\delta n_N + r\sqrt{b}\,\delta n_S]/[C_{\Sigma N}(1-r^2)]$. If $2r\sqrt{b} > 1$, doubling of the number of steps can be detected, but not the non-monotonous charging curve. To access the latter, it is suitable to measure $\delta V_S = e\,[r\sqrt{b}\,\delta n_N + b\,\delta n_S]/[C_{\Sigma N}(1-r^2)]$ as well, with a second point contact close to S, and reconstruct $\delta n_N = C_{\Sigma N}\,[\delta V_N - r\delta V_S/\sqrt{b}]/e$. The parameters $C_{\Sigma N}, r, b$ can easily be measured from the stability diagram obtained in the normal state in the presence of a very weak tunneling between N and S [18]. Notice that the tunneling rates calculated above are much reduced compared to the bare single-electron rate Γ. Therefore the use of point contacts permits, non only a time-averaged [21], but even a time-resolved and directional [22] detection of the charge variations in N and S. On the other hand, cross-correlation shot noise measurements, as in ref. [23], would require higher currents. In practice, a possible setup inspired by ref. [21] is proposed in Fig. 1. It involves a superconducting strip with a Cooper pair box, coupled laterally to an InGaAs/AlGaAs 2DEG, suitably tuning the Schottky barrier present at the interface between the superconductor and the 2DEG. Notice that the geometry of Fig. 1 could be modified, including drain and source such as to allow transport through N [4].

In conclusion, we have proposed a mechanism inducing a controllable negative charging energy, thus attractive correlations, in one or several metallic dots. We believe that such an effect would be useful in view of more complex nanoelectronics devices. The authors are grateful to T. Martin, M. Fogelström, and G. Johansson for useful discussions, and S. Andergassen for careful reading of the manuscript. D. F. and A. Z. were partially supported by the contract AC NANO NR0114, and C. H. was supported by the Swedish Research Council (VR) under grant 621-2006-3072.

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