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Quantum Coherence with a Single Cooper Pair

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Abstract

A metallic electrode connected to electron reservoirs by tunnel junctions has a series of charge states corresponding to the number of excess electrons in the electrode. In contrast with the charge state of an atomic or molecular ion, the charge states of such an "island" involve a macroscopic number of conduction electrons of the island. Island charge states bear some resemblance with the photon number states of the cavity in cavity QED, the phase conjugate to the number of electrons being analogous to the phase of the field in the cavity. For a normal island, charge states decay irreversibly into charge states of lower energies. However, the ground state of a superconducting island connected to superconducting reservoirs can be a coherent superposition of charge states differing by two electrons (i.e. a Cooper pair). We describe an experiment in which this Josephson effect involving only one Cooper pair is measured.

1. Introduction

As shown by several beautiful experiments reported in this volume, the combined quantum states of individual atoms or ions in an electromagnetic cavity and the photons of the cavity can be manipulated and controlled to a high degree of accuracy. These atomic physics systems open the door to the practical construction of complex entangled quantum states of the type on which the theory of quantum computers is based. The question naturally arises as to whether there exists also solid state electronic systems exhibiting quantum states amenable to such manipulations. With the elaborate nanofabrication techniques now available, a large number of coupled electronic devices can be produced which could in principle be well suited for practical implementations of the schemes of quantum computing. However, the flexibility of electronic systems regarding connections between individual elements is also accompanied by severe drawbacks. Although at the microscopic level, most electronic devices in practical applications are based on quantum properties of electrons, electronic degrees of freedom that can be probed externally such as voltages and currents almost never behave quantum mechanically. These variables, which play the role of, for instance, the atomic dipole moment in cavity QED [1], are usually so strongly damped that their quantum decoherence time is much shorter than the time window of experiments.

A notable exception is encountered in superconducting tunnel junction circuits for which, in principle, very low internal electric dissipation can be achieved. In the early 80's, A. J. Leggett [2] already remarked that a superconducting ring interrupted by a tunnel junction (a so-called RF-SQUID) could, under certain experiment conditions, behave as a quantum two-level system analogous to the ammonia molecule. The two "resonant" states for this particular circuit correspond to two opposite values of the magnetic flux threading the ring. Although practical experiments have been proposed [3] to observe the coherent quantum tunneling between these two degenerate flux states, only irreversible tunneling out of a single metastable flux state has been observed in the RF-SQUID [4] and in the related current-biased Josephson junction system [5]. The observation of macroscopic quantum coherence in the RF-SQUID is mainly hindered by the difficulty to control precisely the external flux on the device, which is the key parameter determining the degenerescence of flux states.

More recently, experiments [6–9] have shown that, in contrast to flux states of a superconducting ring which are easily perturbed by the electromagnetic environment, the charge states of a superconducting island connected to the rest of the circuit by tunnel junctions and capacitors might sufficiently be well decoupled from external influences to allow long-lived macroscopic quantum superposition of states. In this article, we report experimental results showing how a quantum superposition of charge states can be prepared in the simplest superconducting island circuit, namely the superconducting box.

2. Theoretical description of the superconducting box

The superconducting box circuit (see Fig. 1) is a simplified version of a circuit first considered by M. Buttiker [10] for Bloch oscillations in superconducting tunnel junctions [11]. It consists of a single superconducting island connected to a superconducting electron reservoir by a tunnel junction with capacitance C_j. Electrons can be transferred from the reservoir to the island by a voltage source U connected between the reservoir and the island via a gate capacitance C_g. Both the island and the reservoir are taken to be good

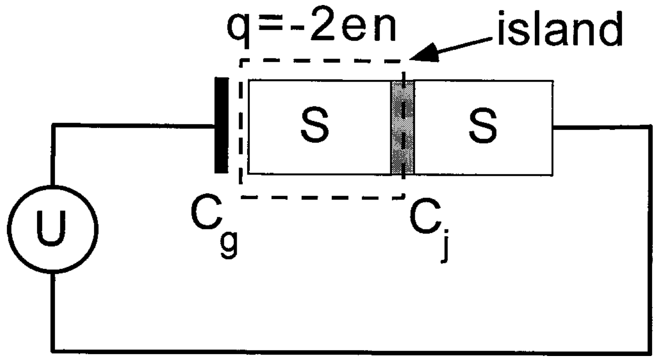

Fig. 1. Schematics of the single Cooper box: a superconducting electrode (island) is in contact with a superconducting reservoir though a tunnel junction (grey zone) with capacitance C_j. Excess Cooper pairs tunnel onto the island in response to an electric field applied by means of the gate capacitance C_g and voltage U.

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BCS superconductors with a gap Δ much larger than the energy $k_B T$ of thermal fluctuations or the Coulomb energy $e^2/2C_\Sigma$ of the island ($C_\Sigma = C_g + C_j$ is the total island capacitance). Under these conditions, we can consider that all electrons in the island are paired [12]. The only remaining degree of freedom of the island is its total number of excess Cooper pairs n which is related to the total charge q of the island by $q = -2en$. The variable n, like the number of photons in a cavity in cavity QED [1], is discrete. However it can take negative as well as positive integer values, $n = 0$ corresponding to an electrically neutral island. The number n can fluctuate quantum-mechanically since Cooper pairs can tunnel in and out the island by Josephson tunneling. We must therefore describe it by an operator $\hat{n}$. We can take as a convenient basis for the charge states of the island the eigenvectors of $\hat{n}$:

$$\hat{n}|n\rangle = n|n\rangle. \tag{1}$$

Using this basis, we can write the electrostatic part of the hamiltonian as

$$\mathscr{H}_{\mathrm{el}} = E_C \sum_n (n - n_g)^2 |n\rangle\langle n| \tag{2}$$

where $E_C = (2e)^2/2C_\Sigma$ is the Coulomb energy of an extra Cooper pair on the island for zero gate voltage and $n_g = C_g U/(2e)$ the dimensionless gate voltage.

The Josephson coupling hamiltonian has the form:

$$\mathscr{H}_J = -\frac{E_J}{2} \sum_n (|n\rangle\langle n+1| + |n+1\rangle\langle n|) \tag{3}$$

where the Josephson energy E_J is macroscopic in the sense that it is proportional to the area of the tunnel junction. This energy can be expressed in terms of the superconducting gap and the tunnel junction conductance G_T in the normal state by the Ambegaskar-Baratoff zelation

$$E_J = \frac{hG_T}{8e^2}\Delta. \tag{4}$$

In absence of the Josephson hamiltonian, the energies of the states of the system are given by a set of parabola shown as dashed curves in Fig. 2(a). The Josephson hamiltonian lifts the degeneracy at the crossings of the parabola and for the case $E_J \ll E_C$ we get the avoided crossings shown on Fig. 2(a). This is the case which we consider in this article. In the opposite limit where $E_J \gg E_C$, the fluctuations of n are so large that we recover the usual situation considered in the conventional description of the Josephson effect, where the phase of the island is the good-quantum number.

At temperatures such that $k_B T \ll E_C$, we can limit our analysis to the two states with the lowest energy. Because of the periodicity of the system with respect to the addition of an extra Cooper pair, we can restrict the dimensionless gate voltage to vary in the interval $0 < n_g < 1$. We can then work in a Hilbert space spanned with the vectors $|0\rangle$ and $|1\rangle$.

In this space, the total hamiltonian $\mathscr{H} = \mathscr{H}_{\mathrm{el}} + \mathscr{H}_J$ is represented by the following matrix:

$$\mathscr{H} = \frac{1}{2}\begin{bmatrix} -E & -E_J \\ -E_J & E \end{bmatrix}. \tag{5}$$

The difference $E = E_C(1 - 2n_g)$ between the electrostatic energy of the two states depends linearly on the gate voltage. The trace of the matrix has been nulled out by an

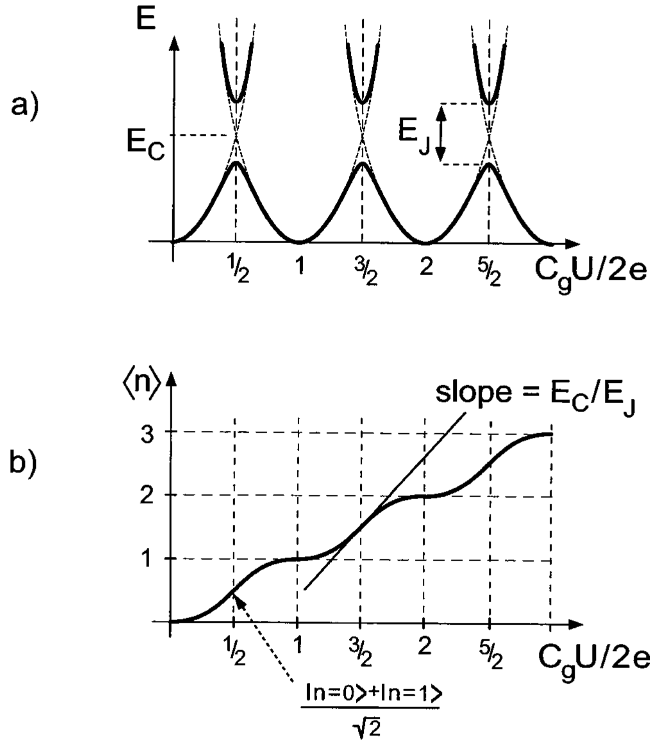

Fig. 2. (a) Electrostatic energy of the single Cooper box circuit of Fig. 1 as a function of U, for several values of the number n of excess Cooper pairs in the island (parabolas in dotted line, $n = 0$–3). In full line, total energy of the box including the effect of Cooper pair tunneling through the junction. (b) Predicted value of the average of n in the ground state of the box, as a function of U.

adequate choice of the zero of energy $E_0 = E_C(1/2 - n_g)^2$. One can thus make a correspondence between the Cooper pair box and a spin $\frac{1}{2}$ in a magnetic field using the Pauli spin matrices:

$$\sigma_x = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \quad \sigma_y = \begin{bmatrix} 0 & i \\ -i & 0 \end{bmatrix}, \quad \sigma_z = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}. \tag{6}$$

The hamiltonian takes the form

$$\mathscr{H} = -\boldsymbol{s} \cdot \boldsymbol{h} \tag{7}$$

where $\boldsymbol{s} = \frac{1}{2}\boldsymbol{\sigma}$ is the spin operator and $\boldsymbol{h}$ an effective magnetic field whose components in the bass (x, y, z) are $[E_J, 0, E]$ (see Fig. 3).

In this correspondence the average z-component of the spin gives the average charge of the island:

$$\langle n\rangle = \tfrac{1}{2} + \langle s_z\rangle. \tag{8}$$

Here the average $\langle O\rangle$ of an operator O is given by the usual expression

$$\langle O\rangle = \mathrm{tr}\,[\exp(-\beta\mathscr{H})O] \tag{9}$$

adequate for thermal equilibrium at a temperature $T = (k_B\beta)^{-1}$.

By simple geometrical considerations (see Fig. 3) one obtains directly

$$\langle s_z\rangle = \tfrac{1}{2}\tanh(\beta|\boldsymbol{h}|/2)\cos\theta \tag{10}$$

where θ is the angle between the field $\boldsymbol{h}$ and the z axis. The average Cooper pair number on the island is thus

$$\langle n\rangle = \frac{1}{2}\left[1 + f\left(\frac{E_C(2n_g - 1)}{E_J}, \beta E_J/2\right)\right] \tag{11}$$

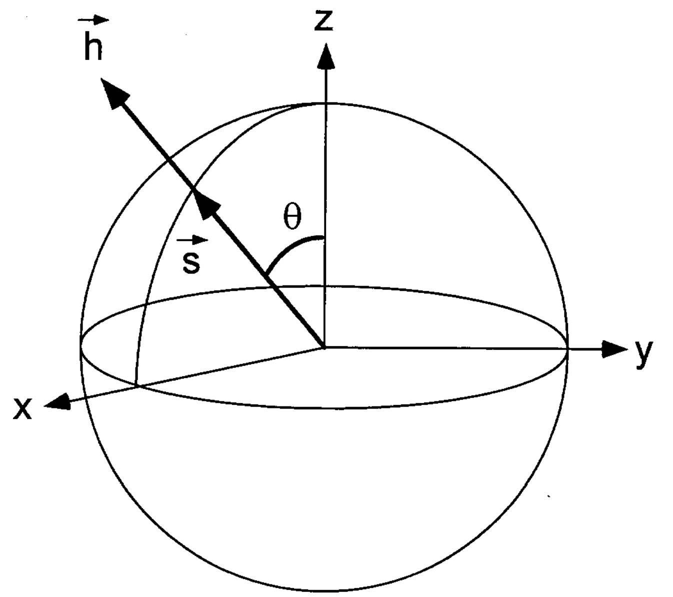

Fig. 3. Geometric representation of the effective spin s associated with the quantum state of the Cooper pair box. Here, the spin is represented in its ground state, i.e. pointing in the direction of the effective field $\boldsymbol{h}$ associated with the combination of the electrostatic field due to U and Cooper pair tunneling.

where the dimensionless rounded-step function $f(\eta, \kappa)$ is given by

$$f(\eta, \kappa) = \frac{\eta \tanh(\kappa\sqrt{1+\eta^2})}{\sqrt{1+\eta^2}}. \tag{12}$$

At the lowest temperatures, when $k_B T \ll E_J$, the system will stay in its ground state for all values of n_g. When taking into account the periodicity of the system with respect to the addition of a Cooper pair, we get for the variations of $\langle n \rangle$ as a function of n_g the staircase shown on Fig. 2(b). At half-integer values of n_g, the slope of the staircase goes through a maximum and takes the value E_C/E_J. At these points, the system is in coherent superposition, with equal weight, of two charge states differing by one Cooper pair. When the temperature increases, this maximum slope decreases and in the regime $E_J \ll k_B T \ll E_C$, takes the value $E_C/k_B T$. The quantum superposition of charge states due to Josephson tunneling thus manifests itself in the fact that the slope of the staircase, which can be measured by a very sensitive electrometer, remains finite as $T \to 0$.

Can a coherent superposition of two different charge states be observed in "real-world" experiments, in which the charge degrees of freedom are coupled to a dissipative electromagnetic environment, which includes the perturbative action of the measuring electrometer? The influence of the electromagnetic environment on single Cooper pair tunneling in the particular case of the superconducting box has been calculated by Neumann *et al.* [13] and we present here their results which are relevant for our experiments.

The electromagnetic environment can be modeled as a series impedance $Z(\omega)$ placed between the voltage source and the gate capacitance. It can be shown [14] that the circuit is equivalent to a pure Josephson element in series with an effective impedance $Z_t(\omega)$ given by:

$$Z_t(\omega) = \frac{\lambda^2}{i\lambda C_j \omega + Z^{-1}(\omega)}$$

where

$$\lambda = \frac{C_g}{C_g + C_j}. \tag{13}$$

In our box experiment the factor λ^2 is smaller than 10^{-2} and the effective series impedance $Z(\omega)$ has a modulus of the order of the vacuum impedance $377\,\Omega$. This results in an effective impedance $Z_t(\omega)$ several orders of magnitude smaller than the resistance quantum $R_K = h/e^2$.

In this low impedance limit, the perturbational calculation of the complex energy shift of the excited state relative to the ground state gives:

$$\Delta\varepsilon = \frac{E_J^2}{\varepsilon} \frac{Z_t(\omega = \varepsilon/\hbar)}{\pi R_K} \tag{14}$$

where ε is the energy difference between the ground and excited state. The quality factor $|\Delta\varepsilon|/\varepsilon$ of the transition between the two states is thus well above 10^3 in our experiments and the influence of the environment can be neglected in the analysis of the effect of quantum fluctuations on the average value of the number of Cooper pairs.

3. Experimental results

We have measured the number of excess Cooper pairs in the island by coupling it electrostatically to a single electron transistor (SET) used as an electrometer [15]. The electrometer measures a time-averaged value $\bar{n}$ which we take equal to the thermal average value $\langle n \rangle$ by assuming ergodicity. Since a SET electrometer has a relatively low cut-off frequency (around 100 Hz) and is also a subject to $1/f$ noise, the charge detection was performed in the frequency range 0.1–100 Hz. In this domain, the detection precision is better of $10^{-2} e/\sqrt{\text{Hz}}$, which ensures a sub-electron accuracy for the island average charge measurements even if the coupling capacitance between the box island and the SET island has been taken, as in our experiment, small compared to the box island capacitance to limit the back-action noise of the SET on the box.

A micrograph of the sample used in the experiment reported below is shown in Fig. 4 together with its corresponding schematic diagram. The superconducting box circuit and the measuring electrometer can be seen at the top and at the bottom of the picture respectively. Both circuits are coupled by a small coupling capacitor placed at the end of the long T-shaped SET island. The box and SET circuits are simultaneously fabricated using a three-angle-evaporation of metallic layers through a nanofabricated trilayer mask on an oxidized silicon chip. Both devices consist of two identical $Al/Al_2O_3/Al$ junctions in series which confers a symmetric structure to the whole circuit (Fig. 4, bottom panel). In contrast with Fig. 1, the box island is coupled to the reservoir by two junctions instead of one. However, this does not change the topology of the circuit nor its degrees of freedom, provided that the impedance of the link between the two junctions is negligible compared with the impedance of the junctions. Furthermore, this feature allows the measurement of the series resistance of the tunnel junctions at room temperature, a crucial information that would be impossible to obtain with the box circuit in its simplest design. The configuration of the superconducting box is recovered by connecting the two junctions to

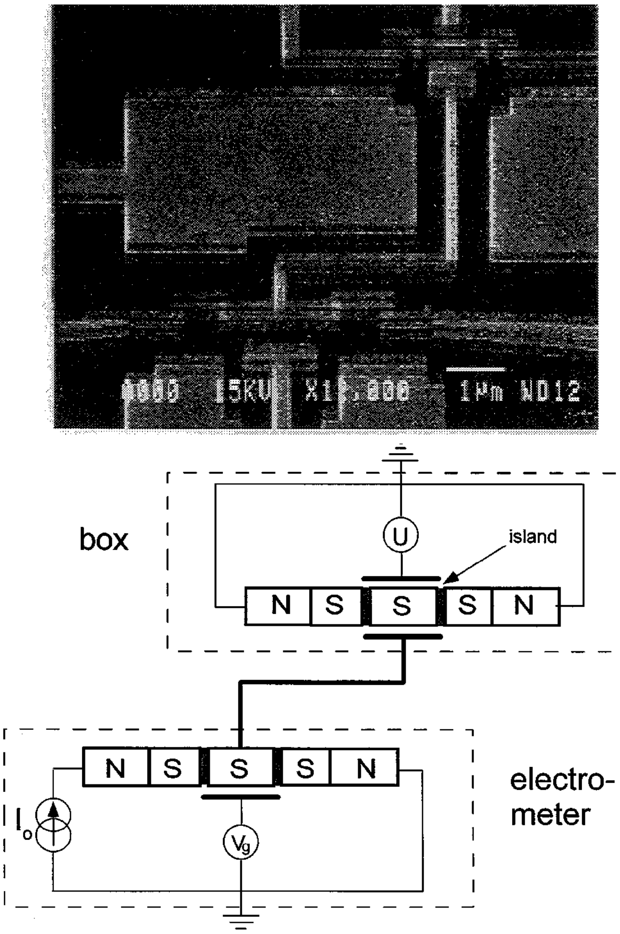

Fig. 4. Bottom: schematics of the experiment showing how the charge of the island of the superconducing box is measured with an electrometer. For adequate bias conditions, the voltage variations across the electrometer are proportional to the variations of the charge of the island. Although the island of the box is actually connected to two superconducting reservoirs in parallel for test purposes, its electrical environment is equivalent to that shown in Fig. 1. Top: electron micrograph of the on-chip implementation of the experiment. Triple evaporation through a shadow mask has been used to define the junctions and the normal ballast electrodes (noted N) used for filtering out-of-equilibrium quasiparticles.

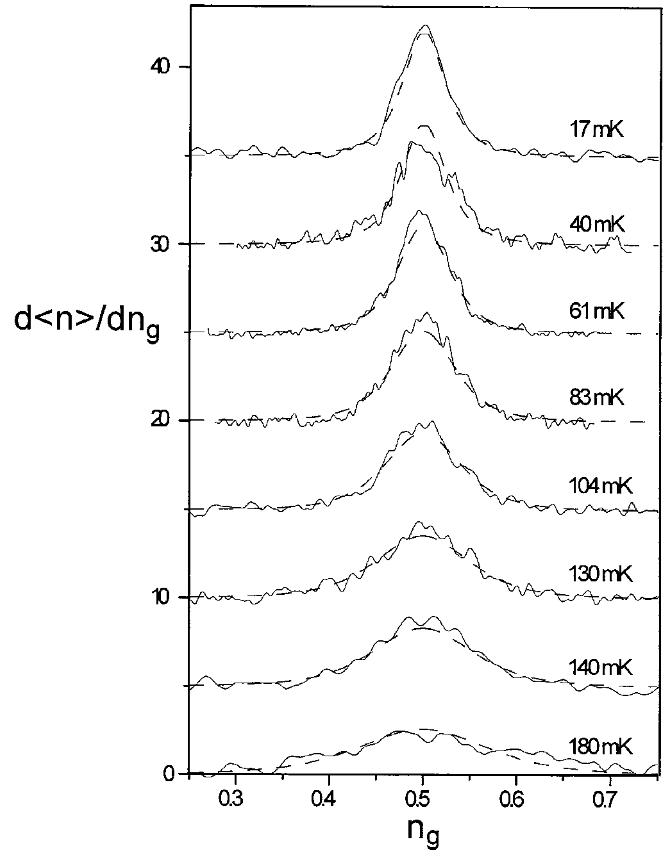

Fig. 5. Full line: derivative of the time-average of n with respect to the dimensionless gate voltage $n_g = C_g U/(2e)$ measured by a lock-in technique. Dashed line: theoretical fit using expressions (11) and (12) with $E_J \to 0$ and leaving T an adjustable parameter.

the same ground. These two junctions can be lumped into one effective junction with capacitance and Josephson energy which are twice as large as the nominal junctions. Implementing copper leads in series with the superconducting Al reservoirs was important to suppress spurious out-of-equilibrium quasiparticles. These normal-metal electrodes close to the device play the role of "filters" for quasiparticles.

A lock-in technique was used to measure the derivative $\partial\langle n\rangle/\partial n_g$ of the staircase of Fig. 2(b) as a function of n_g and T. The results for one step of the staircase are shown in Fig. 5. They show a peak which at first stays constant with increasing temperature and then broadens at higher temperatures. The width of the peak can be quantitatively determined by fitting the peak by the theory curve (eqs (11) and (12)) corresponding to $E_J \to 0$ (no quantum fluctuations of charge) and letting the temperature to be an adjustable $T_{\rm eff}$ parameter (see below for the determination of E_C which enters in the theoretical expression). In Fig. 6 we plot $T_{\rm eff}$ as a function of T. This plot shows that as T is lowered, $T_{\rm eff}$ first follows T and then saturates.

We have further measured the residual width of the low temperature peak by directly measuring the average charge during several sweeps of the gate voltage and accumulating the resulting curves. The staircase obtained at 20 mK in this way is shown in Fig. 7, together with theoretical predictions.

We now explain how we have determined the parameters of the experiment in order to compare experimental results and theoretical predictions with no adjustable parameters.

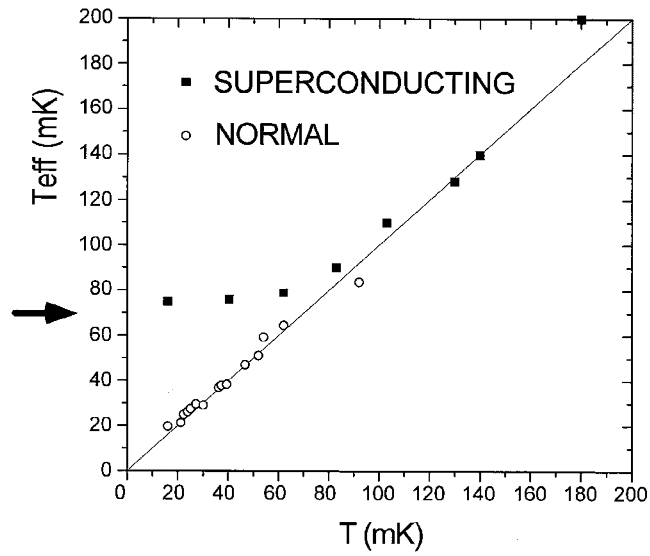

Fig. 6. Effective temperature $T_{\rm eff}$ obtained from the fit of the peaks of $d\langle n\rangle/dn_g$ obtained with the lock-in technique as a function of temperature for the sample in the superconducting (full squares, see Fig. 5) and normal (open circles) states. The arrow corresponds to theoretical predictions (see text).

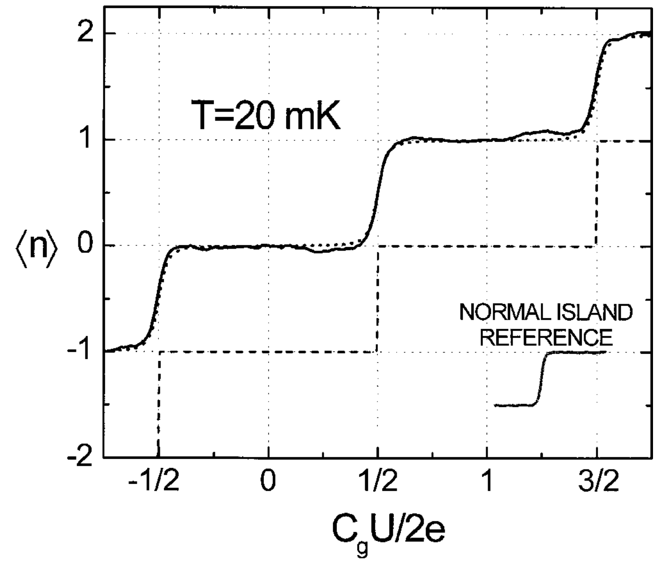

Fig. 7. Full line: direct measurement of the average value of n as a function of U in the superconducting state. Dotted line: theoretical prediction for the superconducting box with experimentally determined parameters. Dashed line: theoretical prediction not taking into account the Josephson energy and showing the effect of thermal rounding alone. Inset in bottom right shows reference step taken in the normal state at the same temperature $T = 20\,\mathrm{mK}$. The thermal broadening of the step is relatively larger than the broadening displayed by the dashed line because the Coulomb energy for electrons is 4 times smaller than for Cooper pairs.

Since the effect of quantum fluctuations due to Josephson tunneling is to round the staircase; it is particularly important to check that spurious effects, such as extraneous electromagnetic noise, do not also significantly contribute to a rounding of the staircase.

4. Comparison between experiment and theory

Four energies are involved in the comparison between experiment and theory:

- the island charging energy E_C,
- the Josephson coupling energy E_J,
- the odd-even free energy difference $\tilde{\Delta}(T)$,
- the thermal fluctuation energy $k_B T$.

The two energies E_C and E_J are determined during sample fabrication by respectively choosing the tunnel junction areas at the electron-beam lithography stage, and transparencies at the oxidation stage. The third energy, $\tilde{\Delta}$ [12], is also sample dependent, but can also be continuously reduced down to zero during the measurement by applying a small magnetic field. The fourth energy should not be determined directly from the thermometers. An *in-situ* control experiment is necessary to ensure that the electrons of the box are indeed thermalized at the temperature indicated by the thermometer under the conditions of the experiment.

4.1. *Determination of E_C and T*

Superconductivity in the aluminum electrodes is suppressed by applying a magnetic field of 0.1 T. The device then becomes a single-electron box [16], the island charge is quantized in units of e ($q = n_1 e$) and the Coulomb stair-case is e-periodic. The reduced gate charge is $n_{1g} = n_g/2$ and the Coulomb energy is now $E_{1C} = E_C/4$. For each experimental curve $\partial\langle n_1\rangle/\partial n_{1g}$ (data not shown), we have determined the best fitting parameter T_{eff}/E_{1C}. The fitting parameter varies linearly with temperature down to the lowest temperatures, providing strong evidence that no extraneous noise source contributes to the rounding of the staircase. The Coulomb energy of the box in the superconducing state, obtained from the slope of the best linear fit of the data points, is $E_C/k_B = 2.5 \pm 0.2\,\mathrm{K}$. This value is in good agreement with our estimate of the tunnel junction capacitance. Our analysis is shown in Fig. 6 we plot the values of T_{eff} in the normal state corresponding to this value of E_C as a function of T. We find that the data in the superconducting state fall at low temperature on a plateau which is clearly out of the error bars of the data in the normal state. The shape of the step of the Coulomb staircase in the normal step is directly shown at the bottom right of Fig. 7.

4.2. *Estimation of the Josephson energy E_J*

The Josephson coupling energy E_J was estimated from eq. (5): $E_J = h\Delta/(8e^2 R_{T\|})$ where $R_{T\|}$ is the parallel combination of the two tunnel resistances of the box. The superconducting gap Δ, deduced from I–V curves, is $\Delta/k_B = 2.33\,\mathrm{K}$. The tunnel resistance of the two junctions in series measured in the normal state is $R_{T\|} = 36.6\,\mathrm{k\Omega}$. Assuming that both junctions are identical, we obtain the value $E_J/k_B = 0.2 \pm 0.02\,\mathrm{K}$ and the ratio $E_J/E_C = 0.08 \pm 0.015$.

4.3. *Estimation of the odd-even free energy difference $\tilde{\Delta}(T, H = 0)$*

This energy determines the presence of unpaired electrons in the island. In the BCS theory, it takes the value $\tilde{\Delta}(T, H = 0) = \Delta - k_B T \ln N$, where $N = vV\Delta$ is the number of Cooper pairs in the islands given as function of the density of states v of the metal and V the volume of the island. It can be deduced from the Coulomb staircase measured at intermediate magnetic fields, in the regime when $\tilde{\Delta}(T, H)$ is reduced below E_C, thus leading to the appearance of intermediate steps in the Coulomb staircase. The analysis of the Coulomb staircase leads to a precise determination of $\tilde{\Delta}(H)$ [17]. The extrapolation of the data to $H = 0$ provides $\lim_{H\to 0} \tilde{\Delta}(T = 20\,\mathrm{mK}, H)/k_B = 0.74\,\mathrm{K}$, which is smaller than the BCS gap $\Delta/k_B = 2.33\,\mathrm{K}$. However, this value is sufficiently large compared with E_J that it is possible in the first approximation to ignore the corrections to the theoretical expressions (11) and (12). We attribute this reduced value of $\tilde{\Delta}$ to the existence of one or several discrete quasiparticle states in the gap of the superconductor. Such states, commonly observed in other similar samples, can even lead to the suppression of the $2e$-periodicity of the Coulomb staircase and jeopardize the observation of quantum coherence effects in superconducing tunnel junction circuits. Their origin is not understood, but might arise from impurities in or at the surface of the aluminum island.

From these estimates of the parameters of the theory, we can compute the effective temperature that would be found at $T = 0$ in the superconducting box experiment. This value $T^*_{\mathrm{eff}} = 70 \pm 10\,\mathrm{mK}$ is indicated as an arrow in Fig. 6 and is in good agreement with the experimental data.

The staircase at 20 mK obtained by the direct measurement method is also in good agreement with the theoretical prediction, as shown in Fig. 7 by a dotted line. Also shown in dashed line is the theoretical prediction at 20 mK without inclusion of the Josephson energy, which allows, in another manner, to compare the observed rounding of the staircase to the rounding due to thermal fluctuations only.

Given all these control measurements, we attribute the rounding of the Coulomb staircase observed at low temperature in the superconducting box to the effect of the quantum fluctuations due to Josephson tunneling. Our measurement provides the first direct evidence that the ground state of a single Cooper pair device can be a coherent superposition of only two charge states. This ground state is "intrinsically quantum-mechanical" in the sense that for $n_g \simeq 1/2$, neither its Cooper pair number nor its Cooper pair phase are well-defined.

5. Perspectives

In quantum computing, the information is coded by a two state quantum system nicknamed "q-bit" by analogy with the classical bit used in ordinary computers. Our experiment shows that the superconducting box provides a solid-state, easy-to-integrate quantum system. The two lowest energy states of a superconducting box could be used to code a single q-bit. An array of superconducing boxes, controlled by independent gate voltages, and coupled to one another by tunnel junctions, could be used to implement a logic function. Such an array is equivalent to an array of coupled spins $\frac{1}{2}$ each placed in a locally controllable magnetic field. A major challenge in the realization of such tunnel junction arrays is the control of the even-odd free energy $\tilde{\Delta}$ which, for every island, needs to be well above the Josephson energy to avoid poisoning of the system by unpaired electrons. Preliminary experiments with small arrays [18, 19] have met unexpected difficulties in this respect, but they could perhaps be circumvented by improved fabrication techniques.

Decoherence results in "fatal errors" in a quantum computer [20]. We have shown above theoretically that the decoherence in the superconducting box is mainly due to the dissipation induced by the electromagnetic environment of the circuit. Using the expression of the decoherence rate (eq. (14)), we estimate that the life-time of a q-bit can be longer than 10 μs. This time would already be sufficient to perform interesting manipulations on the quantum state of a system with several q-bits. Experiments in progress in our laboratory and others (Delft, Stony Brook, NEC) are now aiming at measuring directly the decoherence time of quantum superposition of charge states.

Acknowledgements

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Note added in proof

A. Schnirman and G. Schön (to be published in Physical Review) have recently analyzed theoretically the measurement of the quantum state of a superconducting electron box by a single electron transistor presented in this paper.

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12. The precise condition under which all electrons of the island are paired is $k_B T \ll e^2/(2C_\Sigma) \ll \tilde{\Delta}(T)$, where $\tilde{\Delta}(T)$ is the even-odd free energy which at $T = 0$ and $H = 0$ is equal to Δ in a BCS superconductor. See Tuominen, M. T. *et al.*, Phys. Rev. Lett. **69**, 1997 (1992); Lafarge, P. *et al.*, Nature **365**, 422 (1993).
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