

Quantum Charge Fluctuations and the Polarizability of the Single-Electron Box

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We measure the average charge on the island of a single-electron box, with an accuracy of two thousandths of an electron. Thermal fluctuations alone cannot account for the dependence of the average charge on temperature, on external potential, or on the quasiparticle density of states in the metal from which the box is formed. In contrast, we find excellent agreement between these measurements and a theory that treats the quantum fluctuations of charge perturbatively.

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A general feature of quantum many-body phenomena is the screening of a single degree of freedom by a bath of virtual excitations. The Lamb shift and the Kondo effect are well-known examples, where the discrete states of a hydrogen atom or a magnetic impurity are renormalized by the quantum fluctuations of an environment of virtual photons or virtual spin flips. In single-electron circuits [1], such as the single-electron transistor (SET) [2], the charge pump [3], or the single-electron box [4], the same sort of quantum fluctuations exist in a system which can be controlled and measured electrically. These fluctuations arise from the virtual tunneling of electrons between the metal islands and the metal leads that comprise single-electron devices. Electron-hole pairs, generated by the virtual tunneling, partially screen the charge on the islands and modify the discrete spectrum of charge states. The single-electron box, the simplest single-electron circuit, is the ideal system in which to test the theory of quantum charge fluctuations.

The box has been studied theoretically [5–9] because it is a model system for understanding electron-electron interactions and because the quantum fluctuations in the box are analogous to both the Kondo effect [5] and the Lamb shift. In spite of the extensive theoretical work, few experiments have probed the fluctuations described by Refs. [5–9]. Those experiments that have done so are mostly in semiconductor dots [10–12]; whereas the theory of Refs. [6–9] describes metallic systems, such as our box or Refs. [13,14], in which the tunnel junctions comprise many nearly opaque channels. Because the quantum fluctuations screen an electron with a polarization charge much less than one electron, very sensitive charge measurements are required to resolve the fine structure associated with these fluctuations.

In this Letter, we measure the time-averaged charge on the island of a single-electron box with an accuracy much better than one electron using a radio-frequency SET (rf-SET) [15]. We observe quantum fluctuations of charge, and we modify the strength of these quantum fluctuations by changing the temperature, the external potential, and the quasiparticle density of states of the

metal in which the tunnel junction is embedded. In each case, we find quantitative agreement between our results and the theory of quantum fluctuations.

Our single-electron box is composed of an isolated aluminum island attached to an aluminum lead through a thin insulating layer across which electrons can tunnel. A 1 T magnetic field is applied to keep the aluminum in its normal (nonsuperconducting) state. An additional lead, called the gate lead, lies near the island and changes the electrostatic potential of the island with the application of a voltage V_g to the gate lead through the gate capacitance C_g . The total island capacitance C_Σ^0 is small enough that the addition of a single electron to the island requires a large electrostatic energy

$$U_n = E_C^0(n - n_g)^2, \quad (1)$$

where $E_C^0 = e^2/2C_\Sigma^0$ is the charging energy, n is the number of excess electrons on the box, and $n_g = C_g V_g/e$. The minimum energy is clearly achieved when n is the integer nearest n_g ; when $n_g = 0.5$ the two lowest-energy charge states are degenerate.

Equation (1) ignores the quantum fluctuations, or the effects of the coupling of island and lead through the tunnel junction. The junction couples the charge states to each other and to quasiparticle excitations in the metal on either side of the junction. This alters the spectrum of states in Eq. (1) in three ways. First, the charging energy is reduced (C_Σ^0 is enhanced) from its bare value E_C^0 to a renormalized value $E_C^N = e^2/2C_\Sigma^N$ in the normal state or $E_C^S = e^2/2C_\Sigma^S$ in the superconducting state. Second, when a pair of states are nearly degenerate their energy difference becomes temperature dependent. Finally, the electrostatic energy U_n of the charge states is no longer quadratic in n_g . The magnitude of these three effects is calculated with a theory perturbative in the dimensionless conductance $g = R_K/(4\pi^2 R_j) = (h/e^2)/(4\pi^2 R_j)$ [6,7,9], where R_j is the box junction resistance. By measuring the average charge on the box island Q_{box}/e versus n_g (Coulomb staircase) with an uncertainty less than g , we can compare all three effects with theory.

We measure Q_{box} as a function of n_g by coupling the box island to an rf-SET electrometer through a capacitor C_C [15] [Fig. 1(a)]. The quantity that we measure directly is the charge coupled to the electrometer Q_{elec} versus n_g , which approximates a sawtooth function (Coulomb sawtooth). We infer the Coulomb staircase as $Q_{\text{box}} = (n_g e \kappa - Q_{\text{elec}})/\kappa$ [Fig. 1(c)], where $\kappa = C_C/C_{\Sigma}^N$ is the fraction of the charge on the box that couples to the electrometer. The value of $e\kappa$ is not an independently known parameter; rather, it is determined as the slope of the Coulomb sawtooth at $n_g = 0$, assuming that Q_{box} is independent of n_g at $n_g = 0$. Following this procedure, we extract a value of $\kappa = (3.35 \pm 0.05) \times 10^{-2}$. Our assumption is valid if we interpret C_{Σ}^N (and E_C^N) determined in our experiments as a value renormalized by tunneling, not the bare, geometrical value C_{Σ}^0 , which is a parameter in the theory of Refs. [6,7]. While we cannot prevent

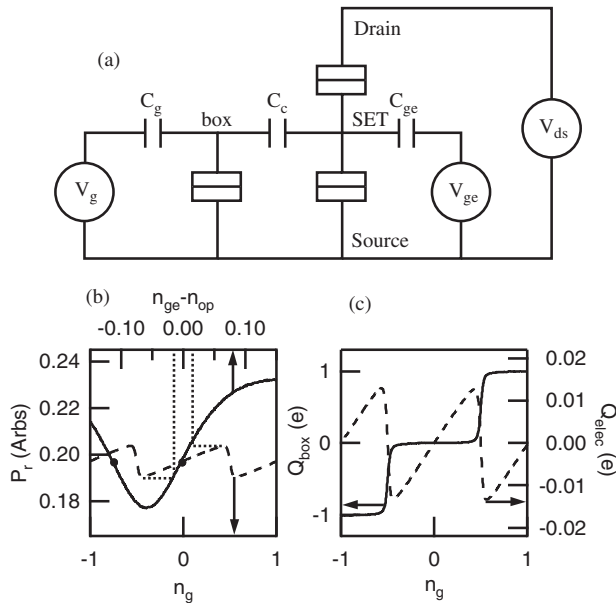


FIG. 1. (a) Circuit diagram of the single-electron box capacitively coupled to an rf-SET electrometer. The tunnel junctions are represented by boxes divided by a horizontal line. The junction capacitance C_J is the dominant component of the total box capacitance, $C_{\Sigma}^0 = C_J + C_g + C_C$. Additional circuit elements (not shown) apply an rf signal between the SET's drain and source and detect the amount of rf power reflected P_r from the rf-SET [15]. (b) Calibration of the Coulomb sawtooth is accomplished by varying the SET's control gate voltage $V_{ge} = n_{ge}(e/C_{ge})$ about a fixed operating voltage $V_{op} = n_{op}(e/C_{ge})$ while the box gate is held at $n_g = 0$. This applies a known charge signal $Q_{\text{elec}} = e(n_{ge} - n_{op})$ to the SET. The plot P_r versus $n_{ge} - n_{op} = Q_{\text{elec}}/e$ (solid line, top axis) is a nonlinear map (implied by dotted lines) that converts P_r versus n_g (dashed lines, bottom axis) into Q_{elec} versus n_g . The electrometer's operating point $n_{op} = 0.44$ and an alternative $n_{op} = 0.56$ and are indicated (two dots). (c) The Coulomb sawtooth, Q_{elec} versus n_g , (dashed line) on the right axis and the Coulomb staircase, Q_{box} vs n_g , (solid line) on the left axis at $T = 30$ mK.

tunneling and measure C_{Σ}^0 , we can suppress tunneling and observe a variation in the total box capacitance.

We find the first evidence of quantum fluctuations by examining the temperature dependence of the Coulomb staircase. We measure the staircase at a high temperature ($T = 500$ mK) and extract a value of $E_C^N/k_B = 1.57 \pm 0.05$ K ($C_{\Sigma}^N = 590 \pm 20$ aF) by assuming thermal broadening, that is, a Boltzmann occupation of the states in [Eq. (1)]. In the range 200–500 mK, we find excellent agreement [Fig. 2(a)] between the measured staircase and thermal broadening in a comparison with no adjustable parameters. Below 200 mK, thermal fluctuations characterized by any single temperature cannot account for the measured staircase [Fig. 2(b)]. As the temperature of the cryogenic apparatus is reduced, away from $n_g = 0.5$ the staircase remains rounded as if the box's temperature were saturating around 130 mK. Nevertheless, the staircase grows continually sharper at

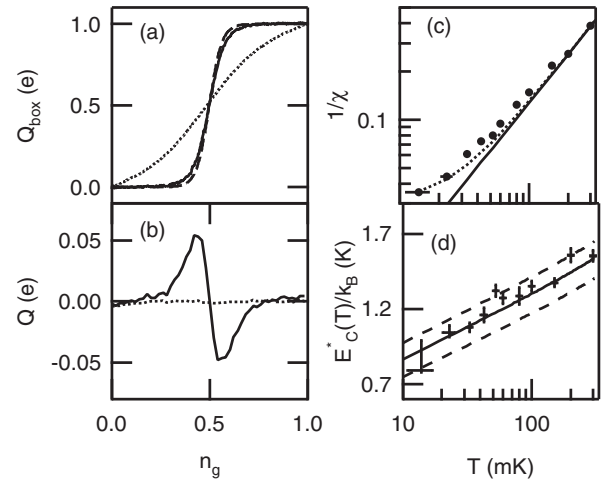


FIG. 2. (a) Coulomb staircases at $T = 500$ mK (dotted line) and $T = 100$ mK (solid line). The charging energy E_C^N is extracted by fitting to the 500 mK data a theoretical staircase (not shown) broadened only by thermal fluctuations. The 100 mK staircase is compared to the thermal fluctuation theory with no adjustable parameters (dashed line). (b) The residuals of the 500 mK fit (dotted line) and of the 100 mK comparison (solid line). (c) Plotted versus T on logarithmic scales are the measured value of $(1/\chi)$ (points), the expression $1/\chi = 2k_B T/E_C^N$ (line) showing the expected behavior in the absence of quantum fluctuations, and the expression $1/\chi = 2k_B \sqrt{(T^2 + T_{sp}^2)}/E_C^N$ showing the expected behavior in the absence of quantum fluctuations but in the presence of a spurious broadening characterized by a phenomenological effective temperature $T_{sp} = 25$ mK (dotted line). A model of temperature-independent spurious broadening does not contain the observed behavior of χ versus T . (d) The quantity $E_C^*(T) = 2k_B T \chi$ (points) and the prediction of [9] (line) versus T with no adjustable parameters. Dashed lines indicate the range of theory consistent with the uncertainties in g and E_C^N . The error bars of two lowest T points account for a systematic rounding introduced by the SET's backaction [16].

electrostatic degeneracy, $n_g = 0.5$, consistent with temperatures below 30 mK. Because the box is most sensitive to external noise at degeneracy, this surprising behavior is both inconsistent with an external source of noise and a qualitative hallmark of quantum fluctuations [10].

The theories of Refs. [5,8,9] predict the slope $\chi = (1/e)dQ_{\text{box}}/dn_g$ of the Coulomb staircase, essentially the polarizability, at $n_g = 0.5$ as a function of temperature [8,9]. Because $\chi = E_C^N/2k_B T$ in the absence of quantum fluctuations, a plot of $1/\chi$ versus T reveals the quantum fluctuations in its deviation from a line with slope $2k_B/E_C^N$ [Fig. 2(c)]. Near $n_g = 0.5$, where the two lowest charges states are nearly degenerate, quantum fluctuations cause a temperature-dependent reduction in the energy separation, $U_1 - U_0 = E_C^*(0.5 - n_g)$, of the levels, described by a reduced $E_C^* = 2k_B \chi T < E_C^N$. Reference [9] implies $E_C^* = E_C^N[1 - 2g(3.154 + \ln(E_C^0/\pi k_B T))] + \mathcal{O}[g^2, (k_B T/E_C^N)^2]$, where $E_C^0 = E_C^N(1 + 4g + \mathcal{O}(g^2))$. Note the similarity to the Kondo effect where the screening of a localized magnetic impurity by itinerant spins leads to a logarithmic in T correction of the impurity's magnetic moment [5]. In Fig. 2(d), we plot $E_C^*(T)$ versus T and find good agreement with Ref. [9], in a comparison with no adjustable parameters. This same effect was observed in SET's by Joyez *et al.* [13].

To make the comparison with theory, we must have an independent determination of the dimensionless conductance of the box, $g = (4.2 \pm 0.2) \times 10^{-2}$, which can be obtained by studying the box in its superconducting state. With no applied magnetic field, the aluminum superconducts, and the parameters of the box, $C_\Sigma^S = 518 \pm 6$ aF and $R_j = 15.4 \pm 0.9$ k Ω , can be extracted by microwave spectroscopy of the coherent two-level system formed by the coupling of Cooper pairs between the lead and the island [18]. What is directly measured is the charging energy in the superconducting state $E_C^S/k_B = 1.79 \pm 0.02$ K and the Josephson energy $E_J/k_B = (\hbar\Delta/8e^2 R_j k_B) F(E_C^0/\Delta) = 0.62 \pm 0.01$ K, where $\Delta/k_B = 2.4 \pm 0.1$ K is the BCS gap in aluminum and $F(E_C^0/\Delta)$ is a function that accounts for Coulomb blockade effects by modifying the usual Ambegakor-Baratoff relation [19]. For our sample $F(E_C^0/\Delta) = 1.25$. Note that in the superconducting state $E_C^S = e^2/2C_\Sigma^S$ is not the same as $E_C^N = e^2/2C_\Sigma^N$ in the normal state. This difference reflects the different quantum fluctuations of a metal with a superconducting or with a normal quasiparticle density of states (DOS).

We are able to tune this influence of the DOS by continuously reducing Δ in the aluminum with an applied magnetic field B_{app} . We observe that C_Σ^S is a function of $\Delta(B_{\text{app}})$ by measuring $\kappa^S = C_C/C_\Sigma^S$, the slope of the Coulomb sawtooth at $n_g = 0$, as the aluminum is driven from the fully superconducting state to the normal state. With increasing B_{app} , κ^S is reduced continuously from a value $\kappa^S = (3.70 \pm 0.05) \times 10^{-2}$ with $B_{\text{app}} = 0$ to $\kappa =$

$(3.35 \pm 0.05) \times 10^{-2}$ in the normal state (Fig. 3). Because both κ^S and E_C^S are proportional to $1/C_\Sigma^S(B_{\text{app}})$ we infer $E_C^N/k_B = [E_C^S(B_{\text{app}})/k_B][\kappa/\kappa^S(B_{\text{app}})] = 1.62 \pm 0.04$ K, which is consistent with the value 1.57 ± 0.05 K extracted from the broadening of the Coulomb staircase at high temperatures.

The theory of the normal box [7] predicts that the effects of tunneling can be treated around $n_g = 0$ as a renormalization of C_Σ^0 to a value larger by the factor $[1 + 4g + 10.93g^2 + \mathcal{O}(g^3)] = 1.18$. The renormalization of the bare capacitance in the superconducting state C_Σ^S/C_Σ^0 is predicted to be smaller than in the normal state because the quasiparticle excitations have a minimum energy Δ , which suppresses the virtual tunneling. The bare capacitance is not an experimentally accessible parameter; however, the perturbative techniques of Ref. [7] can be used [20] to calculate the renormalization of C_Σ^0 for a metal with a BCS, rather than constant, DOS. Inverting this, we infer from the normal state ($C_\Sigma^0 = 498 \pm 16$ aF) and from the superconducting state ($C_\Sigma^0 = 478 \pm 7$ aF) values for the bare capacitance that are consistent with each other. By altering the DOS, we have observed that the capacitance of a tunnel junction is not a property of tunnel junction alone, but also of the spectrum of low-energy excitation in the metal from which it is made.

We have already seen that the electrostatic energy of the box is both a function of the temperature and of the

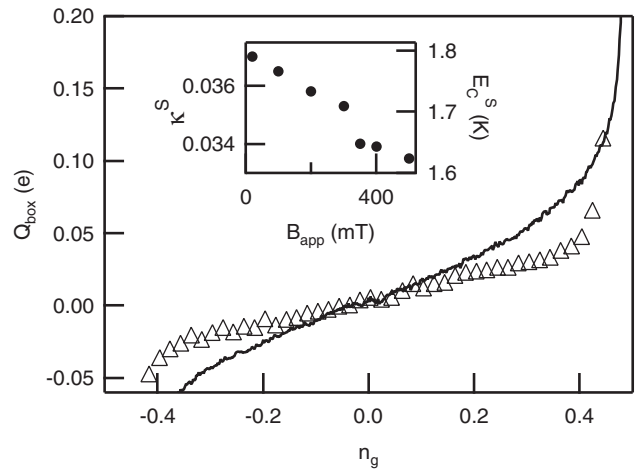


FIG. 3. Coulomb staircases with the box in its normal state (line) and in its superconducting state, $B_{\text{app}} = 100$ mT (triangles). For both, the Coulomb sawtooths have been converted to staircases using $\kappa^0 = C_C/C_\Sigma^0 = 3.9 \times 10^{-2}$, which would be the slope of the Coulomb sawtooth around $n_g = 0$ in the absence of tunneling. The renormalization of C_Σ^0 is visible as the nonzero slope $(1/e)dQ_{\text{box}}/dn_g$ of these plots at $n_g = 0$. The inset shows $\kappa^S(B_{\text{app}})$ and the value of $E_C^S(B_{\text{app}})$ inferred from κ^S , as the aluminum is driven from its superconducting to its normal state. In the superconducting state, a single out-of-equilibrium quasiparticle on the box's island keeps the Coulomb staircase e periodic.

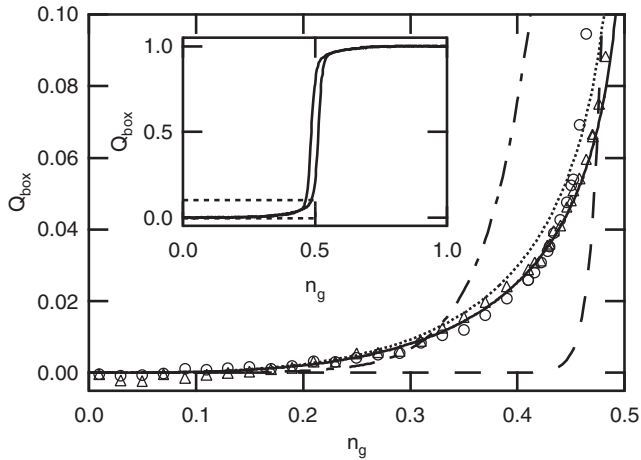


FIG. 4. Coulomb staircases with the box at two different electrometer operating points, $n_{op} = 0.44$ (triangles) and $n_{op} = 0.56$ (circles); $V_{ds} = 0$ for both. The SET's backaction causes these two curves to deviate from each other around $n_g = 0.5$. The theory plots are the charge on the box predicted for thermal fluctuations but no quantum fluctuations at $T = 125$ mK (dashed dotted line) and $T = 29$ mK (dashed line), and for the quantum fluctuations calculated to first order in g (dotted line) and second order in g (solid line) [6,7]. The second order calculation is fit to the data with the adjustable parameter $\kappa = (3.375 \pm 0.001) \times 10^{-2}$, which is better constrained by this fit than by extracting the slope of the Coulomb sawtooth. Note we have plotted both data and theory in the form with $dQ_{box}/dn_g = 0$ at $n_g = 0$ unlike Refs. [6,7]. Inset: the same data over a range $0 < n_g < 1$. The dashed box indicates the region plotted in the main figure.

quasiparticle DOS in metal lead and island. We now show that the ground-state energy deviates from the parabolas of Eq. (1). Because $Q_{box}/e = n_g - (1/2E_C^0)(dU_n/dn_g)$ at $T = 0$ [6], Eq. (1) implies perfectly flat steps in the Coulomb staircase, whereas we observe some curvature around $n_g = 0$ even at $T \ll E_C^N$. The ground-state energy cannot be quadratic in n_g . This modification of the ground-state energy is the Lamb shift in the single-electron box.

It is precisely the detailed shape of the Coulomb staircase at $T = 0$ that is predicted by [6,7] and which provides the most stringent test of the theory (Fig. 4). We find that Q_{box} deviates from a perfect step function by several percent in the range $0 < n_g < 0.45$ [16]. In this region at the base temperature of our cryogenic apparatus, we may consider the box to be in a zero temperature limit and ignore the influence of the electrometer (Fig. 4). To an accuracy of $2 \times 10^{-3} e$, limited by the linearity of the applied gate voltage, we find agreement with this theory. Our measurement is sufficiently accurate and sensitive that the perturbative calculation of Ref. [7] must be carried out to second order to show agreement with our experiment, even for the relatively small value of $g = 4.2 \times 10^{-2}$.

In these experiments, we have used an rf-SET electrometer to measure the polarizability of a mesoscopic

electrical circuit. We have chosen to apply this technique to the single-electron box, a model system for understanding electron-electron interactions whose Hamiltonian is analogous to the Kondo Hamiltonian. We find excellent agreement between our measurements and a perturbative treatment of the quantum fluctuations. The excellent agreement between our measurements and theory both supports this theory and demonstrates the precision electrometry possible with the rf-SET. The technique we demonstrate would be an ideal method for exploring the equilibrium behavior of more complicated mesoscopic circuits, such as semiconductor quantum dots or carbon nanotubes.

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- [1] *Single Charge Tunneling*, edited by H. Grabert and M. H. Devoret, Nato Advanced Study Institutes, Ser. B, Vol. 294 (Plenum, New York, 1992).
- [2] T. A. Fulton and G. J. Dolan, *Phys. Rev. Lett.* **59**, 109 (1987).
- [3] H. Pothier *et al.*, *Europhys. Lett.* **17**, 249 (1992).
- [4] P. Lafarge *et al.*, *Z. Phys. B* **85**, 327 (1991).
- [5] K. A. Matveev, *Sov. Phys. JETP* **72**, 892 (1991).
- [6] G. Göppert *et al.*, *Phys. Rev. Lett.* **81**, 2324 (1998).
- [7] G. Göppert and H. Grabert, *Phys. Rev. B* **63**, 125307 (2001), and references therein.
- [8] H. Schoeller and G. Schön, *Phys. Rev. B* **50**, 18436 (1994).
- [9] H. Grabert, *Phys. Rev. B* **50**, 17364 (1994).
- [10] D. Berman *et al.*, *Phys. Rev. Lett.* **82**, 161 (1999).
- [11] D. S. Duncan *et al.*, *Appl. Phys. Lett.* **74**, 1045 (1999).
- [12] L. W. Molenkamp, K. Flensberg, and M. Kemerink, *Phys. Rev. Lett.* **75**, 4282 (1995).
- [13] P. Joyez *et al.*, *Phys. Rev. Lett.* **79**, 1349 (1997).
- [14] D. Chouvaev *et al.*, *Phys. Rev. B* **59**, 10599 (1999).
- [15] R. J. Schoelkopf *et al.*, *Science* **280**, 1238 (1998).
- [16] In a region $0.45 < n_g < 0.55$ and $T < 50$ mK, Q_{box} depends on the operating point of the electrometer. While we can accurately model the electrometer's influence on the box [17], no theory yet accounts for quantum fluctuations and backaction simultaneously.
- [17] B. A. Turek *et al.* (to be published).
- [18] K. W. Lehnert *et al.*, *Phys. Rev. Lett.* **90**, 027002 (2003).
- [19] P. Joyez, Ph.D. thesis, University of Paris, 1995.
- [20] V. Bouchiat, Ph.D. thesis, University of Paris, 1997.