

## Devices which Transfer Electrons One-by-One

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### Abstract

This paper provides an introduction to the field of single-electron devices, which are electronic circuits across which electrons may be transferred one by one. The paper will describe experiments on the single-junction box and the four-junction box, devices in which electrons were transferred to a metallic island and their presence detected using a single-electron electrometer. The single-junction box, the simpler of the two devices, allows one to control the thermally-averaged number of excess electrons on the metallic island, whereas with the four-junction box one can actually control the transfer of individual electrons.

In 1911, Millikan<sup>(1)</sup> showed that the charge on a drop of oil is always an integer multiple of the electron charge  $e$ ; however, the discreteness of the electron charge is rarely evident in electronic circuits, appearing only as shot noise in the current flowing through a vacuum tube or a semiconductor diode. Screening effects in metals and semiconductors, as well as thermal noise and the spread of the electron wavefunctions, makes the conduction process appear fluid and continuous.

The first realization that the conduction process can be strongly affected by the discreteness of the electronic charge came through experiments on granular metallic thin films, as discussed by Gorter<sup>(2)</sup>. Experiments had shown that the current-voltage ( $I$ - $V$ ) characteristics of these films exhibit a region of low conductance at small bias voltages  $V$ , and that the conductance of this region increases with temperature  $T$ . Gorter postulated that the conduction in these films was due to the hopping of electrons from one metallic grain to the next. If a grain has a self-capacitance  $C$ , then the energy  $E$  to place one electron on the grain is  $E = e^2/2C$ . If the capacitance is small enough that the energy satisfies  $E \gg k_B T$ , then the flow of electrons across the grain will be inhibited, resulting in a small value of conductance. When the temperature is increased to a value larger than the largest value of  $E$  in the conduction path across the film, this effect will disappear.

In 1986 Averin and Likharev<sup>(3)</sup> launched the field of single-electron devices, by developing some of the fundamental theoretical ideas in the field, and by proposing that it was possible to intentionally fabricate devices in which the conduction process is directly affected by the discreteness of the electron charge. The fabrication of such devices became possible through the development of nanolithographic techniques, where the use of the scanning electron microscope (SEM) as a lithography exposure tool allows the patterning of thin metal films with dimensions on the order of 100 nm; such small dimensions allow one to meet the charging condition  $e^2/2C \gg k_B T$ , if the measurements are carried out on a dilution refrigerator at a temperature well below 1 K. The single-electron devices which have been developed to date all have a few basic elements in common; they consist of metal wires which are interrupted either by pure capacitors, which allow no electrons to pass across, or by metal-insulator-metal tunnel junctions, which are simply capacitors whose two plates are placed so close together (about 1 nm) that electrons may cross from one plate to the other by quantum-mechanical tunneling through the insulating barrier. When the two metals are in the

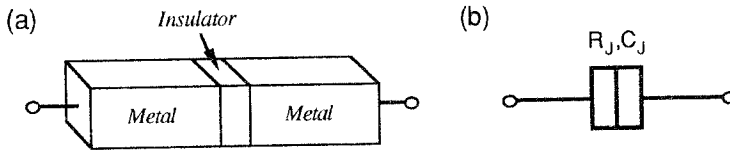


FIGURE 1. (a) Schematic representation of a tunnel junction; the insulator is typically about 1 nm thick. (b) Electrical symbol used to represent a tunnel junction, with tunnel resistance  $R_J$  and capacitance  $C_J$ .

normal state (as opposed to the superconducting state), and are biased with a constant voltage  $V$ , the rate of electron tunneling through the insulating barrier is quite accurately proportional to the voltage: the number of states available for an electron to tunnel into increases linearly with voltage, and by Fermi's rule the rate increases proportionally. The current  $I$  through the junction is therefore given by  $I = V/R_J$ , where the parameter  $R_J$  is called the junction resistance. A schematic drawing of a junction and the electronic symbol used to designate it are shown in Figs. 1(a) and (b); note that the geometry of the junction implies that it has a non-negligible capacitance  $C_J$ . A requirement on the tunnel junction is that its resistance must be sufficiently high to meet the criterion  $R_J \gg h/e^2 \approx 25.8 \text{ k}\Omega$ , as otherwise the electrons are not sufficiently localized on one side of the barrier as opposed to the other<sup>(4)</sup>.

### The Single Junction Box

The most elementary example of a single-electron device, the single junction box, is shown in Fig. 2. The series combination of the capacitor and the tunnel junction forms a metallic island, consisting of the right plate of the capacitor  $C_s$ , the left plate of the tunnel junction, and the wire connecting them, as outlined in the figure. The metallic island is designed to have a very small capacitance  $C = C_J + C_s$ , on the order of 1 fF; in order to place a single electron on this island, one needs to provide an energy  $e^2/2C$  corresponding to a temperature of about 1 K. If this device is cooled to about 20 mK by using a  $^3\text{He}$ - $^4\text{He}$  dilution refrigerator, the number of electrons on the island will be solely determined by the bias potential  $U$  applied across the capacitor and the tunnel junction, and as this potential is increased from zero, the number of electrons will increase one by one in order to minimize the electrostatic energy.

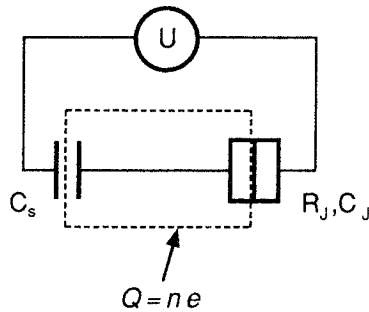


FIGURE 2. Electrical circuit for the single-junction box, consisting of a voltage source  $U$ , bias capacitor  $C_s$  and a tunnel junction; the number of electrons  $n$  on the island is a discrete integer.

More specifically, one can write the electrostatic energy  $E$  of the complete circuit in terms of the number of electrons  $n$  on the island and the charge  $Q = C_s U$ :

$$E_n = \frac{[n(-e) + Q]^2}{2(C_s + C_J)} - \frac{Q^2}{2C_s} \quad (1)$$

The energy as a function of  $Q$  with  $n$  fixed is a parabola, as expected, and as one varies  $n$  across its allowed integer values one finds a family of parabolas. The system will always be in the state of lowest energy, at least at zero temperature. The value of  $n$  will therefore increase by one whenever the value of  $Q$  crosses the values  $-3e/2$ ,  $-e/2$ ,  $e/2$ , etc. The effect of finite temperature will be to make the measured value of  $n$  correspond to the thermal average  $\langle n \rangle$  over all states  $E_n$ , as long as the measurement is over a time much longer than the inverse electron tunneling rate  $\Gamma^{-1}(n \rightarrow n \pm 1)$ , which is at most of order  $R_J C_J$ , or less than 1 ns. In general, the transfer rate of electrons across a junction is given by<sup>(5)</sup>

$$\Gamma = \frac{1}{e^2 R_J} \frac{\Delta E}{1 - \exp(-\Delta E/k_B T)} \quad (2)$$

where  $\Delta E$  is the change in total electrostatic energy, including the work done by the external voltage sources.

The single junction box presents an experimental system in which all of the fundamental elements of this type of device can be found. The authors therefore performed an experiment to demonstrate that these elements were understood, by measuring the variation in the electron number  $n$  as a function of the bias voltage  $U$ ; this was to be done by measuring the voltage developed across the tunnel junction due to the charge  $Q_J$  on its capacitance  $C_J$ . It can easily be shown that the thermally averaged charge  $\langle Q_J \rangle$  on the junction is given by

$$\langle Q_J \rangle = \frac{C_J}{C_S + C_J} (\langle n \rangle (-e) + Q) . \quad (3)$$

The major difficulty in measuring  $\langle Q_J \rangle$  is that by connecting leads of any significant length to the island of the box, one adds a very large ( $\sim 1$  pF) stray capacitance in parallel to the junction capacitance, and in doing so enormously reduce the difference in energy between the states  $n$  and  $n \pm 1$ . The resolution of this problem was to use an electrometer with very small input capacitance, as described below.

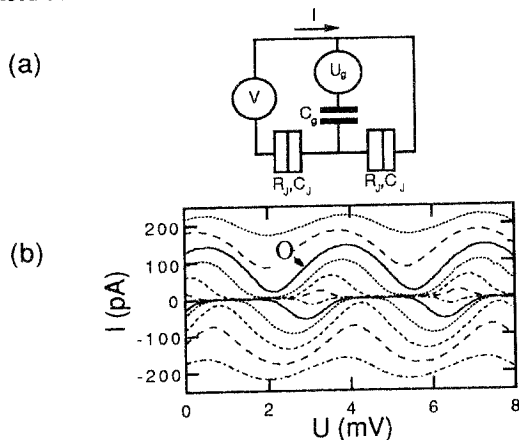


FIGURE 3. (a) Circuit diagram for the electrometer. It is usually operated by holding the bias voltage  $V$  constant and modulating the gate voltage  $U_g$ , while monitoring the current  $I$ . (b) Response  $I$  vs.  $U_g$  for the electrometer, measured at various bias voltages  $V$ , at a temperature of 20 mK. The point  $O$  indicates the optimal bias values for  $U_g$  and  $V$ .

### The Electrometer

The single-electron device which can be used as an ultra-sensitive electrometer was first demonstrated by Fulton and Dolan<sup>(5)</sup>; this device is also known as a single-electron transistor. This device, shown in Fig. 3(a), consists of two series-connected junctions biased with a voltage  $V$ , with the metallic island formed between the two junctions also capacitively coupled through the capacitor  $C_g$  to a gate voltage  $U_g$ . The junctions have tunnel resistances  $R_J \gg R_K \approx 25.8 \text{ k}\Omega$  and capacitances  $C_J$  of the order of 1 fF; the gate capacitance  $C_g$  is typically about 0.1 fF. The detailed behaviour of this device may be worked out with equations similar to those for the single-junction box; we will simply sketch out how this device is used. If the current  $I$  flowing through the junctions is measured as a function of the gate voltage  $U_g$  for a number of different bias voltages  $V$ , one finds the family of curves shown in Fig. 3(b). The current is periodic in the charge  $q$  on the capacitor  $C_g$  with period  $e$ ,

and the modulation amplitude of the current varies with voltage  $V$ , with the maximum variation  $\Delta I \approx e/R_J C_J$  for charge variations  $\Delta q = e/2$  appearing for bias voltages  $V \approx e/2C_J$ . The optimum bias point, with the greatest sensitivity  $\Delta I/\Delta q$ , is indicated by the point  $O$  in Fig. 3(b).

The usefulness of this device can immediately be seen from the characteristics shown in Fig. 3(b): small variations  $\Delta q$  in the charge on  $C_g$  give rise to easily measured variations in the current  $I$  flowing through the junctions. With standard room-temperature amplifiers, the limits on the noise in the measurement of  $I$  enable us to measure equivalent variations in the charge of about  $10^{-4} e/\sqrt{\text{Hz}}$  at 1 kHz, when the electrometer is biased at the point  $O$ ; in other words, if we average the current measurement for 1 second, we can measure a charge variation of  $10^{-4} e$ . The major difficulties in using such a device are its high input impedance (formed by the small gate capacitance  $C_g$ ), and the resultingly large voltages developed at the gate: a charge of  $e$  on  $C_g$  produces a voltage of about 1 mV with respect to the electrometer island. Note that in standard applications, one usually couples the charge to be measured through a second gate capacitor  $C_g'$ ; the capacitor  $C_g$  is used to bias the electrometer so that it is operating at the optimum bias point  $O$ .

#### Experimental Investigation of the Single Junction Box

The measurement of the single junction box was carried out by coupling an electrometer to the box with a coupling capacitance of about 80 aF; the circuit diagram is shown in Fig. 4(a). The electron box was made with two junctions connected in parallel, equivalent to a single junction with twice the capacitance and half the tunnel resistance; this was done so that the junctions could be tested prior to the measurement. The electron box and electrometer were fabricated on a Si chip using SEM lithography and shadow evaporation of Al, as discussed in Ref. (7), and the device was mounted inside a Cu shield on the mixing chamber of a dilution refrigerator; details of the measurement can be found in Ref. (8). The Al could be driven into the normal state with a 0.5 T superconducting magnet. The basic measurement was to bias the electrometer at its optimal point  $O$ , and then observe the current flowing through the electrometer as a function of the bias voltage  $U$  on the box. From the electrometer transfer function  $I(q)$ , the charge on the box island could be determined, and the results are plotted in Fig. 4(b) for both the normal and superconducting states of the device; also plotted are the theoretical predictions from Eqs. (1) and (2).

The sawtooth oscillations in the junction charge  $\langle Q_j \rangle$  in the normal state are fairly close to the predictions; the charge jumps by nearly  $e$  for every increase of  $e$  in the bias charge  $C_s U$  (the jumps are not quite  $e$  due to the factor  $C_j/(C_s+C_j)$  and thermal averaging). The discrepancy between the theory and experiment can be accounted for by taking a system temperature of 60 mK instead of the temperature of 20 mK indicated by the refrigerator thermometry; however, the source of this increased temperature is not understood. What is more surprising is the observation that the sawtooth oscillations in the superconducting state appear identical to those in the normal state: one would naively expect that the oscillations would be  $2e$ -periodic due to the Cooper pairing of the superconducting electrons. The  $e$ -periodicity implies that there is an unknown source of unpaired electrons in the superconducting device.

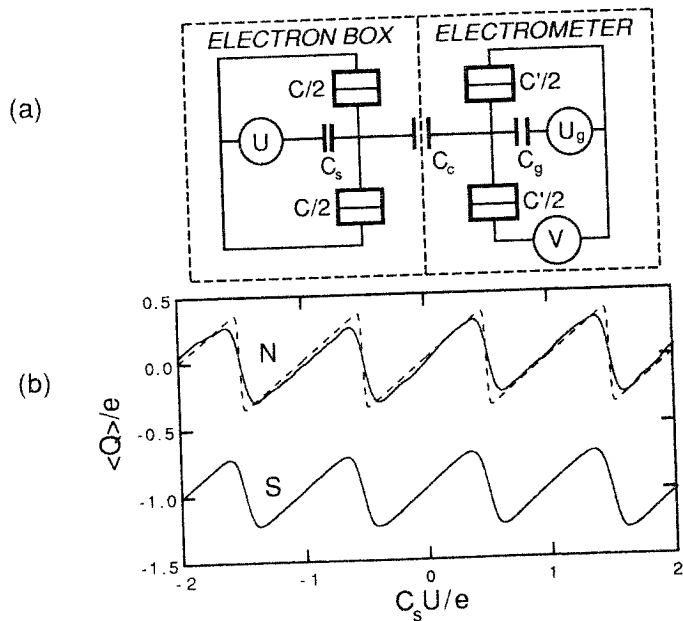


FIGURE 4. (a) Circuit diagram for the measurement of the charge on the electron box with a weakly coupled electrometer; the capacitor  $C_c$  has capacitance of 80 aF. (b) Average charge  $\langle Q \rangle$  on the island of the box as a function of the box bias  $U$ , in the normal (N) and superconducting (S) states. The theoretical prediction for the N curve is shown as a dotted line, calculated at the experimental temperature of 20 mK.

### The Four Junction Box

The single junction box is appealing in its simplicity and in the ability to observe the change in the average charge by one electron on the island; however, the experiment does not measure the rate of tunneling of electrons into and out of the island, as given by Eq. (2), and it also does not allow the observation of the transfer of single electrons into and out of the island, only the change in their thermally-averaged number. In order to measure both of these quantities, the authors developed an experiment in which the single junction of the box was replaced by four series-connected junctions, as shown in Fig. 5(a). Each of the islands  $a$ ,  $b$ , and  $c$  formed between two junctions was coupled through a capacitor  $C_i = 80$  aF to a voltage source  $U_i$ , and the island  $p$  formed by the leftmost junction and the gate capacitor  $C_s$  was capacitively connected through  $C_c = 240$  aF to the electrometer, as in the case of the single junction box. The role of the voltage sources  $U_i$  is simply to tune out the unavoidable background charges induced on the islands, and for the sake of this discussion may be ignored. The box state is then specified by the value of the bias voltage  $U$  and the number of electrons on each of the islands,  $\{n_p, n_a, n_b, n_c\}$ . Note that in the circuit diagram, the island  $p$  is coupled through a capacitance  $C_s = 80$  aF to the voltage  $U$ , but also has a larger capacitance  $C_p = 2$  fF to ground. The energy to add one electron to the island  $p$  is therefore significantly less than to add an electron to any of the islands  $a$ ,  $b$ , or  $c$ . At the bias voltage  $U = 0$ , the stable state is that with no charge on any island, and as the voltage is increased, the state with one electron on the island  $p$  drops in energy until at  $C_s U = e/2$ , it has energy equal to the initial state. For values of  $U$  larger than this, the state with one electron on  $p$  has lower energy than the initial state and the states with one electron on  $a$ ,  $b$ , or  $c$ . At this point it becomes possible for an electron to pass from the right side of the circuit to the island  $p$ , tunneling by way of the virtual states with one electron on each of the intermediate islands: in other words, the electron "co-tunnels" across the four junctions to the island  $p$ . The rate for the co-tunneling process was worked out by Averin and Odintsov<sup>(9)</sup>. For the experiment described here, for values of  $C_s U$  near  $e/2$  the rate of co-tunneling can be well below 1 Hz, and individual tunnel events should therefore be easily observable.

The experimental details of the measurement are very similar to those for the single junction box, and will be published elsewhere<sup>(10)</sup>. In Fig. 5(b) we display the current through the electrometer as a function of time, with the bias voltage  $U$  across the box junctions held fixed. The



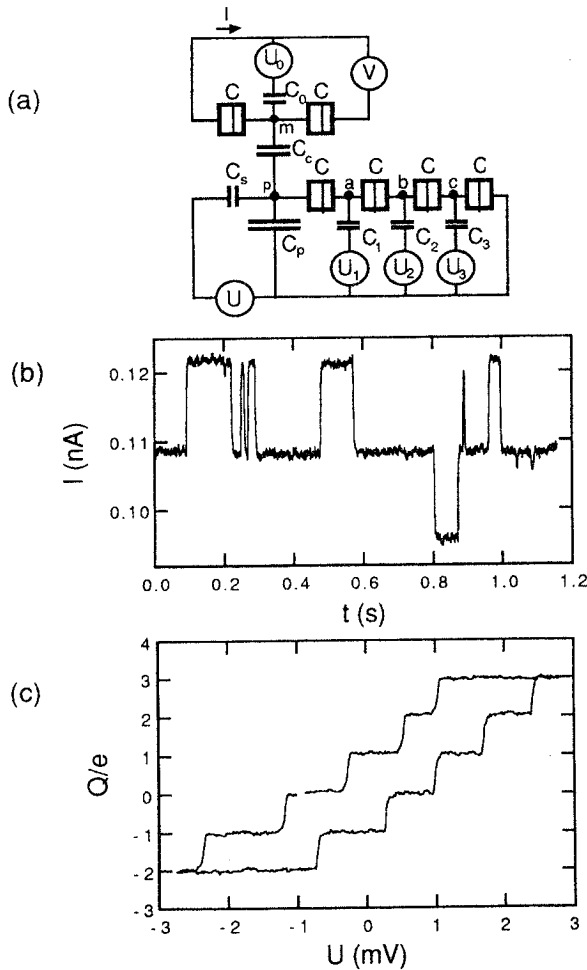


FIGURE 5. (a) Circuit diagram for the 4-junction box, consisting of four junctions connected in series, and an electrometer to measure the charge on the box island. (b) Measurement of the current  $I$  through the electrometer as a function of time; the telegraph signals are due to single electrons hopping on to and off of the island  $p$ . (c) Measurement of the charge  $Q$  on the box island as a function of the bias voltage  $U$  applied across the junctions.

sudden changes of the electrometer current are due to single electrons hopping across the four junctions to or from the island  $p$ . We have therefore successfully observed the transfer of single charges to and from a metallic island. In Fig. 5(c) we show the charge  $Q$  on the island as a function of the bias voltage  $U$ ; the voltage was swept through a triangle wave with a 0.4 s period, and the

hysteresis in the value of charge  $Q$  is due to the slow tunneling rate of electrons, which causes the actual state of the system to lag the stable state determined by the instantaneous value of  $U$ .

Detailed comparison of the tunnel rate predicted by the theory and that found in the experiment indicates a serious discrepancy: the measured rate scales roughly with bias voltage  $U$  as predicted by the theory, but the overall rate is  $10^5$  times higher than expected, even if the theory is calculated using a temperature of 50 mK as opposed to the refrigerator temperature of 20 mK. Adding additional radiofrequency filters, as well as injecting microwave noise into the leads connected to the experiment, had no effect on the tunneling rate. The discrepancy is not at present understood.

Measurements in the superconducting state were performed as well, and just as in the case of the single junction box, only single electron events were observed. A very similar experiment has been performed by Fulton *et al.*<sup>(11)</sup>, in the superconducting state only, and their measurements yielded very similar results to our own. Despite the large discrepancy with the theory, this experiment has indicated that metrological measurements involving the counting of electrons could quite possibly be performed, with errors of the order of one mis-transferred electron per second.

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