Determination of Coulomb-Blockade Resistances and Observation of the Tunneling of Single Electrons in Small-Tunnel-Junction Circuits

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Coulomb-blockade effects greatly enhance the resistance of low-capacitance tunnel-junction circuits at low bias and temperature. Experiments which involve the charging of small capacitances through such circuits are used to determine semiquantitatively the degree of this enhancement. Resistances of $\gtrsim 10^{17}$ Ω have been observed for four-series-junction circuits whose individual junctions have resistances $\sim 10^6$ Ω. Two-series-junction circuits show lesser enhancements. For the higher resistances, one can observe directly the charging proceeding in discrete steps as individual electrons tunnel through the circuits.

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Coulomb-blockade behavior is characteristic of low-capacitance tunnel-junction circuits. One of its signatures is an enhanced resistance in the low-voltage region of the (nonlinear) I-V curve. Thus blockade resistances of $\gtrsim 10^{10}$ Ω commonly occur in circuits whose junctions taken singly have resistances of $\lesssim 10^6$ Ω. It is suspected, further, that the enhancements actually may be much higher than has been measured so far. We report experiments in which a small capacitor ($\sim 10^{-13}$–10^{-15} F) is slowly charged through such a circuit composed of two or four junctions of submicrometer dimensions placed in series. The capacitor voltage is monitored by an adjacent blockade-based electrostatic voltmeter. The RC time delay between the capacitor voltage and the applied voltage gives an approximate blockade resistance of the array. Resistances from $< 10^{12}$ to $> 10^{17}$ Ω have been observed in this way. For the higher resistances (occurring only in the four-junction arrays) currents of $\lesssim 1$ electron/s can be involved. In some such cases the discrete passage of the individual electrons is seen in real time. Such high resistances are desirable for potential applications in metrology (e.g., the "single-electron turnstile" [1] or related devices [2] as a standard of current) or perhaps in memory devices.

To review [3] briefly, consider the Giaever-Zeller-type [4] circuit of two tunnel junctions in series, with a region of low capacitance C between junctions. (Multijunctions circuits behave similarly but are more complicated in detail.) The current flow is impeded by the electrostatic energy needed to move a tunneling electron to the low-capacitance region. In our experiments, for example, C is ~ 0.1–1 fF, and the energy barrier of $\sim e^2/2C$ is ~ 0.1–1 meV. At temperatures $T < 1$ K the I-V curves are correspondingly nonlinear in the region of $V \sim e/C$. For $V \gg e/C$ the resistance approaches the usual series resistance of the junctions, but for $V \ll e/C$ it is much higher, due to the barrier. Furthermore, the I-V shape is sensitive to any "modulation voltage" V_M on an electrode having mutual capacitance C_M with the low-C region. As V_M increases, the high-resistance region alternately shrinks to nothing and reexpands to full size cylindrically, with period e/C_M.

The semiclassical model predicts a thermally activated blockade resistance proportional to $\exp[(e^2/2C)/kT]$ at low V and T (with V_M set to maximize the blockade). This is because the energy of a tunneling electron imparted by the bias plus any thermal energy is usually small compared to $e^2/2C$. For metrology purposes the ratio of blockade resistance to that of the individual junctions should be $\gtrsim 10^8$. The semiclassical model predicts that this will occur at an attainable T, e.g., $T \sim 0.05$ K for $e/C \sim 0.2$ mV. A quantum treatment [5] suggests, however, that such enhancements will be hard to realize with a two-junction device, due to what is called "macroscopic quantum tunneling of charge." Multiple-junction devices are predicted to be more promising in this respect. Experimental support for this picture has been obtained [6].

The experimental circuit is sketched in Fig. 1. The series array S of two or four junctions is the device under test. It connects voltage V_J to an otherwise-isolated patch of thin film K of area ~ 10–1000 μm^2. The capac-

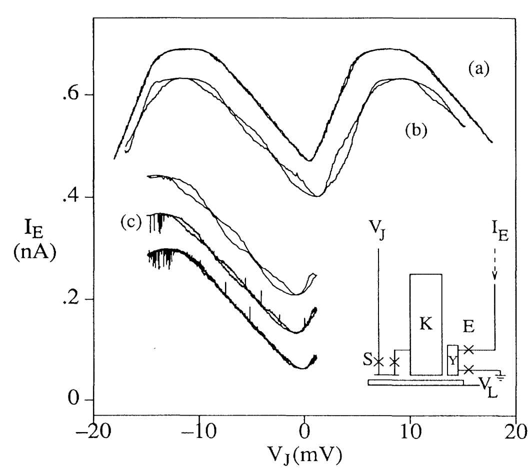

FIG. 1. Cyclic curves for a two-junction array. The curves are offset vertically to avoid overlap. Curve a, I_E vs V_L (expanded horizontally ×20). Curve b, I_E vs V_J at 1.3 mV/s. Curves c, I_E vs V_J at three sweep rates, 1.3, 0.13, and 0.07 mV/s from top to bottom. Inset: A sketch of the circuit.

itance of K, C_K, is charged or discharged through S as V_J is changed. A nearby two-junction circuit E has a central region Y coupled to K with capacitance C_{EK}. As described above, the shape of the I-V curve of E is modulated periodically by the voltage of K, V_K, with period e/C_{EK}. Hence E serves as a voltmeter to monitor V_K. All the films lie on an oxidized degenerately doped silicon substrate L. The capacitance between K and L constitutes the bulk of C_K. The voltage of L, V_L, modulates the shape of the I-V curve of S, periodically for two junctions (with period e/C_{SL}) and quasiperiodically for four junctions. Both V_K and V_L affect E in the same way but with different periods e/C_{EK} and e/C_{EL}. The junctions are of the usual [7] Al-oxide-Al thin-film form, of typical dimensions $\lesssim 0.1$ μm and resistances of $\sim 10^5$–10^7 Ω.

A number of such circuits have been studied. For two-junction arrays the area of K was usually made comparatively large, ~ 1000 $\mu\mathrm{m}^2$, in expectation of a lower blockade resistance. Four-junction arrays generally used smaller areas for K, 2–100 $\mu\mathrm{m}^2$. The I-V of S could not be measured directly. We believe the resistances and capacitances are similar to those of the electrometers, judging from the I-V curves of cofabricated and two- and four-junction monitor circuits. Generally, the Coulomb-blockade behavior displayed in the measured I-V's was of the well-developed form expected from the semiclassical model, allowing a fairly accurate determination of the resistances and (for two-junction circuits) capacitances. Resistances and capacitances of nominally identical cofabricated circuits usually differed by less than $\times 2$ and $\times 1.25$, respectively. Estimates of geometrical capacitance compared reasonably well to measured values. The range of T was 0.1–1 K. The Al electrodes are superconducting in this range, but may be driven normal by application of a magnetic field.

In the superconducting state the I-V curve of a single junction shows a well-known increase in resistance at low V and T from quasiparticle freeze-out. For $T < 1$ K and $\Delta = 0.2$ mV the expected increase is proportional to $\exp(2.3/T)$. We suspect this exponential behavior may not continue much below $T \sim 0.2$ K. In this regime quasiparticle freeze-out is extreme in that no quasiparticles are present most of the time in the isolated low-capacitance region whose volume is $\sim 10^{-15}$ cm^3. (This might even depend on whether the number of electrons present is odd or even.) The current flow, even though $\lesssim 100$ electrons/s, may induce a nonequilibrium density. Pair tunneling may also act to lower the resistance.

In a typical experiment E is biased such that its current I_E is sensitive to the capacitor voltage V_K (and V_L). One then sweeps V_J in a sawtooth over ~ 1–20 mV in ~ 1–200 s and I_E is plotted versus V_J over one or more cycles. Such cyclic plots comprise most of the data to be shown [8]. As discussed, V_K often lags behind V_J, such that hysteresis occurs in this plot. The voltage across S is $V_S = V_J - V_K$ and the current through S is $I_S = C_K dV_K/dt$. Ideally, one could extract the I-V of S from data at various sweep rates, but our data quality is inadequate for that. We can only speak of an approximate resistance $R = V_S/I_S$. (Since the I-V is nonlinear, the value of R will depend on V_S.)

In Fig. 1, S is a two-junction array in the superconducting state at $T = 0.15$ K. Here C_K is ~ 0.2 pF, as estimated from geometry and direct measurements of smaller capacitances. Curve a is a cyclic (both sweep directions) plot of I_E vs $V_L(\times 20)$ showing the oscillatory response period (period 0.95 mV) of E to substrate voltage. Curve b is a cyclic plot of I_E vs V_J. The oscillatory shape (period 21 mV) is much like that of curve a, but regions of hysteresis occur periodically. At the maxima of these regions, V_K departs from V_J by $\lesssim 0.6$ mV and lags by ~ 0.5 s so that roughly $R \sim 3\times 10^{12}$ Ω. In the intermediate region where there is minimal hysteresis we estimate that $R < 5\times 10^{11}$ Ω. The periodic occurrence of hysteresis is due to the waxing and waning of the blockade resistance of S. This is induced through C_{SL} as described above, except that V_J changes and V_L is fixed rather than vice versa. The period, ~ 4.5 mV, is comparable to that found in the I-V of monitor devices of similar geometry. Curves c are taken at a different bias for E and a shorter range of V_J with three different sweep rates of V_J. Hysteresis which is well developed at 1.3 mV/s dwindles at 0.13 mV/s and disappears in the noise at 0.07 mV/s. R seems to increase by only about a factor of 2 from $V_S \sim 0.6$ mV (upper curve) to $V_S \sim 0.2$ mV (middle curve). At higher temperatures the hysteresis decreased and was largely gone by 0.4 K. In the normal state, obtained by applying a magnetic field, no hysteresis was observed at $T \geq 0.12$ K. For E the I-V shape gives $e/C \sim 0.3$ mV and the individual junction (normal state) resistances are $R_J \sim 5\times 10^5$ Ω. Assuming similar numbers for S, the semiclassical model predicts (at $T = 0.15$ K) a maximum blockade resistance in the normal state of $\sim 6\times 10^{10}$ Ω. The resistance in the superconducting state at the intermediate points, where the blockade is suppressed by modulation voltage, should be just the junction series resistance, $\sim 10^6$ Ω, augmented by superconducting quasiparticle freeze-out to (perhaps) $\sim 5\times 10^{12}$ Ω. The predicted maximum blockade resistance in the superconducting state (at $V_S = 0$) is $\sim 3\times 10^{17}$ Ω. One would not expect, then, hysteresis in the normal state, and also perhaps not in the superconducting state at the intermediate points (depending on the extent of quasiparticle freeze-out). The $< 10^{13}$ Ω maximum observed resistance, however, is far below that predicted. Since four-junction arrays on the same chip had resistances $\gtrsim 10^{16}$ Ω this discrepancy seems not to be the result of experimental difficulties. The predicted [5] quantum limit on the blockade resistance of a two-junction circuit is $\sim R_J[\kappa R_J/(\hbar/e^2)]$, where κ is a number ~ 10. Even a conservative estimate of $R_J \sim 10^9$–10^{10} Ω for the superconducting junctions still gives a much larger resistance than was observed. Pair tunneling may be involved [9].

Figure 2 shows similar plots for a four-junction S. Here K was 5.0 μm^2, giving $C_K \sim 1$ fF. The parameters for E were $e/C \sim 0.25$ mV and $R_K \sim 2\times10^6$ Ω. Curves a–c are taken in the superconducting state at $T=0.31$, 0.46, and 0.61 K. At the lower T unevenly spaced regions of hysteresis occur which dwindle and mostly disappear at the higher T. Curves d and e show comparable data for the same S in the normal state at $T=0.29$ and 0.62 K. (The larger signals in the superconducting state reflect the greater nonlinearity of the I-V curve.) Hysteresis is evident at the lower T, but has vanished at the higher T. The interpretation of these data is much as before, except that the resistances are much higher, consistent with Ref. [5]. (The nonperiodic response to V_J is due to unequal capacitances within S.) At $T=0.3$ K in the region of hysteresis the difference $V_J - V_K$ showed a decay time of several hundred seconds in the superconducting state, or $R>10^{17}$ Ω. In the normal state decay was faster, taking ~ 100 s. In both cases the decay was faster towards the edges of the hysteresis region than the center.

For samples having $C_K \lesssim 1$ fF one could expect to see further blockade effects due to C_K itself [10,11], with V_K taking on discrete values separated by e/C_K. Such effects were marginally visible in the sample of Fig. 2. A much clearer example is shown in Fig. 3 for a four-junction array. The detectors had $R_J \sim 1$ MΩ and e/C ~ 0.25 mV. The area of K was ~ 3 μm^2. The upper plot shows I_E vs V_J, here for a single sweep. A staircase structure is superimposed on the oscillatory voltmeter response (period 3.4 mV). The step spacing is 0.40 mV with some jitter. Roughly, one might think of S as a single junction whose resistance slowly varies with V_J. This should be reasonable, provided no electrons can be trapped within S, particularly in the portion adjacent to K. Then the electrostatic energy needed to charge K with one electron varies in a piecewise quadratic fashion with V_J, being $e^2/2C_K \sim 0.2$ meV in the middle of the steps and shrinking to zero in between. In this picture the occupation of K by electrons is in thermal equilibrium at fixed V_J, and the time-averaged shape of the step structure is determined by T. This seems to be only qualitatively true here. In some regions of V_J the step structure is well defined with flat steps, sharp transitions, and regular spacings, while in other regions it is more blurred. Possibly, the blurred regions occur where the internal structure of S plays a role. Interaction with the voltmeters may also be occurring. In some regions of well-defined steps hysteresis is observed with a faster sweep. The lower curves of Fig. 3 (which in the upper curve are in the region of $V_J=0.75$–0.8 mV) show such a case at three temperatures. At lower T the steps overlap, with some telegraphlike jumping back and forth. Here again the hysteresis demonstrates that S is high in resistance. The transitions between steps denote the passage of single electrons through S onto K. Comparable structure of the same period occurs in the normal state, but no hysteresis is observed in this temperature range.

In summary, we have observed that Coulomb-blockade resistances for multiple junctions in series can be so high that currents of only a few electrons/s will flow, and that this flow can sometimes be observed directly. For two junctions in series the resistances tend to be lower, al-

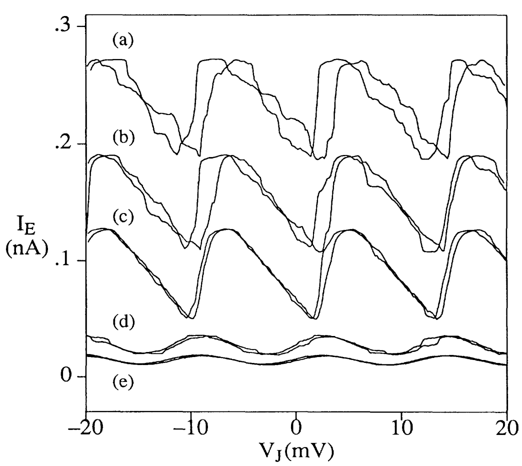

FIG. 2. Cyclic curves for a four-junction array. Curves a–c, I_E vs V_J in the superconducting state, at $T=0.31$, 0.46, and 0.61 K, and sweep rate = 1.2 mV/s. Curves d,e, I_E vs V_J in the normal state, at $T=0.29$ and 0.62 K, and sweep rate = 0.4 mV/s. Curves have been displaced vertically to avoid overlap.

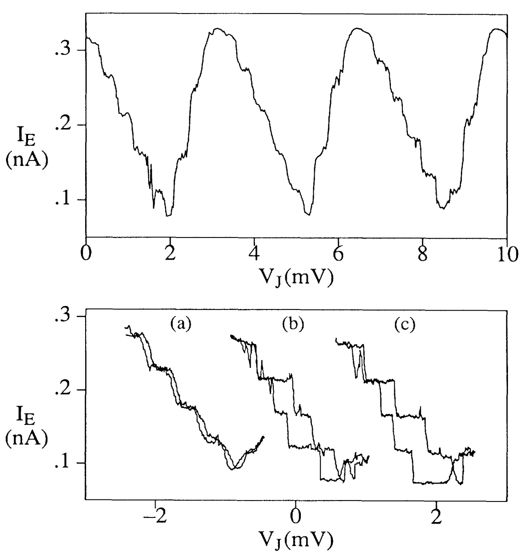

FIG. 3. Upper curve: Single-trace plot of I_E vs V_J for a four-junction array at $T=0.30$ K; sweep rate = 0.2 mV/s. Lower curves: Cyclic traces in a hysteretic region at $T=0.51$, 0.34, and 0.30 K for a–c; sweep rate = 2 mV/s. Curves have been displaced horizontally to avoid overlap.

though still substantial. This difference may be due partially to quantum effects.

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[8] It would be possible and in some ways better to unfold the oscillatory voltmeter response and plot V_K vs V_J. We prefer to deal with the unprocessed I_E vs V_J plots. The relation between I_E and V_K is nearly linear in much of the region of interest anyway.

[9] We have looked for RC delays with ~ 1-MΩ single junctions in the superconducting state using $C_K \sim 1$ pF. For $T \geq 0.1$ K we have seen no delay to our resolution of ~ 0.1 s.

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