Observation of Macroscopic Quantum Tunneling through the Coulomb Energy Barrier

L. J. Geerligs,[a] D. V. Averin,[b] and J. E. Mooij
Department of Applied Physics, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands
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The conductance of linear arrays of two and three normal-metal small tunnel junctions is studied for bias voltages V below the Coulomb-blockade threshold. At low temperature, we find evidence for macroscopic quantum tunneling of the electric charge (q-MQT) through the Coulomb energy barrier. For double junctions the tunneling rate scales as V^3, and approximately as the product of the junction conductances, as predicted by the theory of inelastic q-MQT.

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Quantum behavior of the macroscopic degrees of freedom of small tunnel junctions has recently attracted much attention.[1] Experimental investigations have been largely devoted to macroscopic quantum tunneling (MQT) of the Josephson phase difference[2,3] φ that can take place in superconducting tunnel junctions with large quasiparticle conductance $R_t^{-1} \gg R_Q^{-1}$ ($R_Q \equiv h/4e^2 \approx 6.5$ kΩ). In such junctions φ behaves as an almost classical variable. In particular, for small bias current the phase can be trapped in a minimum of Josephson potential energy, resulting in a zero-voltage supercurrent. Quantum fluctuations of φ give rise to a nonvanishing probability of quantum tunneling through the Josephson potential barrier, and make the zero-voltage state metastable. This macroscopic quantum tunneling of φ (φ-MQT) has been convincingly demonstrated in several experiments.[3]

In the dual case of tunnel junctions with small conductance ($R_t^{-1} \ll R_Q^{-1}$), the electric charge q on the junction capacitance, conjugate to φ, evolves nearly classically.[1] At low temperature this results in Coulomb blockade of tunneling: For low bias voltage the tunnel current is suppressed, since tunneling would increase the Coulomb energy of the junction capacitance. However, small quantum fluctuations of the charge give rise to a nonvanishing probability of quantum tunneling through the Coulomb energy barrier, and thus make the zero-current Coulomb-blockade state metastable. Only one electron (one Cooper pair for a superconducting junction) is transferred through the system in one act of this quantum tunneling. Nevertheless, electron transfers in these small junctions are essentially macroscopic processes, because the tunneling electron polarizes the junction electrodes in virtual states below the energy barrier, so that the Coulomb energy is given by usual macroscopic electrostatics. This implies that all free electrons of the junction electrodes do participate in the tunneling process. In this respect it is not the tunneling of a single electron but rather that of a macroscopic variable, the electric charge q of the junction. Because of the analogies of the macroscopic quantum tunneling of the electric charge through the Coulomb energy barrier to φ-MQT,[4] we will denote this process by q-MQT. Apart from its fundamental importance, q-MQT has practical implications. Proper functioning of a number of "single-electronic" devices[5] depends on the reliable trapping of an extra single-electron charge between small tunnel junctions. The q-MQT process decreases the reliability of operation of these devices. For example, it is a source of possible inaccuracy of current quantization in the recently reported single-electron turnstile device.[6]

In this Letter we report for the first time on the experimental observation of macroscopic quantum tunneling of the electric charge through the Coulomb energy barrier, in linear arrays of two and three normal-metal junctions. In such systems an act of q-MQT consists of a finite number of consecutive tunneling events. In an array of two junctions an electron is transferred via one intermediate state, in which an extra electron or hole charges the central metal electrode between the two junctions. The Coulomb energy of this intermediate state is equal to E_1 or E_2 if the first tunneling event occurs across the left or right junction, respectively:

$$E_1 = (e/C_\Sigma)[e/2 + Q_0 - V(C_2 + C_g/2)], \quad \text{(1a)}$$

$$E_2 = (e/C_\Sigma)[e/2 - Q_0 - V(C_1 + C_g/2)], \quad \text{(1b)}$$

where $C_\Sigma = C_1 + C_2 + C_g$, C_1 is the capacitance of the left junction, C_2 of the right one, and C_g of the gate [Fig. 1(a)]. Q_0 is the background charge of the central electrode which can be changed by the gate voltage V_g: $Q_0 = C_g V_g +$const. For bias voltage inside the Coulomb gap, $V < V_{th}$, the energy of the intermediate state is positive (this condition defines the threshold voltage V_{th}), and the thermally assisted classical consecutive tunneling is exponentially suppressed at low temperatures. However, even for $V < V_{th}$ there is a finite probability for an act of quantum tunneling with a virtual occupation of the central electrode.

For our metal junctions with relatively large electrodes, the main contribution to q-MQT is given by a process in which two different electrons tunnel through the two junctions. In this inelastic process the q-MQT creates an electron-hole excitation on the central electrode [Fig. 1(b)].[4,7] In metal particles smaller than about 10 nm or semiconductor heterostructures, the

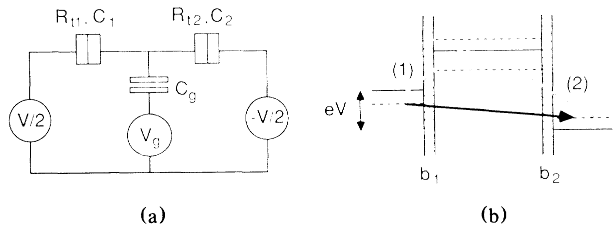

FIG. 1. (a) Equivalent circuit of a symmetrically biased two-junction array, as used in the experiments. A square divided by a vertical line indicates a tunnel junction. (b) Macroscopic quantum tunneling of the charge occurs via occupation of a virtual intermediate state. An electron tunnels to an excited state in the central island (1). The resulting virtual state decays by a different electron tunneling across the second junction (2), leaving a hole and the excited electron, but decreasing the charging energy. b_1 and b_2 indicate the junction barriers, and solid lines represent the Fermi levels in the electrodes.

energy-level spacing in the central electrode need not be very small compared to the characteristic charging energy. In this case an "elastic" contribution to q-MQT may become significant.[8,9]

The rate of q-MQT decreases with increasing tunnel resistance. It has been calculated in a perturbative approach, i.e., for $R_t \gg R_Q$.[4,7] For two junctions it is

$$\gamma_{\mathrm{MQT}} = \frac{\hbar}{2\pi e^4 R_{t1} R_{t2}} \left\{ \left(1 + \frac{2}{eV} \frac{E_1 E_2}{E_1 + E_2 + eV} \right) \times \ln \left[\left(1 + \frac{eV}{E_1} \right) \left(1 + \frac{eV}{E_2} \right) \right] - 2 \right\} eV , \qquad (2)$$

and for a linear array of N junctions (with index i) at bias voltage low compared to V_{th},

$$\gamma_{\mathrm{MQT}} \propto V^{2N-1} \prod_{i=1}^{N} \frac{R_Q}{R_{ti}} . \qquad (3)$$

Equation (2) is valid for $V < V_{\mathrm{th}}$, except in the vicinity of the threshold voltage. As a result of the discreteness of charge transfer in an act of q-MQT, γ_{MQT} decreases only as a certain power of the relevant parameter R_Q/R_t that determines the strength of the quantum fluctuations of q. The rate of the quantum tunneling of a continuous variable, e.g., φ-MQT, or q-MQT in a single junction shunted by an Ohmic conductance,[4] decreases exponentially.

We have studied q-MQT in four double junctions, with R_t between 41 and 347 kΩ. The junctions are made of overlapping aluminum strips, approximately 60 nm wide and 20 or 40 nm thick, and have an area of about (60 nm)^2. The resistance of the aluminum-oxide barrier is controlled by varying the oxidation pressure. Inside an array, the junctions are about 1 μm apart. The metal electrodes between the junctions can be polarized

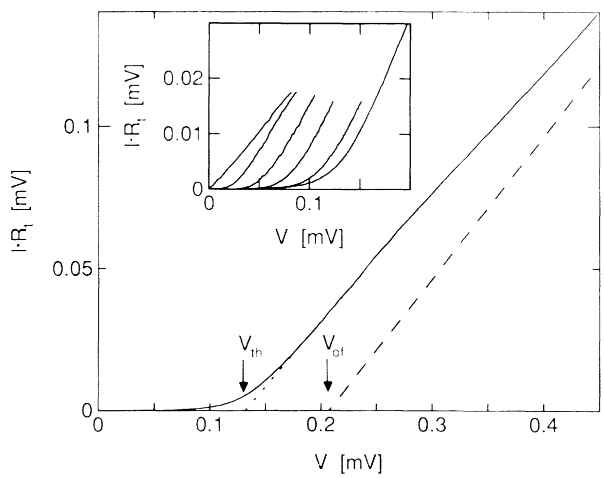

FIG. 2. I-V curves of a double junction with $R_t = 78$ kΩ. The maximum Coulomb gap is obtained by adjusting V_g. The dashed line gives the asymptote to the I-V curve that is used to determine $R_\Sigma = 2R_t$. V_{of} and V_{th} (see text) are indicated. The inset shows that the Coulomb gap can be continuously controlled with gate voltage (curves are given for, from right to left, $Q_0 \approx 0$, $0.1e$, $0.2e$, $0.3e$, $0.4e$, and $e/2$).

(i.e., the background charge Q_0 can be controlled) by a gate electrode at 1.5-μm distance, with a coupling capacitance C_g of about 0.07 fF. The samples were thermally anchored to the mixing chamber of a dilution refrigerator. For all measurements low-pass filters were used that were also thermally anchored to the mixing chamber. However, we found no significant difference for filtering with RC filters only, as compared to more careful high-frequency filtering. A magnetic field of 2 T was applied to bring the junctions into the normal state.

In Fig. 2 we show a typical I-V curve. The Coulomb gap is clearly visible, but even at the lowest temperatures (below 20 mK), the gap is rounded and there is significant current for voltages below V_{th}. The dashed line gives the asymptote to the I-V curve determined on a scale of a few mV. In the classical theory[5] this asymptote should intersect the zero-current axis at e/C_Σ, and have a slope $R_\Sigma = R_{t1} + R_{t2}$. The Coulomb-blockade threshold voltage should also equal e/C_Σ, provided that it is maximized by adjusting Q_0,[5] for instance, with V_g. In Table I we give $R_t = R_\Sigma/2$ and C_Σ, the latter determined from both the asymptote and the threshold voltage, for all double junctions. The capacitances obtained from the

TABLE I. The parameters of the four double junctions.

R_t (kΩ)	$C_\Sigma = e/V_{\mathrm{of}}$ (fF)	$C_\Sigma = e/V_{\mathrm{th}}$ (fF)	$C_{\Sigma,\mathrm{best\ fit}}$ (fF)
41	0.92	1.38	1.19
78	0.77	1.21	0.95
117	0.63	0.88	0.71
347	0.68	0.91	0.72

high-voltage asymptote (e/V_{of}) are smaller by a factor of 1.5 than those obtained from V_{th}. Several reasons can account for this difference. First, the Coulomb gap could be partially suppressed by imperfect adjustment of the gate voltage, or by thermal and quantum fluctuations of the charge, leading to a smaller V_{th}, and thus larger capacitance e/V_{th}. Second, the I-V-curve offset V_{of} at high voltages could be larger than the low-voltage offset, since additional channels of inelastic electron tunneling can become available with increasing voltage.[10] Another possible reason for a decrease in the effective capacitance with increasing voltage is capacitance renormalization due to Coulomb blockade. At low voltages the capacitance is increased by $\delta C=(4/\pi)^2(R_Q/R_t)C$,[11] while at high voltages $\delta C \propto (V_{th}/V)^2 \rightarrow 0$. The fact that the difference between the low-voltage and high-voltage capacitances is larger for junctions with lower R_t indicates that at least part of this difference can be attributed to the latter reason. We cannot quantitatively take into account all these factors, although it follows that the relevant capacitance for q-MQT should lie within the bounds set by V_{th} and V_{of}. For comparison with theory we will therefore use the capacitance as a fitting parameter.

In Fig. 3 we show the log(I)-log(V) curves for the four double junctions.[12] Classical tunneling current would increase exponentially with voltage. In contrast, the experimental log(I)-log(V) curves yield straight lines with a slope equal to 3, as expected for q-MQT [Eq. (3)], except for high or very low voltages. In Fig. 3(a) we compare the measured I-V curves with calculations for q-MQT from Eq. (2), assuming both junctions to be equal. The measured current and voltage are scaled to e/R_tC_Σ and e/C_Σ, respectively, making it possible to observe the effect of R_t on the q-MQT rate in one plot. All I-V curves can be very well fitted by theory in a broad voltage range inside the Coulomb gap, and the corresponding best-fit values of C_Σ lie (as they should) between the high-voltage and the low-voltage capacitances (Table I). The theoretical curves are only slightly shifted vertically if the junctions are assumed to be unequal in size. The resistance of junctions in different devices on the same substrate never differed more than 30%. From Table I it is also clear that the junction capacitance is well controlled. The smaller slope of the log(I)-log(V) curves at $V\rightarrow 0$ is probably caused by the fact that, if $eV \gg k_BT$ is no longer satisfied, crossover to a linear I-V curve occurs.[7,9] The larger slope for high voltages may be due to the crossover to thermally assisted charge transfer. In the regime where the classical tunneling rate becomes of order γ_{MQT} there will be more current than predicted from q-MQT. Inequality of the two junction capacitances or a not completely symmetric voltage bias would similarly increase the current at high voltages. We cannot exclude that these effects play a role in the experiments.

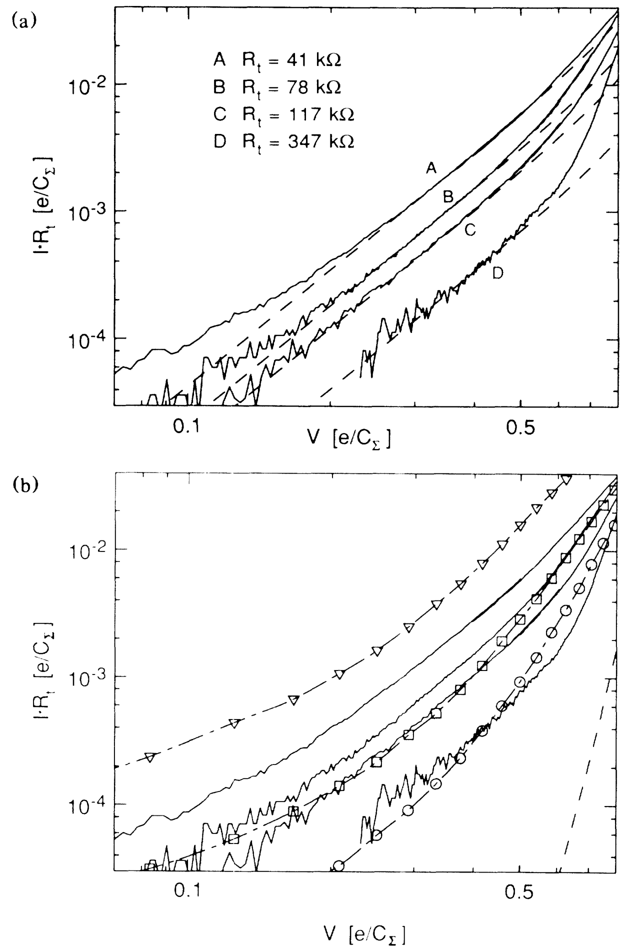

FIG. 3. Comparison of I-V curves with theory, for four double junctions. The measurements (solid curves) have been scaled with R_t and C_Σ, the latter being a fit parameter. The current increases with increasing R_t^{-1}; from top left to bottom right curve, R_t equals 41, 78, 117, and 347 kΩ, and C_Σ equals 1.19, 0.95, 0.71, and 0.72 fF. (a) The dashed curves give the predictions from q-MQT. (b) Comparison with predictions from thermally assisted tunneling. Dashed curve: $T=0.02e^2/C_\Sigma$ and $Q_0=0$. Dash-dotted curves: $T=0.04e^2/C_\Sigma$ and $Q_0=0$ (circles), 0.1e (squares), and 0.2e (triangles).

The observed sub-Coulomb-gap current could alternatively be explained by an effective sample temperature that is significantly higher than the mixing-chamber temperature, e.g., because of noise and interference. Therefore, in Fig. 3(b) we compare the same measurements with calculations for classical thermally assisted charge transfer, with the same scaling as in Fig. 3(a). There is clearly not a convincing agreement. To obtain rough similarity of calculations to measurements, it is necessary to assume a misadjustment of the gate voltage corresponding to a Q_0 of as much as 0.2e, with, in addition, systematically more error for the lower-R_t samples. Also, a high temperature of about 70 to 100 mK must be

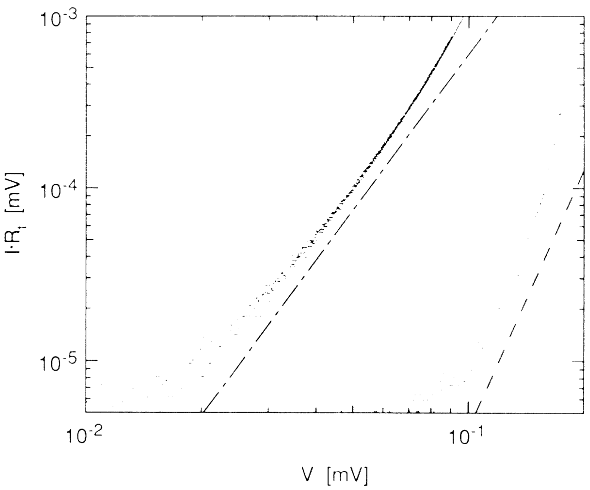

FIG. 4. I-V curves for a double junction with $R_t = 78$ kΩ (left) and three junctions in series with $R_t = 84$ kΩ (right). As a guide to the eye, lines for which $I \propto V^5$ (dashed) and $I \propto V^3$ (dash-dotted) have been drawn, representing the q-MQT prediction for the slope of the log(I)-log(V) curves.

used to obtain a calculated curve that is at least in the range of the measurements. In previous experiments on similar devices (e.g., Ref. 6), using the same low-pass filters, we have found that the effective temperature of our devices is not larger than about 50 mK. The effective temperature of electrons in a central electrode will be higher due to creation of the electron-hole excitations. We find that our observations cannot simply be explained by heating. We will address the complicated matter of cooling of the hot electrons created by tunneling across the Coulomb gap in a separate publication.

Comparison of the log(I)-log(V) curves of a double junction and a three-junction array of almost equal R_t, in Fig. 4, also provides support of q-MQT. The prediction from Eq. (3) is that for a double junction $I \propto V^3$, whereas for three junctions $I \propto V^5$. As a guide to the eye, two lines give the expected slopes, which are in fair agreement with the results. Because of the higher exponent of I, the voltage range in which the current is neither unobservably small nor for a significant amount due to thermally assisted transfer is smaller for the longer array.

We conclude that for the first time we observed macroscopic quantum tunneling of the electric charge through the Coulomb energy barrier in linear arrays of two small tunnel junctions. As a result of this tunneling a finite current flows through the array even in the Coulomb-blockade regime. The current is in quantitative agreement with theoretical predictions.

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[a]Present address: Department of Physics, University of California at Berkeley, Berkeley, CA 94720.

[b]Permanent address: Department of Physics, Moscow State University, Moscow 119899 GSP, U.S.S.R.

[1]See, e.g, the review by G. Schön and A. D. Zaikin, Phys. Rep. (to be published).

[2]A. O. Caldeira and A. J. Leggett, Ann. Phys. (N.Y.) **149**, 374 (1983).

[3]D. B. Schwartz, B. Shen, C. N. Archie, and J. E. Lukens, Phys. Rev. Lett. **55**, 1547 (1985); J. M. Martinis, M. H. Devoret, and J. Clarke, Phys. Rev. B **35**, 4682 (1987), and references therein.

[4]D. V. Averin and A. A. Odintsov, Phys. Lett. A **140**, 251 (1989).

[5]K. K. Likharev, IBM J. Res. Dev. **32**, 144 (1988).

[6]L. J. Geerligs, V. F. Anderegg, P. A. M. Holweg, J. E. Mooij, H. Pothier, D. Esteve, C. Urbina, and M. H. Devoret, Phys. Rev. Lett. **64**, 2691 (1990).

[7]G. Schön and D. V. Averin, in Proceedings of the NATO Advanced Study Institute on Coherence in Mesoscopic Systems, Les Arcs, France, April 1990 (Plenum, New York, to be published).

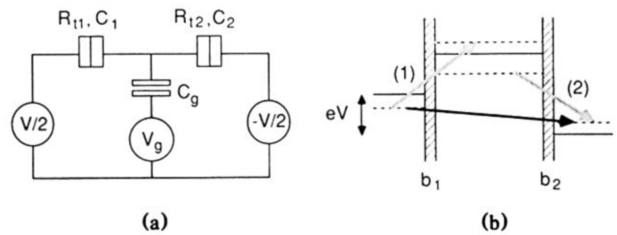
[8]L. I. Glazman and K. A. Matveev, Pis'ma Zh. Eksp. Teor. Fiz. **51**, 425 (1990) [JETP Lett. (to be published)].

[9]D. V. Averin and Yu. V. Nazarov, Phys. Rev. Lett. **65**, 2446 (1990).

[10]Yu. V. Nazarov, Zh. Eksp. Teor. Fiz. **95**, 975 (1989) [Sov. Phys. JETP **68**, 561 (1989)].

[11]E. Simánek, J. Phys. Soc. Jpn. **54**, 2397 (1985).

[12]The measurements have been compensated for offset voltages by shifting the intersection of the $Q_0 = 0$ and $Q_0 = e/2$ curves to the origin.



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