## Metrological Accuracy of the Electron Pump

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We have operated a five-junction electron pump with an error for transferring electrons of approximately 0.5 part per  $10^6$ . The error predicted from existing theory is several orders of magnitude smaller, thus implying that our present understanding of the Coulomb blockade is incomplete. We conjecture that the errors arise from photon-assisted tunneling, where the photon energy is supplied by noise from the environment.

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The Josephson and quantum Hall effects enable very accurate metrological standards of voltage and resistance to be made. Recent progress on Coulomb blockade devices allow charge to be determined by the counting of electrons [1—3] and may similarly give an intrinsic standard of capacitance and a new measurement of the fine structure constant [4]. The performance of this new standard is limited by the ability to transfer electrons through a circuit with very small errors. Although previous experiments have tested the electron pump [2] and a related device, the electron turnstile [3], to an accuracy of about  $10<sup>3</sup>$  parts per million (ppm), metrological applications require errors lower than 1 ppm. Proposed digital circuits [1) require presumably even lower errors.

In this Letter we present experimental data for an electron pump that operates with errors slightly less than 1 ppm, thus demonstrating the feasibility of using the device for metrological applications. We explain several important design criteria and measurement techniques that are necessary to achieve this accuracy. The experiment also tests fundamental assumptions and our understanding of Coulomb blockade phenomena. We find that our measured errors are significantly higher than predicted, thus showing the need for a greater understanding of all possible electron-tunneling processes.

A schematic of the experimental circuit is shown in Fig. 1(a). Our electron pump is made from a linear array of five ultrasmall tunnel junctions and four capacitive gates, where the voltage  $V_{gi}$  applied to gate i polarizes the island between two adjacent junctions with charge  $Q_i = C_{qi} V_{qi}$ . An appropriate time sequence of island polarizations, as shown in Fig. 1(b), then causes an electron to tunnel sequentially through all of the junctions [5]. When an electron is transferred through the pump it charges the capacitance  $C_p$  with a resulting change of the pump voltage  $\Delta V_{\text{pump}} = e/C_p$ . This voltage change is measured with a voltmeter based on the single-electron transistor which has an input gate capacitance  $C_g$ . The values of  $C_p$  and  $C_g$  were chosen so that a single electron can be detected by the circuit [6, 7], while not significantly altering the voltage bias across the pump due to the tunneling of a few electrons.

The operation of the pump can be broken down into a sequence of tunneling events through the individual junctions. Figure 1(c) plots the capacitive energy  $E_n$ of an electron on island  $n$  at a bias where the tunneling of the first junction is allowed. The lowest energy state has an electron in the first island, and the pump operates without error when this state is filled by an electron tunneling through the first junction. This corresponds in Fig. 1(c) to a  $0 \rightarrow 1$  transition (solid arrow). Previous calculations have shown that pumping errors can be classified and predicted by three error processes [5, 8]. The first is due to errors from thermally assisted tunneling over the Coulomb barrier and corresponds to the  $5 \rightarrow 4 \rightarrow 3 \rightarrow 2 \rightarrow 1$  transitions (dashed arrows) in Fig  $1(c)$ . This gives a pumping er-



FIG. 1. (a) Electrical schematic of the circuit. The boxed symbol is an ultrasmall tunnel junction. The application of appropriate pulses to gate lines  $V_{gi}$  pumps an electron through the device. This changes  $V_{\text{pump}}$  by  $e/C_p$ , which is then detected by the voltmeter. (b) Sequence of charge polarizations applied to the metal islands between the pump junctions that produces the optimal pumping. Traces  $Q_2, Q_3, Q_4$  are offset for clarity. The sequence corresponds to an electron pumping out and then into the device. (c) Coulomb energy  $E_n$  versus an electron on island  $n$ , at time indicated by dashed line in (b). Solid, dashed, and dotted arrows are the wanted, thermal activation, and cotunneling processes, respectively.

ror  $\epsilon_{\text{th}} = 20 \exp[-0.8e^2/2Ck_BT]$ , where T is the electron temperature. Errors below 0.001 ppm can be attained at 100 mK with tunnel junctions having capactron temperature. Errors below 0.001 ppm can be at tained at 100 mK with tunnel junctions having capacitance  $C < 0.4$  fF. Errors from the simultaneous tunneling of electrons through four junctions [9, 10] (cotunneling) correspond to the  $5 \rightarrow 1$  transition (dotted arrow). This error is less than 0.001 ppm for pumps with five or more junctions each with a resistance  $R_t$  above 60 k $\Omega$ . A third source of error arises from missing the desired tunneling process due to pumping at too fast a rate. For a total pumping time of  $\tau_{\text{pump}}$ , the error is  $\epsilon_{\tau} = \exp(-a\tau_{\text{pump}}/R_tC)$ , where  $a \approx 0.02$ .

The sample was fabricated using standard techniques of electron beam lithography and double angle evaporation. A magnet placed under the substrate brought the Al tunnel junctions into the normal state. An electrical contact to the pump was necessary for the measurement of  $C_g$ ,  $C$ , and  $R_t$ , and the tuning of the crosscapacitance cancellation of the gates as discussed below. We directly measured  $V_{\text{pump}}$  through a mechanical switch that could be connected in situ. The switch was made from a magnetically controlled needle that contacted a 100  $\mu$ m by 100  $\mu$ m Au pad on the sample which was part of the thin film capacitor  $C_p$ . We used a sapphire substrate because the mechanical contact of the switch produced stress-induced charge noise with an oxidized Si substrate. The gate capacitance  $C_g = 1$  fF of the voltmeter was formed from interdigitated metal electrodes [11] and restricted the operating temperature to below 200 mK. The capacitance  $C_p$  was determined from a measurement of  $\Delta V_{\text{pump}} = e/C_p$  to be 33 fF. The current-voltage characteristic of the pump gave a measurement of  $C = 0.43 \pm 0.02$  fF from the Coulomb gap, and  $R_t = 300 \pm 15$  k $\Omega$  from the differential resistance well above the gap. Here we assume all junctions have the same  $C$  and  $R_t$  because of the reproducibility of our lithography and because our predictions do not vary greatly with small asymmetries in  $C$  and  $R_t$ .

Our actual circuit is slightly more complicated than depicted in Fig. 1 because of stray capacitances. We have computed that small strays can be accounted for by adding their capacitance to the junction capacitance [12]. The stray capacitances of the islands were reduced to less than 0.15 fF by fabricating the pump with island dimensions approximately 0.8  $\mu$ m. This design, however, created a significant cross-capacitance between a given gate line and adjacent islands. For example, the capacitance between gate line 1 and island 2 was almost 40% of the capacitance between gate line 1 and island 1. This cross-capacitance affects the operation of the pump since a gate voltage polarizes charge in more than one island. We have calculated that the optimum biasing [5] is given by a charge polarization  $Q_1$  through  $Q_4$  as shown in Fig. 1(b). This optimal polarization can be produced from the voltage gates by electronically canceling the crosscapacitance with small negative voltages applied to sdjacent gate lines. The magnitude of the cancellation voltage is first estimated by computing a cross-capacitance matrix. An inversion of this matrix then gives the appropriate ratios of the gate voltages. A further adjustment for the actual device is based on the current-voltage characteristics of the pump being e periodic in the charge polarization of any island. The relative magnitudes of the cancellation voltages are changed so that, at constant current bias, there is no change in  $V$  versus  $V_{gi}$  when a change in polarization of e is made to an island.

At low temperatures, small voltages, and when the islands are not polarized, the Coulomb gap blocks the tunneling of electrons through the pump. Hence, the number of electrons on  $C_p$  and  $V_{\text{pump}}$  are held constant. We name this state the "hold" mode. In Fig. 2(a) we plot the time dependence of  $V_{\text{pump}}$  for the hold mode. The voltage is constant over several seconds, apart from noise intrinsic to the voltmeter. However, quantized shifts in voltage  $\Delta V_{\text{pump}} = e/C_p$  are observed [6, 7], for example, at  $t \approx 0.3$  s in Fig. 2(a). A shift implies that an electron has tunneled through the pump, which corresponds to an error.

If the bias voltage  $V_{\text{pc}}$  of the pump capacitor is varied when the pump is in the hold mode, then  $V_{\text{pump}}$ should change linearly with  $V_{\text{pc}}$ . This is what was observed for small voltages, but beyond a critical voltage  $V_{\rm cr} \sim 460~\mu \rm V$   $V_{\rm pump}$  ceased to change. This occurs because for large  $V_{\text{pump}}$ , electrons can tunnel and consequently discharge  $C_p$ . The critical voltage depends on the bias applied to the gates and is determined by the voltage at which the tunneling rate of the pump  $\Gamma$  is



FIG. 2. Plot of pump voltage versus time. Arrows indicate  $\Delta V_{\text{pump}} = e/C_p$ . Traces (b)–(g) are offset for clarity. (a) Pump in hold mode. (b) Pumping of  $+2e$ , then  $-2e$ , where  $\tau_{\text{pump}} = 290$  ns and  $\tau_{\text{cycle}} = 20$  ms between pumping of two electrons. (c) Same as (b) but with pumping of  $\pm 4e$ . (d) Same as (c), but with  $\tau_{\text{cycle}} = 16 \text{ }\mu\text{s}$  between pumping of four electrons. (e) Same as (d), but with  $Q_4$  adjusted from optimal biasing by 0.30e. (f) Same as (d), but with  $Q_4$  adjusted by 0.42e. (g) Pumping of  $\pm 1e$  with  $\tau_{\text{pump}} = 290$  ns and  $\tau_{\text{cycle}} = 0.5 \ \mu \text{s}$ . Error rate of 1 s<sup>-1</sup> gives 0.5 ppm pumping error.

equal to the electron charging rate of  $C_p$ . For our experiment this gives  $\Gamma = (dV_{\text{pc}}/dt)/(e/C_p) \sim 1000 \text{ s}^{-1}$ . Thermal activation gives a tunneling rate in the hold mode and zero charge bias  $\Gamma \approx (R_tC)^{-1} \exp[-\Delta E/k_BT]$ , where  $\Delta E = 1.2(e^2/2C)(1 - 2C|V|/3e)$  for  $|V| < e/C$ . From our measured parameters of  $\Gamma$  and  $R<sub>t</sub>C$  we find  $\Delta E/k_BT \approx 16$ . This also predicts that the critical voltage should decrease linearly with  $T$ . We adjusted the gate biases of the pump for maximum critical voltage and then measured the critical voltage versus temperature between 40 and 200 mK. The data fell on a line and had intercepts at zero voltage of 200 mK and zero temperature of 590  $\mu$ V. These data correspond to a second measurement of  $C$  for biasing within the Coulomb gap and give a value of  $C = 0.38 \pm 0.06$  fF.

Figure 2 also shows data for the pumping of electrons, where the offset of the gates was adjusted to zero charge bias which gives minimum pumping errors [5], as discussed below. In Fig. 2(b) we plot  $V_{\text{pump}}$  versus time, where we pump two electrons in or out of the pump every 20 ms. The pump sequence for these data took only 390 ns per electron, with the rest of the time spent with the pump in the hold mode. The charging and discharging of  $C_p$  are clearly observed. Figure 2(c) shows similar data, but now for the pumping of four electrons. As expected, the voltage change for (c) is twice as large as for (b). In Figs.  $2(b)$  and  $2(c)$ , the observed error rate is not significantly greater than for (a), where the pump is maintained in the hold mode. In order to test the pump to a higher accuracy, we pump electrons in and out at a higher rate. Figure 2(d) shows data taken for four electrons pumped in or out of the pump every 16  $\mu$ s. Although the electrometer does not have sufficient bandwidth to detect the fast voltage changes, the constant average voltage of  $V_{\text{pump}}$  indicates that exactly four electrons were repeatedly pumped in and out of the device for the 2 s span of the trace. Thus no pumping errors were seen with a total number of pumped electrons of  $5 \times 10^5$ .

This measurement of the accuracy assumes that pump errors are random; that is, pumping errors do not cancel. This is a good assumption since each pump sequence acts independently. As a test that the experiment actually measures pumping errors, we show in Figs.  $2(e)-2(f)$  the result of an increased error rate of pumping. Figure 2(e) is data taken similarly as for (d), but the charge bias on island 4 is adjusted away from its optimal value by 0.30e. Here single-electron errors are clearly observed with a rate of about 10 s<sup>-1</sup>. In (f) this bias is further adjusted to give a correspondingly higher error rate. The errors in this case occur frequently enough that single-electron errors are not observed, but the large fluctuations of  $V_{\text{pump}}$ as compared to (a) imply that single-electron errors are present. Figure 2(e) shows that the error rate increased only by a factor of 10 when one gate was adjusted away from the optimal bias by the large amount  $\pm 0.30e$ . This indicates that the pumping error rate is not sensitively dependent on the gate biases. Likewise, we found it easy to adjust the charge biases for minimum pumping errors.

Figure  $2(g)$  shows our best results, where an electron was pumped in or out of the device every  $\tau_{\text{cycle}} = 0.5 \,\mu\text{s}$ .<br>With a measured error rate of about 1 s<sup>-1</sup>, a pumping error of 0.5 ppm is obtained.

In Fig. 3(a) we plot the pumping errors  $\epsilon$  versus pumping time  $\tau_{\text{pump}}$ . The data with large errors were taken with a large hold time between pumping sequences in order to have an error rate of about  $10 s^{-1}$ , which was easily measurable. The exponential dependence on pumping time for small  $\tau_{\text{pump}}$  is in agreement with theory, but for large  $\tau_{\mathrm{pump}}$  the error becomes independent of time at about 1 ppm. The observed error at small  $\tau_{\text{pump}}$  follows  $\epsilon_{\tau} = \exp(-0.012\tau_{\text{pump}}/R_{t}C)$ , as compared to the predicted error  $\exp(-0.02\tau_{\rm pump}/R_{t}C)$ . The agreement is good, with the operating speed of the pump 40% lower than predicted. This is not unexpected because the device is probably not biased perfectly.

Figure 3(b) plots the pumping errors versus pump voltage for two values of  $\tau_{\text{pump}}$ . Here we observe a rapid decrease in pump accuracy as the voltage increases. The voltage at which the pump accuracy approaches zero is only slightly lower than  $e/2C$ , the voltage where we expect lower order cotunneling to occur [5]. This observed behavior is consistent with computer analysis of the pump errors with stray capacitances and shows that the pump has to be operated with voltages lower than about e/4C.

The error rate for pumping in Fig. 2(g),  $1 s^{-1}$ , is higher than for the hold mode, about  $0.1$  s<sup>-1</sup>. A larger



FIG. 3. (a) Pumping error  $\epsilon$  versus  $\tau_{\text{pump}}$  at  $V_{\text{pump}} \approx 0$ . Dashed line is prediction with  $C = 0.43$  fF,  $R_t = 300$  k $\Omega$ , and  $a = 0.012$ . Uncertainty in  $\epsilon$  for each point is due to systematic uncertainties in biasing the pump and is approximately a factor of 2. (b) Error  $\epsilon$  versus magnitude of  $V_{\text{pump}}$  for two values of  $\tau_{\text{pump}}$ . Uncertainty is the same as in (a). Arrow indicates e/2C.

error rate is expected during pumping because the errors arise from an energy barrier corresponding to only four junctions. However, in both cases the observed error rates are at least 2 orders of magnitude greater than predicted from thermaL activation and cotunneling. In order to focus on the cause of these large error rates, we will discuss the simpler case of the electron leakage rate through the pump for the hold mode. This mode also has the advantage that no power is dissipated in the pump, so the electron temperature should be close to the refrigerator temperature. Understanding and decreasing the leakage rate in the hold mode will presumably improve the accuracy when pumping electrons.

We fabricated two similar devices which did not have the pump and found that the measurement of the voltage across  $C_p$  versus time did not contain the single-electron shifts. This indicates that the leakage errors we observe are indeed due to the tunneling of electrons through the pump. A previous experiment for a four-junction pump in the hold mode [7] measured tunneling rates of about  $10 s<sup>-1</sup>$ . We measured four different samples with fivejunction pumps; all had error rates from 2 to 0.1 s<sup>-1</sup>. These rates did not change with relatively large changes in bias of  $\Delta Q_i \sim 0.2e$  and  $\Delta V_{\text{pump}} \sim 200 \ \mu\text{V}$ . The pump error rate in the hold mode decreased from about  $1 \text{ s}^{-1}$ when the sample was first cooled and to about  $0.1 \text{ s}^{-1}$ after the sample remained at 40 mK for 1 week. The leakage rates also can fluctuate with time. For example, in Fig. 2(a) we find two errors occurring together at  $t \approx$ 0.3 s followed by a large time interval in which additional errors are not observed.

The leakage rates were approximately proportional to T from 40 mK to 100 mK. This is inconsistent with predictions because thermal activation predicts an exponential dependence on temperature and thermally enhanced cotunneling gives a dependence of  $T<sup>8</sup>$ . Thermally enhanced cotunneling and thermal activation predict rates of  $10^{-9}$  s<sup>-1</sup> nneling and thermal activation predict rates<br>and  $10^{-21}$  s<sup>-1</sup>, respectively, at 40 mK. Even<br> $\frac{1}{2}$ with  $T = 80$  mK, the predicted rates of  $10^{-6}$  s<sup>-1</sup> and  $10^{-5}$  s<sup>-1</sup> are still much smaller than measured. The measurement of  $V_{\text{cr}}$  gives the argument of the exponential for<br>thermal activation to be  $\Delta E/k_BT \approx 16$  at  $T = 200$  mK. At 80 mK then  $\Delta E/k_BT$  should be 2.5 times larger, thus giving a negligible thermal leakage rate.

These errors must arise from a process other than cotunneling or thermal activation. We think the most likely candidate is photon-assisted tunneling and cotunneling [13]. This mechanism takes into account that the effective temperature of the environment (the leads attached to the device) can be higher than the temperature of the electrons in the tunnel junctions, the temperature that is normally considered in thermal and cotunneling calculations. Noise from the environment provides the photon energy that is needed to make two one-junction transitions or a two-junction cotunneling transition, which subsequently allows an electron to tunnel through the entire pump. This process is similar to thermal activation, but here the energy is supplied by the environment. We calculate that even low microwave power can cause significant error rates. For example, our observed leakage rates can be explained only if  $10^{-6}$  of the thermal noise power at 50 GHz which is generated at 4 K reaches the device.

All experimental leads include carefully designed radio and microwave filters [14]. Since the electron leakage rates were unchanged when we removed the microwave filters that were at the sample temperature, we do not suspect that the noise is due to the 4 K thermal source. We think that the noise may arise from charge traps that are present in the sample mount or substrate, which slowly relax with time. Since the energy release from trap states can decrease with time [15], this hypothesis is consistent with our observation that the leakage rate decreased by a factor of 10 after 1 week. If the noise source is intrinsic to materials at low temperatures and cannot be eliminated, we think a pump with seven junctions should still be able to reach errors below 0.001 ppm.

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- [1] D. V. Averin and K. K. Likharev, in Mesoscopic Phenomena in Solids, edited by B. Al'tshuler, P. Lee, and R. Webb (Elsevier, Amsterdam, 1991), Chap. 6; Single Charge Tunneling, edited by H. Grabert and M. H. Devoret (Plenum, New York, 1992), Chaps. 2 and 3.
- [2] H. Pothier, P. Lafarge, C. Urbina, D. Esteve, and M. H. Devoret, Europhys. Lett. 17, 249 (1992).
- [3] 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).
- [4] E. R. Williams, R. N. Ghosh, and J. M. Martinis, J. Res. Natl. Inst. Stand. Technol. 97, 299 (1992).
- [5] H. D. Jensen and J.M. Martinis, Phys. Rev. B46, 13<sup>407</sup> (1992).
- [6] T. A. Fulton, P. L. Gammel, and L. N. Dunkleberger, Phys. Rev. Lett. 67, 3148 (1991).
- [7] P. Lafarge, P. Joyez, H. Pothier, A. Cleland, T. Hoist, D. Esteve, C. Urbina, and M. H. Devoret, C.R. Acad. Sci. Paris 314, 883 (1992).
- [8] H. Pothier, P. Lafarge, D. Esteve, C. Urbina, and M. H. Devoret, IEEE Trans. Inst. Mess. 42, 324 (1993).
- [9] D. V. Averin and A. A. Odintsov, Phys. Lett. A 140, 251 (1989).
- [10] D. V. Averin and Yu. V. Nazarov, Phys. Rev. Lett. 65, 2446 (1990).
- [11] G. Zimmerli, R, L. Kautz, and J. M. Martinis, Appl. Phys. Lett. 61, 2616 (1992).
- [12] H. D. Jensen and J. M. Martinis (to be published).
- [13] R. Bauernschmitt and Yu. V. Nazarov, Phys. Rev. B 47, 9997 (1993); J. M. Martinis and M. Nahum, Phys. Rev. B 48, 18316 (1993).
- [14] J. M. Martinis, M. H. Devoret, and J. Clarke, Phys. Rev. B 35, 4682 (1987).
- [15] R. C. Richardson and E. N. Smith, Experimental Techniques in Condensed Matter Physics at Low Temperatures (Addison-Wesley, Reading, MA, 1988), Chap. 3.2, p. 124.