Meas. Sci. Technol. **14** (2003) 1237–1242 PII: S0957-0233(03)57589-0

Electrical metrology with single electrons

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Received 16 January 2003, in final form 4 April 2003, accepted for publication 17 April 2003
Published 16 July 2003
Online at stacks.iop.org/MST/14/1237

Abstract
This paper is mostly a review of the progress made at NIST in pursuing a capacitance standard based on the charge of the electron. We briefly introduce the Coulomb blockade, which is the basic physical phenomenon allowing control of single electrons, describe two types of single-electron tunnelling (SET) device and describe the metrology goals and payoffs achievable from SET devices. We then discuss the electron-counting capacitance standard (ECCS): the motivation, previous experimental work on various critical elements, present status and future prospects. This last part includes using the ECCS for a practical representation of capacitance, as well as pointing out that we can close the quantum metrology triangle without needing a large-value current standard. Finally, we briefly review other SET-based metrological applications.

Keywords: electrical metrology, single-electron devices, ECCS

1. Introduction

1.1. Applications of SET devices

The ability to make devices that monitor and control single electrons has excited a large number of potential applications. One obvious one is for the future of semiconductor electronics: *The International Technology Roadmap for Semiconductors* [1] lists single-electron tunnelling (SET) devices as one of the candidate technologies for the time after advances in CMOS circuits have stopped. Among the areas researched have been floating gate-based memory, and various logic architectures [2].

It is clear that the market forces driving development of electronics are much larger than those for metrology. In that context, it is telling that the only application which is nearing fruition is a standards one, and in particular the electron counting capacitance standard (ECCS), which is the main subject of this paper; this may be an illustration of how metrologists can pursue a technology too complex for more general applications!

In general, the metrology applications are for a standard of charge, capacitance or current. The scientific and metrological payoffs of one or more of these applications include the following (some of these are described in greater detail later):

Turnkey capacitance standard. In analogy with the Josephson and quantum Hall standards, the ECCS offers the potential to form a better fundamental capacitance representation (*but not a realization* [3]) than artifactual standards (i.e., silica dielectric capacitors). Because it is not a realization of the SI farad, it cannot replace the calculable capacitor.

Fine-structure constant α. By combining the ECCS with the calculable capacitor and the Josephson voltage standard (and assuming that the Josephson formula is exactly correct), a value of α can be measured [4]. This approach is complementary to the approach using the quantum Hall resistance (QHR) standard [5]; in particular, by assuming that the formula for the QHR is exactly correct, we can obtain h/e^2 in SI units, and thus α from $\alpha = \mu_0 c(2h/e^2)$. The approach using the ECCS may be valuable in that we can compare it to other various methods of measuring α, but it is unlikely to improve upon the current recommended value, which has a relative uncertainty of 3.7×10^{-9} [6].

[3] www.eeel.nist.gov/811/femg/set.html
[4] Electronics and Electrical Engineering Laboratory, Technology Administration, US Department of Commerce. Official contribution of the National Institute of Standards and Technology, not subject to copyright in the United States.

Closing the 'quantum metrology triangle' [7]. By combining the delivered charge or current from a SET pump with both the Josephson voltage and QHR standards, we can test the consistency of the three fundamental relations (the SET pump delivers exactly 1 e, the Josephson standard develops a voltage of exactly $2\ ef/h$ and the quantum Hall standard develops a resistance of exactly h/ne^2).

1.2. *A brief review of Coulomb blockade and single-electron tunnelling (SET)*

The ability to monitor or control the motion of *single* electrons in a conductor is an amazing feat. It is made possible by the 'Coulomb blockade' [8], which refers to the physical phenomenon that occurs when the energy to charge a conductor with *one additional electron* [9] becomes a significant impediment to charge transfer.

In particular, for an isolated conductor with total capacitance to all other conductors of C_Σ, the 'Coulomb blockade energy' is

$$E_C = e^2/2C_\Sigma.$$

This represents the energy necessary to add one additional electron to the conductor (leaving aside external voltages); more generally, E_C represents the typical energy scale of the effect, and is thus important for comparison to effects of temperature, radiation etc.

About 15 years ago, a controllable device, the single-electron tunnelling transistor (SETT), was invented [10]. This device was fabricated using metal lines of aluminium, with the isolated conductor (the 'island') separated from the environment by AlO_x tunnel junctions. The lumped circuit element of the SETT is shown in figure 1(A), and a micrograph of a typical device from our group is shown in figure 1(B).

Figure 1(A) shows the basic features necessary to achieve SET action: an isolated conductor connected to the environment by high-resistance tunnel junctions. The high resistance is necessary to ensure that electrons transfer onto or off the central conductor in units of 1 e; if the tunnel junctions do not have a resistance larger than the resistance quantum, $h/e^2 \approx 26\,000\ \Omega$, then the electrons are not well localized on the island, and thus quantum fluctuations of the charge suppress the Coulomb blockade [8].

In addition to the resistance criterion, the value of the capacitance, through E_C, must be balanced against the temperature of operation. A general rule of thumb for operation of a SETT is that $3k_BT \lesssim E_C$; for a SET pump to achieve low-error-rate operation such as we describe later requires much lower temperatures. For the typical $Al/AlO_x/Al$ SETT, we can achieve a C_Σ as small as about 0.1 fF; this corresponds to a maximum temperature of a few kelvins. To increase the signal to noise ratio, and more importantly, to minimize errors, we typically operate the devices below about 0.1 K; thus, typical device parameters are C_Σ of 0.1–1.0 fF, junction resistance of 100 kΩ–1 MΩ and temperatures below 1 K.

1.3. *SET transistors*

Similar metal-based (usually Al) SET devices have formed the bulk of the devices used for SET metrology, and we thus discuss in some detail the elements in figure 1. For devices satisfying the above criteria, and operated in a properly shielded environment (SET devices are highly susceptible to interference from radiation at or above $E_C/h \approx 20$ GHz), the device shown in figure 1 acts as a transistor. In particular, if we apply a bias voltage of order 1 mV from source to drain, the current through the device is by 'sequential tunnelling'—only one additional electron can tunnel on to the island, and one electron must then tunnel off before the next one can tunnel on. This fact leads to the typical current value of $1e/RC \approx 10$ nA.

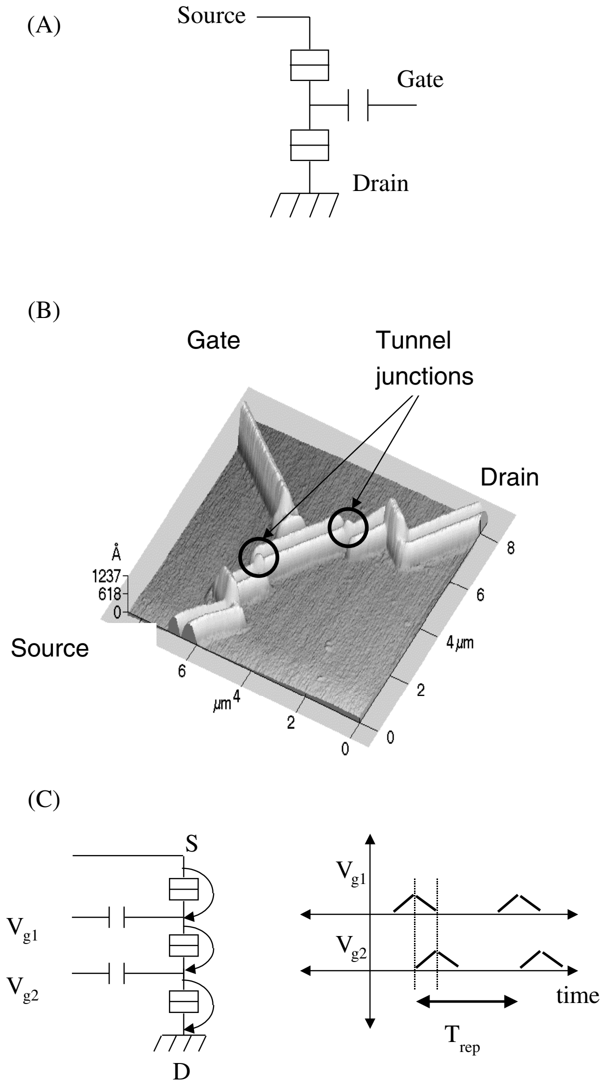

Figure 1. (A) Circuit model for the SETT. This device has source, drain and gate in analogy with a FET; the two rectangles represent ultra-small tunnel junctions, for which the capacitance is small enough that Coulomb blockade is important. (B) Atomic force micrograph of an $Al/AlO_x/Al$ SETT. The substrate is a Si wafer; there are duplicate lines in the direction perpendicular to source and drain, as produced by the self-aligned fabrication. (C) Left: circuit model for SET pump, with three tunnel junctions, two gates and two islands. With the proper application of gate pulses, one electron will transfer sequentially from source to drain. Right: schematic representation of the proper gate pulses. The height of the pulse on V_{g1} is that necessary to move one electron from the source to the top island. As the pulse on V_{g1} ramps down, a similar pulse on V_{g2} ramps up, inducing an electron to tunnel to the lower island. Finally, as V_{g2} ramps down, an electron tunnels to the drain.

In addition, this current is controllable by the static voltage applied to the gate, or the charge deposited on the gate, with a charge sensitivity of about 10^{-4} e in a 1 Hz bandwidth. This makes the SETT by far the most sensitive charge electrometer (although the very small gate capacitance makes the effect of stray capacitance particularly deleterious, and can thus reduce the effective resolution by several orders of magnitude [11]).

This 'transconductance', i.e., the sensitivity of source–drain current to gate voltage, leads to the terminology of the SETT, and the notation of source, drain and gate, both in analogy with standard field-effect transistors. This also obviously leads to suggestions of the use of such devices as logic or memory.

We should mention that essentially all of the metrological work in SET devices [12] (except for the SET surface acoustic wave (SAW)—see the last section) has been performed in Al/AlO$_x$/Al devices. However, there have been a wide variety of materials systems in which Coulomb blockade has been claimed, including carbon nanotubes and granular Si films. The major reason for these investigations has been to raise the maximum operating temperature. Perhaps the best and most controlled work of this type has been done in CMOS-compatible Si-on-insulator devices, with SETT action up to 100 K in some cases [13].

1.4. SET pumps

Having discussed the SETT, we now turn to the other SET device important for metrology—the SET pump [14]. Amazingly enough, this device gives us the capability to move and store single electrons! Figure 1(C) shows the simplest version of a SET pump; it is again the SETT, with now one additional tunnel junction and gate, and thus two islands. The addition of the second island allows us to trap charge: as in the SETT, one electron is free to tunnel from the source to the first island. However, no electron can tunnel off this island until we apply voltage to the second gate and lower the energy barrier for this process. With a sequence of gate voltages [15] such as that shown in figure 1(C), we can lower the energy barrier for tunnelling at one junction at a time and pass an electron through an array of any number of junctions. It is important to note that the pump operates without any voltage across the array, and that the current can flow in either direction, depending on the order in which the gates are pulsed.

With this type of device, we can transfer one and only one electron per cycle (time T_{rep}). At first glance, it seems that the most attractive metrology application of the SET pump would be as a current standard: cycle the device at frequency f to achieve a fundamental representation (not realization) of current $I = ef$. However, there are several reasons why that is in fact not the most attractive option at present: first, there is the simple observation that, unlike the cases of voltages or resistances or charges or capacitances, *devices that store an unchanging current are not readily available*. Thus, as a practical representation, capacitance is often a more convenient standard than current. In addition, because the Josephson and quantum Hall standards can already be used to define a current (by $I = V/R$), a SET-based current representation would only be attractive if it were simpler and less expensive than either the Josephson or quantum Hall standards (this looks unlikely at present).

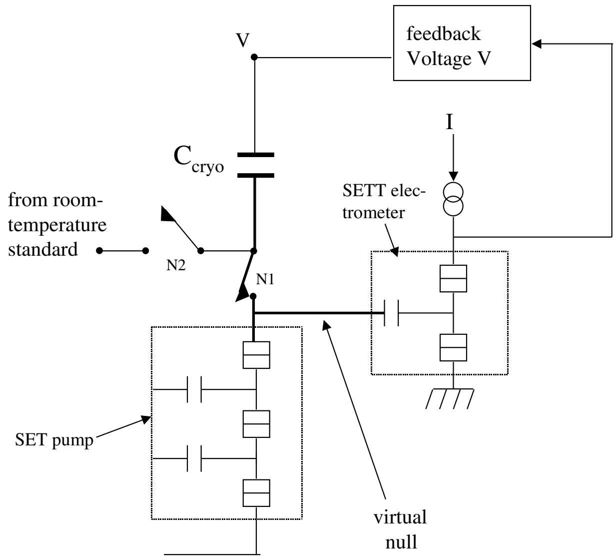

Figure 2. Circuit model for the ECCS [4, 21]. A counted number of electrons are passed to the central node using the SET pump. By ramping the feedback voltage, a virtual null is maintained at this node, using the SETT electrometer as a null detector. The virtual null ensures that all electrons move to the plate of C_{cryo}; the voltage across C_{cryo} is defined by $Ne = C_{\text{cryo}}V$. In a second phase of the experiment, N1 opens and N2 closes, allowing comparison to a room-temperature standard. This is used either to calibrate C_{cryo} from the calculable capacitor, or to use the ECCS to calibrate a customer's artifactual standard.

Finally, a more fundamental difficulty with a SET-based current standard is the achievable value of current: as discussed above, the typical RC time constant is fairly large in these devices; to deliver a current with error rate of about 10^{-8} leads to a maximum current of about 10 pA [15]. This value is quite small, and it appears that it will be difficult to make a useful standard from it. Several groups are considering the feasibility of closing the quantum metrology triangle with this current [16–20]; others are pursuing various ways of substantially increasing the value (see the section below).

2. The electron-counting capacitance standard (ECCS)

2.1. Motivation

The limitation on the current value of SET pumps has been appreciated for a long time. Before either author joined NIST, co-workers had proposed a different approach to a metrological use of the SET pump [4]: *if we pump 1 pA on to a 1 pF capacitor for 1 s, 1 V will develop across the plates of that capacitor*. We will discuss in detail the ECCS for much of the rest of this paper.

2.2. ECCS: the experiment

Figure 2 shows the schematic circuit for the ECCS [4, 21]. The main elements are the SET pump used as a charge source, the SETT used as a null detector and the cryogenic capacitor C_{cryo}. There are two distinct phases of operation of this circuit, and we choose between them using the cryogenic needle switches N1 and N2.

In the first (charging) phase, with N1 closed and N2 open, the pump transfers electrons onto the inside plate of C_{cryo}. As this is happening, the voltage across the pump must be kept near zero to avoid errors. We accomplish this by using the SETT null detector and feedback electronics to ramp the voltage at the outside plate of C_{cryo} at a rate which precisely matches the speed of the pump. This also ensures that all charge transferred through the pump appears across C_{cryo} and not across the stray capacitance from the central node to ground. After about 10^8 electrons have passed through the pump in one direction, we stop pumping and measure V (about 10 V), then pump the same number of electrons in the opposite direction, stop and measure V again (about -10 V). We repeat this process 10–100 times, with each cycle taking about 100 s, to obtain a single value of C_{cryo}.

In the second (bridge) phase, with N1 open and N2 closed, we compare C_{cryo} with another capacitor at room temperature using an ac bridge. This allows us determine the value of a conventional room-temperature capacitor in terms of e, and that capacitor can then be used as a basis for practical calibrations.

We have been aiming for a relative uncertainty of 10^{-8}. Each of the critical elements in this circuit has required substantial work over the past years, and we list some details of each.

SET pump. We require the pump to transfer exactly one electron every cycle (instead of two or zero). Some of the sources of pump error include temperature (we operate below 0.1 K), frequency (repeat frequency below 10 MHz, limiting current to 1 pA) and co-tunnelling errors (requires seven junctions and six islands/gates). With these criteria satisfied, we have achieved an error rate of about four extra or deficit electrons for every billion pumped [22, 23].

Cryogenic capacitor. We require small drift in time, frequency and voltage dependence, and also small leakage. To see the last, we note that we require that the capacitor loses no more than 10^{-8} of its charge; with $C_{\text{cryo}} \approx 1$ pF, for a cycle time of 100 s, we require a parallel leakage resistance of $10^{22}\ \Omega$. We have discovered that a vacuum-gap capacitor, using sapphire supports and Cu electrodes [24], has no frequency and voltage dependence (at the 10^{-6} level so far), and a parallel resistance of at least $10^{21}\ \Omega$ [24, 25].

SETT electrometer. We require the null detector to have a high enough voltage (or charge) sensitivity to achieve 10^{-8} resolution. For the ECCS, the sensitivity of the SETT electrometer is probably no better than a conventional low-noise null detector [26]. However, we also require that the null detector has very low leakage, for the same reason as the capacitor; this drives the choice of a SETT electrometer. The resolution limit of the electrometer used as a null detector is about 10^{-7} [27]; this resolution limit is currently the source of the random uncertainty for the ECCS [21]. The limit is set by 'charge offset' noise, from charged defects moving in the near vicinity of the SETT, and thus causing a fluctuation in time. Reducing this noise is an actively pursued topic for several research groups including NIST.

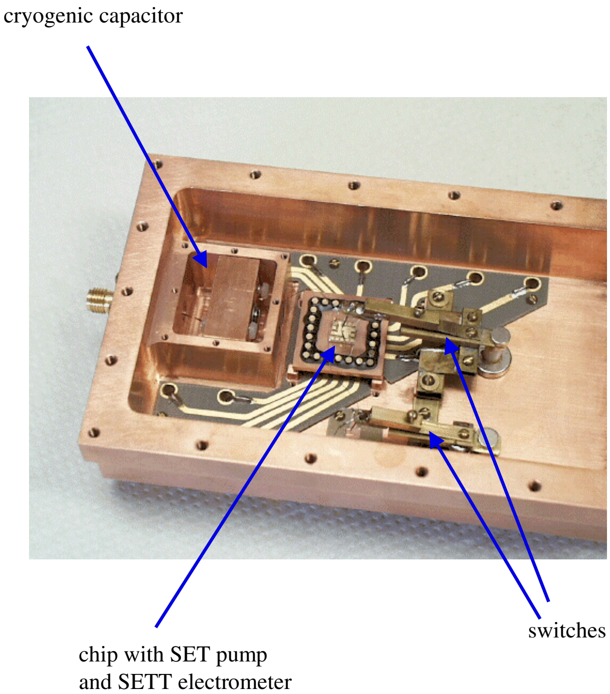

Figure 3. Critical elements of first ECCS prototype, showing the cryogenic capacitor, SET chip and switches.
(This figure is in colour only in the electronic version)

2.3. ECCS: present status and future

2.3.1. Present status. We have assembled a prototype of the ECCS as schematically indicated in figure 2, incorporating the results of the research described above for each of the individual elements [21]. The prototype is shown in the photograph in figure 3. We have indeed succeeded in pumping electrons onto the capacitor and measuring the voltage. As mentioned just above, the limiting random uncertainty arose from the charge offset noise floor of the SETT null detector, with a relative uncertainty of about 0.3×10^{-6}. We have also compared the value of the cryogenic capacitor to a commercial room-temperature standard (operated with a sinusoidal excitation at 1 kHz); this comparison agreed with that derived from pumping electrons, again with a relative uncertainty of about 10^{-6}.

This preliminary result was quite promising, especially given the frequency dependence of the full system: the electron pumping phase uses a non-sinusoidal excitation at a frequency below 1 mHz, and the agreement with a sinusoidal measurement at 1 kHz suggests that the ECCS, and in particular the cryogenic capacitor, has a very small frequency dependence over a remarkably wide range.

Given the promising preliminary result, we have more recently made some advances in the cryogenic capacitor, and in the calibration of that capacitor [28]. First, we have increased the value by about a factor of ten, and are now using a value of $10(1 + 0.03)$ pF. This increase yields two improvements: the first is an improvement in the ultimate use as an artifact for calibrating capacitance meters. Because the noise floor of these meters is typically not dependent on the measured value, the increase in the value will reduce the relative uncertainty of the calibration by the same amount. The second is, for a value near 10 pF, we can measure the value of the cryogenic capacitor to a relative uncertainty [28] less than 5×10^{-8}, by using the inherent tunability of the calculable capacitor, which is only possible for values near 10 pF.

An additional advance is in examination of the frequency dependence of the cryogenic capacitor (not yet published). As mentioned above, we have been able to put an *experimental* upper bound on the frequency dependence, over a limited range of frequencies, of about 10^{-6} [25]. This is not sufficient, both due to the limited range, and because we desire a smaller uncertainty. We have very recently modelled the capacitor in order to determine possible non-ideal sources of frequency dependence. We have concentrated mostly on the possible deleterious effects from surface films, which can introduce both a frequency-dependent real part of the dielectric constant, as well as an imaginary part (dielectric loss); both of these effects will lead to a frequency dependence of the capacitance. Our conclusion is that, at low temperatures, over the whole range of interest, from roughly 0.01 Hz to 1 kHz, there is a relative frequency dependence less than 5×10^{-8}.

2.3.2. Future. We have two goals, one practical and one scientific, requiring different levels of uncertainty.

Practical representation. We feel that to form an attractive practical representation, similar to the Josephson voltage and quantum Hall resistance standards, requires a total relative uncertainty of at most 10^{-7}. We are close to that goal for the random uncertainties in our preliminary prototype. If we can reduce the random uncertainty another factor of three, we will require the following to form a useful practical representation.

(1) *Formation of a robust standard.* The reliability of various components must be improved, and the operation of the standard should be as automated as possible to make it easy and inexpensive to use. We are also exploring the possibility of using a cryogenic platform that is more compact than a conventional dilution refrigerator.
(2) *Uncertainty analysis.* A full analysis of all aspects of the standard is being conducted, with especial emphasis on possible differences between the value of C in the electron counting phase and in the bridge phase.
(3) *Reliability and tunability of cryogenic capacitor.* In our first prototype, the capacitor suffered from occasional hysteretic changes in its value, probably due to mechanical shocks [21]. A cylindrical geometry has alleviated this problem [28, 29].

Measurement of α, and closing the quantum metrology triangle. As described above, the ECCS can be used to measure the fine-structure constant. Unlike the expectations of many, *the capacitance standard can also be used to close the quantum metrology triangle* [30]. As described in Piquemal and Geneves [30], from the ECCS, we obtain

$$Q = CV \Rightarrow I = C\Delta V/\Delta t,$$

where I is the SET pump current used in the ECCS, and ΔV is the voltage change over a ramp time Δt. By an impedance comparison of the quantum Hall resistor and cryogenic capacitor (using a quadrature bridge at frequency ω), we can obtain

$$C = 1/(\omega R).$$

Comparing these two equations yields

$$I = \left\{\frac{\Delta V}{R}\right\} 1 \Big/ (\omega \Delta t).$$

We thus see that, by producing or measuring I, ΔV and R with the SET, Josephson and quantum Hall effects, *we can close the quantum metrology triangle without needing a large-value SET current standard.* To contribute significant new knowledge in a measurement of α or a closure of the quantum metrology triangle, we feel that a total uncertainty of about 10^{-8} must be achieved. In relation to the items listed above, we will thus need to reduce the random uncertainty (mostly from the noise floor of the SETT) by a factor of 30 from its present limit, in addition to assessing all of the systematic uncertainties at the same level.

One new requirement for the measurement of α, and to achieve an uncertainty of 10^{-8}, will be to compare C_{cryo} to the calculable capacitor. This will require either (i) a tunable cryogenic capacitor, or (ii) using the inherent tunability of the calculable capacitor, as mentioned above.

3. Other metrological experiments with single electrons

In this section we give brief descriptions of several other efforts to use SET devices for metrology, mostly to provide a larger value of current. More detailed descriptions of most of these ideas, as well as more complete references to the literature, can be found in a previous paper [23].

(1) *SET-SAW for gigahertz electron pumping.* A two-dimensional electron gas in a heterostructure of GaAs/AlGaAs, similar to those used to make quantum Hall devices, can be patterned into a narrow channel. If this channel is electrostatically squeezed beyond the point where current can flow through it, there is an energy barrier for electrons to travel from one reservoir to the other. When a SAW moves through the channel, an electrostatic potential wave moves with it. If the amplitude of the SAW is large enough, each period creates a small well that can carry one electron over the energy barrier. For a SAW wavelength of about 1 μm, these wells are small enough that the repulsion between electrons ensures that only one electron can occupy each well. The current induced through the channel by a SAW of frequency f is then $I = ef$. Experiments on SET-SAW devices have shown that the current agrees with the expected value with a relative uncertainty of about 10^{-4}–10^{-2} [31, 32]. The speed limit for SET-SAW devices is not precisely known, but is thought to be about 10 GHz or higher.
(2) *RF-SETT for passive electron counting.* The devices described previously in this section are designed to *generate* an accurate current, but it may also be possible to simply *observe* individual charges as they flow through a device. A promising system for such a scheme is a long array of tunnel junctions in which charge flows in the form of solitons with regular spacing [33]. An electrometer can

detect the passage of each soliton if it is fast enough and has low enough noise. The best speed and noise performance has been achieved by integrating an SETT into a radio-frequency resonant circuit and monitoring the damping of this circuit [34]. This device, called an RF-SETT, is the most promising way to create an accurate current based on passive electron counting.

(3) *R-SET pump to allow fewer junctions*. The R-SET pump differs from the conventional SET pump in figure 1(C) by the addition of resistors at one or both ends of the chain of junctions. The purpose of these resistors is to suppress a certain class of unwanted tunnelling processes, known as cotunnelling events, because they involve simultaneous and coherent tunnelling of electrons at two or more junctions. It is expected that the resistors will allow the R-SET pump to perform as well as the conventional pump with fewer than seven junctions; thus, it should have an advantage in slightly higher speed (slightly higher value of current), and in simplicity of the driving electronics (at the expense of a more complex fabrication process). Experimental tests of the R-SET pump have recently begun [35].

4. Conclusions

We have reviewed our progress in the ECCS, and have shown that we can pump electrons onto the plate of a capacitor, and thus form a representation of the farad using the defining relation for capacitance $Q = CV$. We hope that, within the next 2 years, we will demonstrate a useful representation at an uncertainty of about 10^{-7}. Furthermore, if we can improve this to 10^{-8}, we can also close the quantum metrology triangle, without needing a large-value current standard.

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