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LONG-TERM CHARGE OFFSET NOISE IN COULOMB-BLOCKADE DEVICES

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While Coulomb-Blockade devices have shown promise as qubit systems or qubit state detectors, large scale integration of these devices is severely hampered due to the low-frequency charge offset (Q_0) drift in time. We note that an extrapolation of the 1/f noise to lower frequencies cannot account for the large ($\delta Q_0 \sim 1$ e) changes. We have investigated possible approaches towards reducing the Q_0 noise in $Al/Al_2O_{3-x}/Al$ based Coulomb-Blockade (CB) devices to a manageable level. In $Al/Al_2O_{3-x}/Al$-based CB devices, the maximum stability was typically 1-3 days, nearly independent of processing techniques or annealing treatments. In our best result, $\delta Q_0 < 0.1$ e (except for a single excursion) for 12 days. Encasing devices in a continuous metal vault (a nano-Faraday cage) did not however improve the Q_0 noise. We also discuss one specific identified cause of the transient relaxation of the drift. By extending conventional, two-level system theory (previously used to explain the behavior of macroscopic disordered materials), the phenomenon is shown to be caused by atomic structural relaxations of nearby insulating regions.

1 Introduction

The variety of Coulomb-Blockade (CB) based phenomena observed over the past 15 years rely on the generally accepted postulate that similar quasiparticles (e.g. electrons in a normal metal or Cooper Pairs in a superconductor) have nearly the same electronic charge e^*. Therefore the electrostatic energy U of a quasiparticle island is similarly quantized, $U \sim e^{*2}/2C$, where C is the sum capacitance of the system. While usually negligible, the energy can dominate the dynamics of a device at low enough temperatures if the quasiparticle island weakly couples to the surrounding environment[1].

By exploiting the CB phenomenon, a number of potentially useful devices have been or are being developed such as a Single Electron Tunneling Transistor (SETT)[1] an electron Pump[2] and a CB based single photon source[3]. In addition, CB devices may also factor strongly in a solid state-implemented quantum computer -- assuming one could be built -- either directly as a qubit[4] or indirectly as a qubit state detector[5].

At present the number of useful CB devices has remained relatively low, partly because these devices are strongly affected by nanometer length scaled movements of small packets of charge in insulators located either near or in the devices, a phenomenon called the Charge-Offset (Q_0) noise. First investigated soon after the initial CB confirmation, the power spectral density of the noise is typically found to be approximately inversely proportional to frequency[6] between $\sim 10^{-1}$ Hz and 10^3 Hz, similar to the noise observed in other small scale electronic devices. The Q_0 noise deleteriously affects a CB device in several ways. At relatively high frequencies ($> 10^{-1}$ Hz) the Q_0 noise limits the sensitivity of CB based charge electrometer, and thus the sensitivity of recently proposed detector for single nuclear spin qubit systems. In addition, recent evidence strongly suggests that the Q_0 noise at very high frequencies ($\sim 10^9$ Hz) is chiefly responsible for decoherence in recently demonstrated Single Cooper Pair Box qubits[7].

In addition to 1/f noise, there is also a long-term drift in which Q_0 can change by ~ 1 e*; this cannot be attributed to the low-frequency extrapolation of the 1/f noise. Indeed, a simple estimate shows that one would need to wait $10^{3,000,000}$ years for $1/f$ charge noise with typical amplitude to yield $\delta Q \sim 1e^*$. The long-term drift means that, over a sufficiently long time scale (typically 1-3 days), the device switches between the ON state (e.g. maximum current in a SETT) and the OFF state. While the low frequency Q_0 noise of

individual CB devices can be compensated, it becomes increasing difficult to implement this partial solution as the system size increases because the number of compensation circuits is proportional to the system size and the time between compensation circuit readjustment is inversely proportional to system size. Therefore the low frequency ($< 10^{-3}$ Hz) Q_0 drift prevents large scale device integration.

Interestingly, while the low frequency Q_0 drift presents such a serious problem, it has received far less attention than its higher frequency counterpart, chiefly because practitioners have generally assumed that the *1/f* spectra, a ubiquitous feature above ~ 0.1 Hz, extends down to $f = 0$. Thus conventional wisdom has until now dictated that the 10 Hz noise is sufficient to characterize the noise over *all* frequency regimes.

We have undertaken an investigation of the charge offset noise over a broad frequency range ($\sim 10^{-5}$ Hz to 10^2 Hz) in Al/Al$_2$O$_{3-x}$/Al-based CB devices. Our effort has focused on reducing the Q_0 noise and concurrently understanding what is driving the phenomenon. In Al/AlO$_x$/Al based CB devices we were unable to significantly reduce the low frequency Q_0 noise using a variety of different approaches. Therefore it seems that Al/Al$_2$O$_{3-x}$/Al-based CB devices will likely not be sufficiently stable to permit large scale CB device integration. We have however recently found that a new class of silicon based single electron tunneling devices are substantially more stable, thus pointing the way towards large scale integration of at least a subset of CB devices.

In addition, we have discovered that the low frequency ($< 10^{-3}$ Hz) charge offset noise often behaves quite differently than its higher frequency counterpart because it is driven by a different mechanism. Thus a measurement of the 10 Hz noise in an Al/Al$_2$O$_{3-x}$/Al based CB device may not reliably determine whether large scale integration of similarly fabricated CB devices is feasible.

Table 1. Attempts to reduce the low-frequency Q_0 noise.

- Thermal vs. e-beam evaporation
- Annealing in Forming gas
- Annealing in inert gas
- Using Ozone to oxidize tunnel junction insulators
- Thermal cycling (4-300 K)
- Nano-Faraday cage with O_2
- Nano-Faraday cage with O_3
- Cooling device with or without electrical short protection

2   Attempts to Reduce Charge Offset (Q_0) Noise & Drift

Until recently Al/Al$_2$O$_{3-x}$/Al-based CB devices were investigated principally because this material system has already demonstrated its utility in a variety of different applications. To fabricate the devices a standard double angle deposition process was utilized[8]. Early in the investigation, a thermal evaporator was used to deposit the Al films ($P_{base} \sim 2$ x 10^{-6} Torr; $P_{depo} \sim 2$ x 10^{-5} Torr; rate ~ 2.0 nm/sec). We later switched to an e-beam evaporator ($P_{base} \sim 1$ x 10^{-7} Torr; $P_{depo} \sim 5$ x 10^{-7} Torr; rate ~ 0.2 nm/sec) but were unable to detect any difference in the charge offset noise level between the two methods.

While imperfections in the insulator almost certainly cause the Q_0 noise, its exact location (and moreover its microscopic form) remains somewhat controversial[9-13]. One faction maintains that the noise must originate in the tunnel junctions because the electric fields are much more intense. Another faction maintains that the noise originates outside the tunnel junctions because the volume of insulating material and its associated imperfection density is likely much larger. Experimental evidence has implicated both sources, possibly owing to the different techniques used to fabricate the devices.

Our early work did not attempt to isolate the location of the noise, and instead attempted to improve all insulators near or in the CB devices. We first operated under the assumption that the Q_0 noise in our $Al/Al_2O_{3-x}/Al$-based CB devices was driven by charge traps which were similar in nature to charge traps SiO_2. Borrowing a technique for charge trap passivation[14] in SiO_2, we annealed the SETTs in Forming gas for 30 min at 350 °C. Unfortunately however, annealing in a reducing environment did not significantly improve the low frequency Q_0 noise. We also considered the possibility that the Q_0 noise was caused by oxygen deficiencies at the Al/Al_2O_{3-x} interface. In the magnetic tunnel junctions this problem is particularly deleterious, as unoxidized Aluminum reduces the magnetoresistance[15]. To improve the interface, magnetic tunnel junctions can be annealed in an inert environment[15]. While the $Al/Al_2O_{3-x}/Al$ tunnel junctions contain only a single metallic element (unlike magnetic tunnel junctions), we suspected that annealing in an inert environment might also sharpen the interface, possibly because the annealing temperatures are well below the aluminum/oxygen phase separation temperature but high enough to permit significant atomic motion. We therefore annealed the devices in a relatively inert environment (nitrogen), also at 350 °C for 30 min. Again however, the low frequency charge offset noise did not markedly change. Recent evidence has suggested that by switching to from O_2 to an O_2/O_3 mixture, the interface state trap density in Silicon can be reduced[16] and the quality of large area Niobium Josephson junctions similarly improved[17]. We thus attempted to improve the small area tunnel junctions in the CB devices by switching from pure O_2 to (5% O_3)/O_2 but were again unsuccessful at reducing the charge offset noise. Finally we considered the possibility that the charge offset noise arose from mechanical stresses occurring, presumably, from thermal contraction mismatches between the various materials in or near the device. However thermally cycling the devices between 4 K and 300 K did not affect the long term charge offset noise.

More recently we screened noise sources more than ~ 100 nm away by, for the first time, encasing SETTs inside 'nano-Faraday' cages. In these structures, the cage consists of two Aluminum thin film plates which sandwich the SETT. While the sides of the nano-Faraday cage are open, the SETT is still well-shielded from outside noise because the gap between the plates is an order of magnitude smaller than the distance between the SETT and the nearest plate edge. Because $Al/Al_2O_{3-x}/Al$ based CB devices generally cannot tolerate post-fabrication processing, the entire structure was fabricated with a single lithographic mask using an extension of the conventional angled deposition technique.
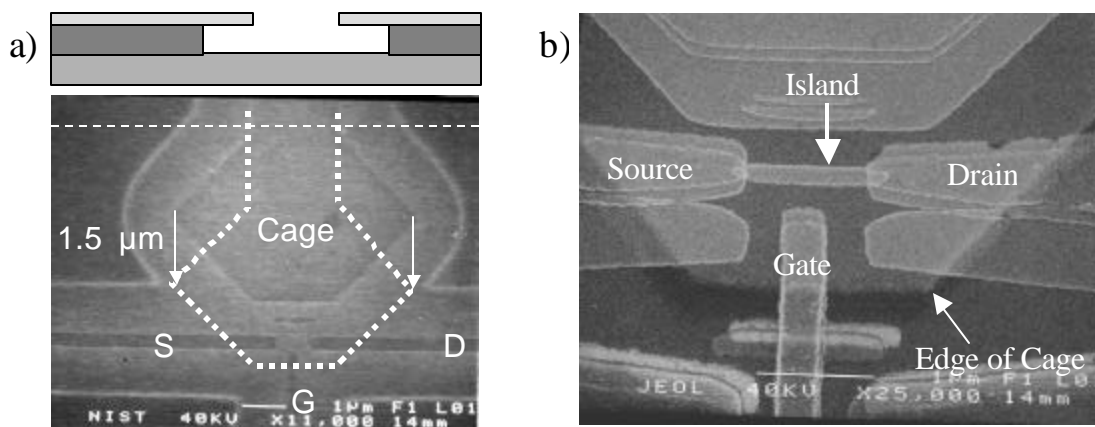

Figure 1. a) SEM Micrograph of lithographic mask used to fabricate nano-Faraday cage. A cross-section of the mask through the horizontal dashed line is shown above. The large hexagonal opening at the top of the mask is used to fabricate the two mask plates. b) Micrograph of SET transistor encased in nano-Faraday cage. Fabrication was stopped prior to depositing the top layers of the cage to permit viewing.

Figure 1a shows a micrograph of the lithographic mask. By depositing Aluminum at a shallow angle the bottom plate of the cage is placed underneath the SETT device area. Al_2O_{3-x} is then e-beam evaporated from an Al_2O_3 charge in the presence of $\sim 1 \times 10^{-5}$ Torr O_2 to electrically isolate the cage from the SETT. The SETT is then fabricated using double angled Aluminum evaporations at near normal incidence. To cap the device, Al_2O_{3-x} is again deposited before depositing the top layer of the cage.

Because this technique requires a large mask undercut to insure that the bottom of the cage lies comfortably underneath the SETT and because ~ 10 depositions are required to fabricate these structures, film stress and/or heating caused the masks to severely warp as shown in Fig. 2b. To stiffen the mask and provide the additional heat sinking, a 40 nm SiO layer was inserted between the bottom PMGI layer and the top PMMA layer. Additionally, a 20 nm Au layer was deposited prior to fabricating the cage bottom to decrease adhesion between deposited material and the mask. Combining these two techniques, mask warping was considerably reduced (Fig. 2c). Absent the cage top to permit examination, a SEM micrograph of a nano-Faraday caged SETT is shown in Fig. 1b.
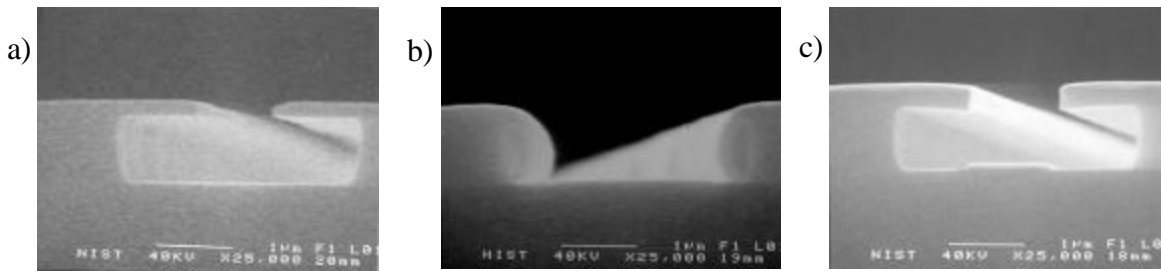

Figure 2. a) Cross-section of large undercut lithographic mask before deposition. b) Severe warping of conventional PMMA/PMGI bi-layer mask after depositing ~50 nm Al.. c) Decreased warping of PMMA/SiO/PMGI tri-layer mask after depositing ~ 20 nm Au/50 nm Al..

As anticipated, the nano-Faraday caged SETTs are far more robust than their uncaged counterparts. While conventional $Al/Al_2O_{3-x}/Al$-based CB devices are extremely sensitive to static electricity, the caged SETTs could be examined for several minutes using a SEM operating at 40 kV without destroying the tunnel junctions. In addition, caged devices could be left in ordinary tap water (which rapidly destroys uncaged devices) for over 24 hours affecting the tunnel junction resistance. Thus encasement of $Al/Al_2O_{3-x}/Al$-based CB devices should likely permit limited post-fabrication processing. Unfortunately however, both the short and long term Q_0 noise in the caged devices was approximately a factor of two higher. The additional noise was caused by defects in the deposited Al_2O_{3-x} film used to electrically isolate the SETT from the cage. Attempts to reduce the noise, including changing the background O_2 pressure, switching from O_2 to (5% O_3)/O_2 and varying the deposition rate did not unfortunately affect the charge offset noise level. Because the e-beam deposited Al_2O_{3-x} films are robust noise sources however, they proved to be remarkably useful towards determining the origin of a component of the charge offset noise.

3 Understanding the Charge Offset (Q_0) Noise Origin

While evaluating our attempts to reduce the charge offset noise in $Al/Al_2O_{3-x}/Al$ based CB devices, we observed that the low frequency ($< 10^{-3}$ Hz) Q_0 noise tended to decrease strongly over time. To parameterize the behavior, we measured the periodic transconductance of a SETT over extended periods of

time to determine the phase shift and thus the charge-offset (Q_0). In Fig. 3a, Q_0 vs. time is shown for a conventional Al/Al$_2$O$_{3-x}$/Al based SETT at T ~ 30 mK. A drastic change of the Q_0 stability as a function of time is clearly evident. Unfortunately though, we were unable to consistently, unambiguously observe the effect using conventional double angle deposited SETTs. To controllably introduce noise, Al$_2$O$_{3-x}$ was deposited at well-defined locations relative to the Al/Al$_2$O$_{3-x}$/Al SETT using an extension of the nano-Faraday caged fabrication technique.

We can quantify the time dependence of the characteristic switching rate by determining the characteristic time $\mathbf{\Gamma}_Q^{-1}$, before the charge offset deviates outside a predetermined window size (e.g. $\delta Q_0 = 0.1$ e). Introducing controlled noise sources permitted us to determine $\mathbf{\Gamma}_Q^{-1}$ as arising from a specific source.
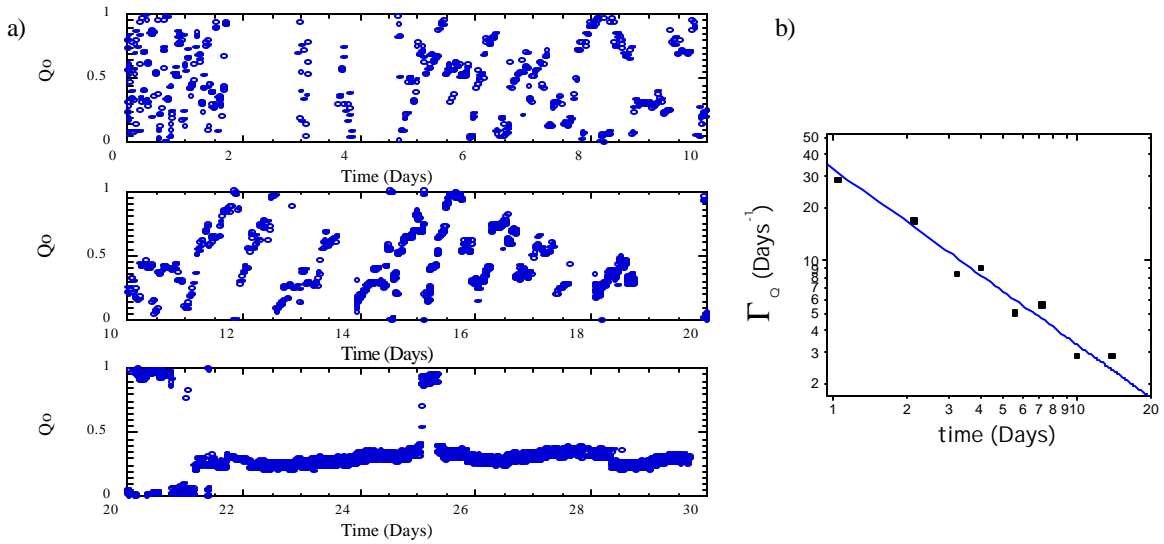

Figure 3. a) Dependence of the charge-offset (Q_0) over time in a SETT without a deliberate noise source. b) Characteristic switching rate Γ_Q as a function of time since fabrication for a SETT with a deliberate noise source. This response was also observed in a fraction of the devices without deliberate noise sources, such as in Fig. 3a.

In Fig. 3b the switching rate $\mathbf{\Gamma}_Q$ is shown as a function of time since fabrication for a SETT with a deliberate noise source. For the first several weeks, *the characteristic switching rate is inversely proportional to time since fabrication.* The inverse time dependence was observed in all Al/Al$_2$O$_{3-x}$/Al based SETTs with deliberate noise sources and in most SETTs without deliberate noise sources, albeit usually less pronounced.

The Q_0 transient response was recently attributed to the same mechanism responsible for anomalous heat evolution in disordered materials[18]. For almost thirty years it has been recognized that disordered materials contain large numbers of two-level systems (TLS) which in general are broadly distributed in energy and relaxation time[19]. At temperatures high compared to the maximum TLS energy splitting $\mathbf{D}$, the two states of the TLS are statistically equally populated and thus do not contribute appreciably to the specific heat. When a disordered material is quenched from high temperatures however, TLS with $\mathbf{D} > k_B T$ relax, releasing packets of heat which can be calorimetrically detected. Experiments performed over the past twenty years have demonstrated that in a broad range of materials with varying degrees of disorder, *the heat evolution is inversely proportional to time since quenching[19].*

By extending conventional TLS theory, we are able to explain the transient response of the charge offset noise. It seems likely that the insulators surrounding the SET device contain large number of TLS

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(whose microscopic origin is not critical for the model). Each time a TLS relaxes, it emits a packet of energy. This process can change Q_0 in at least two ways. First, the atoms constituting the TLS, which likely move during the relaxation, may change their charge configuration. The charge reconfiguration will then induce a charge change on the central island which is then changes the source drain current of the SETT. Second, the packet of energy from the relaxing TLS, likely in the from of a phonon, may travel some distance before being absorbed in another insulating region. The energy may then cause the insulator to rearrange which again would be detected using the SETT. Inserting rough estimates of the TLS density and characteristic volume, we have obtained relatively good agreement between theory and experiment.

At sufficiently long times, the $Al/Al_2O_{3-x}/Al$-based CB devices cease to significantly improve with age. For the vast majority of these devices, a Q_0 change larger than ~ 0.1 e is typically observed every 1-3 days. In a single, non-repeatable instance, partially shown in Fig. 3a, *Q_0 remained stable, except for a single excursion, for twelve days* - our best result. The $Al/Al_2O_{3-x}/Al$ Q_0 baseline noise is a surprisingly robust value: it is insensitive even to high temperature anneals and processing modifications. Thus regardless of the baseline noise source, $Al/Al_2O_{3-x}/Al$ based tunnel junction systems will probably not be sufficiently stable for even moderate scale CB device integration.

Recent results have shown however that Silicon based SETTs fabricated using a pattern dependent oxidation process have an exceptionally small low frequency Q_0 noise[20]. One device drifted by δQ_0 ~ 0.005 e over several days. Further research on the mechanism responsible for the remarkably low charge offset noise is being pursued.

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