Single-Electron Charging Effects in One-Dimensional Arrays of Ultrasmall Tunnel Junctions

L. S. Kuzmin,[a] P. Delsing, and T. Claeson
Physics Department, Chalmers University of Technology, S-412 96 Göteborg, Sweden

K. K. Likharev
Department of Physics, Moscow State University, 119899 GSP, Moscow, U.S.S.R.
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Coulomb blockade of single-electron tunneling and high sensitivity to an external electric field has been observed for 1D series arrays of ultrasmall-area ($<0.1\times0.1\ \mu m^2$) tunnel junctions, made of $Al/Al_xO_y/Al$, at helium temperatures. In particular, the dc voltage V across the array responds strongly to a voltage U_g applied to a control electrode. For an array of thirteen junctions, the voltage gain $K_V=|\delta V/\delta U_g|$ of the resulting sub-single-electron transistor reached 0.2, while the charge sensitivity of the device was as high as $2\times10^{-4}\ e/Hz^{1/2}$ at a frequency $f=10$ Hz.

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Today, with modern submicron fabrication technologies, it is possible to make tunnel junctions with capacitances small enough to observe new effects connected to the tunneling of single electrons and Cooper pairs (for reviews, see Refs. 1 and 2). These effects arise when the elementary charging energy $e^2/2C$ of the junction capacitance C becomes larger than the energy k_BT of the masking thermal fluctuations and the tunnel resistance R is larger than the quantum unit $R_Q=h/4e^2\approx6.5$ kΩ to avoid smearing by quantum fluctuations ($h/\tau\le e^2/2C$, where $\tau\approx RC$).

Under these conditions, tunneling of a single electron (or a single Cooper pair in a Josephson tunnel junction) results in a noticeable recharging of the junction capacitance, such that the probability of tunneling of the next electron (pair) is drastically affected.

Outside this so-called Coulomb blockade range, i.e., at nonvanishing dc currents for $|V|>V_t$, several new effects have been predicted depending upon the detailed configuration. Here, we report experimental results on the Coulomb blockade in 1D arrays of very small tunnel junctions and how it is influenced by external fields. The response is larger than reported previously for pairs of junctions and gives promise of electrometers and transistors controlled by sub-single-electron charge changes.

The simplest single-electron charging effects, including the Coulomb blockade and the dc voltage offset at large V, were observed in granular structures consisting of many tunnel junctions with random parameters.[3,4] However, more subtle and interesting phenomena like periodic oscillations in the dc I-V curves and a large sensitivity to external electric fields have been clearly observed only recently in experiments with single pairs of electrostatistically coupled tunnel junctions.[5-8] Experimental confirmation of other predictions, including quantized (equidistant) dc voltage steps due to phase locking to an external rf field, has not yet been unquestionably observed.[9,10] Hence there is a need of further experiment.

Theory[2,11] says that a very promising structure for experimental observation of virtually all charging effects is a one-dimensional array of a large number, $N\gg1$, of small tunnel junctions connected in series [Fig. 1(a)]. Here, the charging effects lead to the formation of single-electron (or Cooper-pair) solitons behaving qualitatively like the well-known sine-Gordon solitons. The Coulomb repulsion of the solitons and their interaction with external electric fields should result in the formation

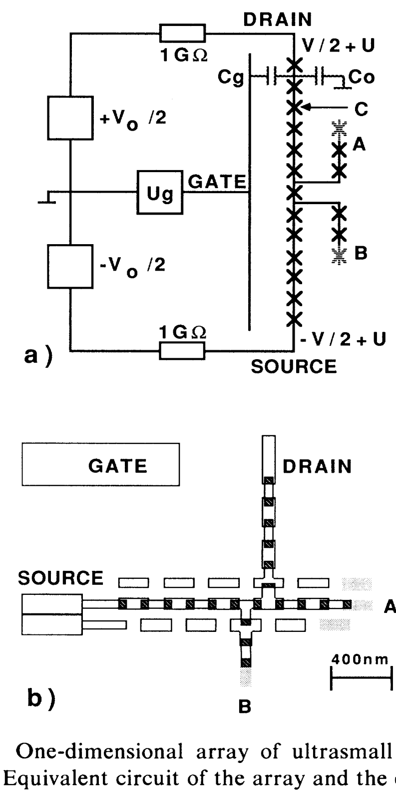

FIG. 1. One-dimensional array of ultrasmall tunnel junctions. (a) Equivalent circuit of the array and the dc supply circuit; (b) scheme of the array geometry. The additional metal islands disconnected from the array are formed as a side effect of the Dolan technology and do not affect the array properties.

of quasiperiodic trains of solitons, either pinned to particular "sites" (metallic electrodes) at $|V| < V_t$ or moving continuously along the array $|V| > V_t$.

The present work has used 1D arrays of $Al/Al_xO_y/Al$ tunnel junctions with areas less than 0.01 μm^2. Figures 1(a) and 1(b) show a typical configuration of the array; additional branches A and B have been used to measure the dc voltage of a single junction inside the array. A comparison of the behaviors of a single junction and one junction inside an array will be published separately.

The structures were fabricated on unoxidized silicon substrates in a single vacuum cycle using the shadow evaporation technique developed by Dolan.[12] The hanging bridge masks were patterned in a two-layer (polymethylmethacrylate/copolymer) resist by e-beam lithography. Junction sizes were estimated to be less than 0.1×0.1 μm^2. The tunnel barriers were grown by thermal oxidation of the base aluminum electrodes ($\sim$30 nm thick) in oxygen at a pressure of 0.05 mbar during 10–20 min. This gave a tunnel resistance of a single junction of order 10^6 Ω ($\gg R_Q$). A typical thickness of the counter electrode was 50 nm.

Several arrays, containing between 2 and 41 junctions each, were studies at temperatures ranging between 1.2 and 4.2 K. A few junctions were also measured below 1 K in order to confirm the good quality of their tunnel barriers. As Al became superconducting, $T_c \approx 1.22$ K, an energy gap opened up, giving a small quasiparticle current in the sub-gap-voltage region.

Figure 1(a) shows the dc supply circuit used to record the I-V curves of the arrays. The circuit was symmetric such that its midpoint was connected to the metal substrate holder which, in turn, contacted the silicon substrate. An independent dc bias voltage U_g could be applied to the special "gate" electrode [Figs. 1(a) and 1(b)]. We could also irradiate the array of microwaves via a coaxial cable and an antenna loop close to the substrate.

Figure 2 shows a dc I-V curve of an array containing $N = 13$ junctions. Note that the irregular shape is not due to noise. It is reproducible but it is dependent upon the direction of the current sweep. The curve, which was registered for increasing bias, has to be rotated 180° to resemble the decreasing-bias curve. One can readily see that it oscillates between two linear "extremal" curves. The outer of these two curves hits the voltage axis at the point $(V_t)_{max} \approx 1.8$ mV [the inner extremal curve is somewhat less well defined, but crosses the voltage axis at $(V_t)_{min} \approx 1.4$ mV]. At large $|V|$, the asymptotic I-V curves are offset by $2V_{off} \approx 3.4$ mV. All these features could be observed at higher temperatures ($T \approx 4$ K) as well; however, the quasiperiodic oscillations became considerably more pronounced at lower temperatures.

The simple theory[11] predicts that for a fixed average electric potential U of the array, its I-V curve should exhibit the Coulomb blockade of tunneling ($I \rightarrow 0$ for $T \rightarrow 0$ and $|V| < V_t$) with the maximum threshold

$$(V_t)_{max} = V_t(U=0, \pm e/C_0, \pm 2e/C_0, \ldots) \approx e/(CC_0)^{1/2}, \quad (1)$$

where C is the capacitance of a single junction and C_0 is the capacitance between each single metallic electrode of the array and the environment ("ground plane"). The factor $(CC_0)^{1/2}$ arises because the typical size of the single-electron soliton (expressed in the number of junctions) equals $\sim 2\lambda_c$, where[13] $\lambda_c = (C/C_0)^{1/2}$ for $C_0 \ll C$, and only the electrodes inside the soliton range yield a contribution to the Coulomb blockade effect. Beyond the threshold ($|V| > V_t$) the I-V curves should gradually (and *without oscillations*) approach their linear asymptotes $V = IR_\Sigma + V_{off}$; the offset is given by the value[14]

$$V_{off} = (N - \lambda_c)e/2C, \text{ for } N/\lambda_c > 2. \quad (2)$$

Using the observed values of $(V_t)_{max}$ and V_{off} and Eqs. (1) and (2), we obtain $C \approx 4 \times 10^{-16}$ F and $C_0 \approx 2 \times 10^{-17}$ F. These values compare favorably with the numbers we get using the area and the expected specific capacitance[15] to calculate C and elementary formulas to estimate C_0.

The oscillations of the dc I-V curves between the two extremal curves can be readily explained as well. The I-V curves should be very sensitive to extra charges Q on

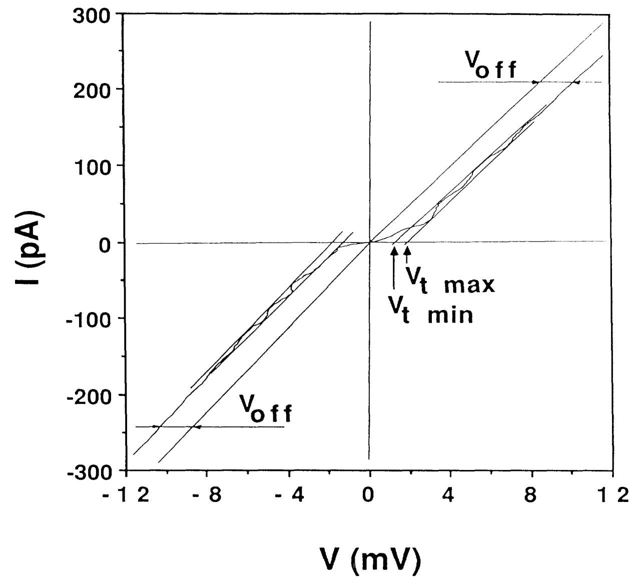

FIG. 2. A typical dc I-V curve of an array of $N = 13$ $Al/Al_xO_y/Al$ junctions, each with area 0.09×0.09 μm^2. $T = 1.65$ K. Thin straight lines shows extremal curves between which the voltage oscillates as the current is increased, and serve only as guides for the eye.

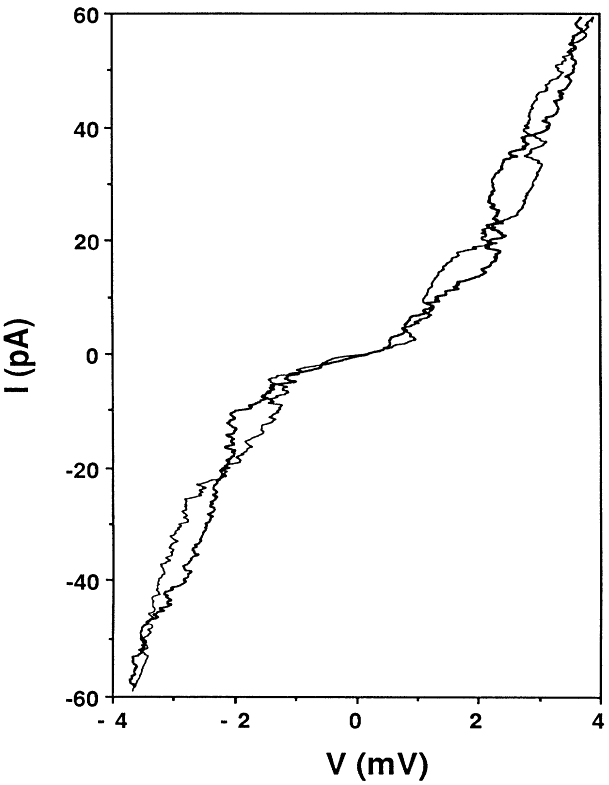

FIG. 3. Effect of the "gate" voltage U_g on the dc I-V curve of the same array as shown in Fig. 2 ($T=1.35$ K). The thick and thin curves were plotted (with increasing bias voltage) for $U_g=0$ and 12 mV, respectively.

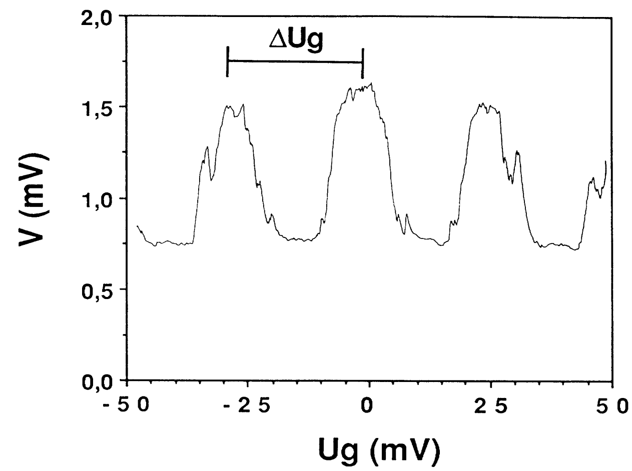

FIG. 4. dc voltage across the same array as a function of the gate voltage U_g for a fixed value of the dc current $I\approx 5$ pA ($T=1.34$ K).

the metallic electrodes, and hence to the average potential U of the array. They will oscillate as a function of U with the period[11] $\Delta Q=e$, i.e., $\Delta U=e/C_0$.

Now note that even a small asymmetry $\eta \ll 1$ of the 1-GΩ resistors used in the dc supply circuit [Fig. 1(a)] leads to a noticeable variation of U proportional to the array current: $U=\eta V_0 \approx \eta \times (2\ \text{G}\Omega)\times I$.

This effect results in oscillations of the I-V curve between the two extremal curves with maximum and minimum V_t; we have found that a value $\eta \approx 0.08$ is sufficient to explain the results shown in Fig. 2. A measurement using a dummy short instead of the array gave an unbalance voltage of about $0.04V_0$.

This interpretation gets additional support from experiments with the gate voltage U_g. A variation of U_g of $\Delta U_g/2$, where the period $\Delta U_g \approx 27$ mV, resulted in a change of the phase of the voltage oscillations of π (Fig. 3). Exactly this behavior (with $\Delta U_g=e/C_g$, where C_g is the capacitance between the gate and the single electrode) follows from the theory.[2] The value $C_g \approx 6 \times 10^{-18}$ F deduced from the experimental ΔU_g is in reasonable agreement with a crude estimate, 5×10^{-18} F, obtained from the structure geometry [Fig. 1(b)] and a dielectric constant of $\epsilon=12$ of the silicon substrate.

A spectacular demonstration of the periodic response to the gate voltage is shown in Fig. 4. The array voltage, at a fixed dc current, is given as the gate voltage is varied. The maximum derivative $K_V=|\delta V/\delta U_g|$ was roughly 0.2; this "voltage gain" of our structure considered as a "single-electron tunneling transistor" controlled by sub-single-electron electric charge variations is a factor of ~ 20 larger than that achieved earlier by Fulton and Dolan[6] in a similar structure with $N=2$. Taking into account our circuit [Fig. 1(a)] and theoretical results,[11] we can estimate the maximum K_V for the array: $(K_V)_{\max}=|\delta V/\delta U_g|_{\max}=2C_g/(C_0+C_g)$. Using the estimated values of C_0 and C_g, we obtain $(K_V)_{\max}\approx 0.5$, a bit higher than the measured K_V. The coupling between the gate and the array could be improved and we believe that values $K_V>1$ can be readily obtained for any $N\geq 2$ using configurations with an overlapped gate electrode.

We have also made a rough estimate of the low-frequency noise level of our "transistor." The rms noise output voltage within 1 Hz bandwidth was measured. We found that the charge sensitivity $\delta Q=(\delta V_{\text{rms}}/K_V)C_g$ can be as high as $2\times 10^{-4}\ e/\text{Hz}^{1/2}$ at a frequency $f=10$ Hz, where the $1/f$ noise is still dominant. This result gives indirect support to the theoretical prediction of the white-noise-range sensitivity $\delta Q\approx 10^{-5}\ e/\text{Hz}^{1/2}$ (Ref. 2).

The array response to external microwave fields of frequencies between 0.5 and 5 GHz is a very complex function of the dc bias point and the field amplitude. The results will be reported later. No quantized voltage steps[2] were observed. Numerical simulations of the array dynamics[11] with the parameters listed above have shown that one would not expect to detect the steps in our experiments: They should be well pronounced only at temperatures below ~0.3 K.

In conclusion, we have observed several peculiar features of the dc I-V curves of one-dimensional arrays of ultrasmall tunnel junctions formed between normal-metal electrodes. The basic features, including the Coulomb blockade of tunneling at lower voltages, the offset of the curves at larger voltages, and the extremely high sensitivity of the voltage across the array to external electrostatic fields, can be well explained using the existing theory[2,11] of the correlated single-electron tunneling, especially if nonuniformities in the array are taken into account. Some features, however, do not find such a ready explanation. They include a noticeable (~15%) difference of the asymptotic slopes of the I-V curve for positive and negative dc voltages, and some dependence of the amplitude and the period of the voltage oscillations upon the prehistory of the bias evolution (the oscillations are more pronounced when one sweeps V from the origin to larger values, see Fig. 2).

The latter effect may be qualitatively explained as a result of a slow drift of charged impurities embedded inside the tunnel barriers under the action of large dc voltages.[5] This drift may lead to random variations of the background values of Q and hence to a randomization of the voltage oscillations. At smaller V, the background charges apparently relax to similar equilibrium values $Q=ne$, so that the coherence of the voltage oscillations in various junctions is restored and their net amplitude is increased. Of course, the relaxation phenomena need more detailed studies.

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[a]On leave from Department of Physics, Moscow State University, Moscow, U.S.S.R.

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