Time-Correlated Single-Electron Tunneling in One-Dimensional Arrays of Ultrasmall Tunnel Junctions

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We have obtained convincing experimental evidence of the time correlation of single-electron tunneling (SET) events, i.e., of SET oscillations with average frequency f_s fundamentally related as $f_s = I/e$ to the dc current I. The experiments have been carried out with one-dimensional N-junction arrays of Al/oxide/Al tunnel junctions of area $S \approx 0.006\ \mu m^2$ at $T \approx 50$ mK. The correlation showed up as resonance peaks of the dynamic resistance dV/dI as a function of I, arising at $I = I_n = nef$ ($n = \pm 1$ and ± 2) for irradiation of the $N = 15$ and 19 samples by microwaves of frequency $f = 0.7$–5 GHz.

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As the area of a tunnel junction is decreased, together with its capacitance and tunnel conductance, single-electron charging effects become increasingly important. For sufficiently small C and G ($C \ll e^2/k_BT$, $G \ll R_Q^{-1} = 4e^2/h$) the charging can lead to a considerable correlation of tunneling events either in space, or in time, or in both.[1,2]

The former type of correlation has been observed reliably in recent experiments[3-7] with double tunnel junctions (i.e., $M/I/M/I/M$ structures) of submicron area. Experimental evidence of the latter (time) correlation, obtained in experiments with disordered and irreproducible granular structures,[8,9] was, on the contrary, much more questionable.[2] A reliable observation of this effect represents an important task. In fact, a complete time correlation of the single-electron tunneling (SET) events in a structure means a periodic (rather than stochastic) transfer of electrons, with a frequency f_s fundamentally related as $f_s = I/e$ to the dc current I transferred through this structure. This phenomenon is (qualitatively) dual to the ac Josephson effect, and can become quite important for both fundamental physics studies and several applications in what has been nicknamed "single-electronics."[1,2]

According to the theory,[1,2] the time correlation should be observable in single tunnel junctions (i.e., $M/I/M$ structures) biased by a fixed current. Several attempts[10-12] to observe this effect (and/or the similar correlation of the Cooper-pair tunneling events[2,13]) have, however, failed or at best given indirect evidence.[14] This is presumably due to a parasitic effect of the stray capacitance $C_0 \gg C$ between the junction leads.[15] It ruins the constant current bias and, as a consequence, the tunneling-event correlation.

A possible way[1,2,16] to circumvent this problem is to use one-dimensional arrays of $N \gg 1$ junctions connected in series, i.e., $M/I/M \ldots M/I/M$ structures. In this case, the stray capacitance C_0 is effectively detached from each junction by its $N-1$ neighbors, and both space and time correlation of tunneling is possible.[16] Our first experiments[15,17] with such arrays confirmed the reliability of the theoretical predictions, but did not reveal the time correlation, presumably because of a relatively high temperature in those experiments. The purpose of this paper is to report the first reliable observation of the time correlation. It has become possible by using millikelvin refrigeration.

The methods of the array fabrication were similar to those described by us elsewhere.[15,17] Five independent arrays with N from 15 to 21 junctions each were fabricated on Si substrates. The junction area was 70×90 nm^2 and the period 100 nm. The individual junction resistance was ~ 200 kΩ. A comparison of the array resistances indicated that the spread of the junction parameters did not exceed 10%–20%.

dc I-V curves and dynamic resistance $R_d = dV/dI$ could be measured with a well screened and filtered symmetric circuit and lock-in techniques, at temperatures from 4 to 0.05 K using a dilution refrigerator. Microwaves could be fed to the arrays via a coaxial cable, a cooled 10-dB attenuator and a three-turn coil. The coupling was very poor, ~ -60 dB at best. This was satisfactory only at certain resonance frequencies; at other frequencies the microwave power needed to affect the junction I-V curve heated up the sample holder noticeably. The poor coupling also eliminated any external or room-temperature rf noise coming down the coaxial cable.

Figures 1(a) and 2(a) show data obtained for a nineteen-junction array (almost similar results have been obtained for a fifteen-junction array on the same substrate). As the temperature T is lowered, the dc I-V curve becomes nonlinear, with a drastic suppression of the conductance below some threshold value V_t behind which solitons are formed.[16] The nonlinearity is present even if T is larger than the superconducting transition temperature $T_c \approx 1.2$ K of the aluminum electrodes. $e/2C$ is comparable to $2\Delta/e$ and the superconducting gap

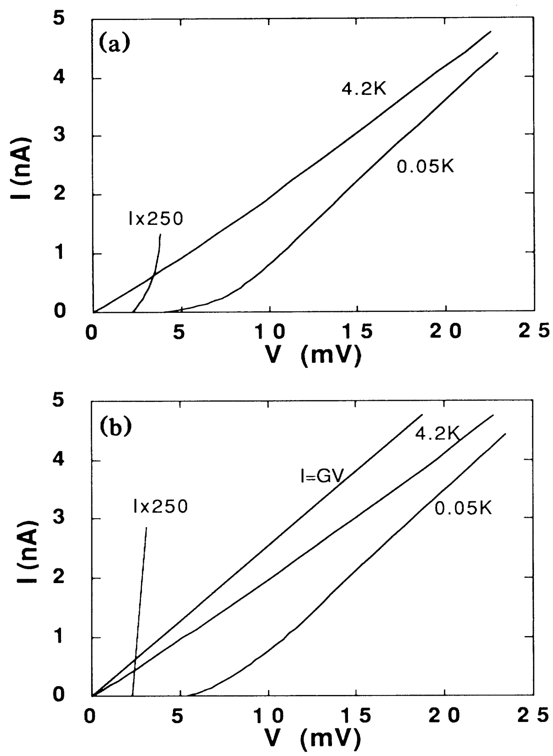

FIG. 1. dc I-V curves of a nineteen-junction array of submicron tunnel junctions for two values of temperature: (a) experiment; (b) calculations according to the theory (Ref. 15) (see text for parameter values). A blowup of the current scale for the 50-mK curve shows that the current is practically zero at low bias.

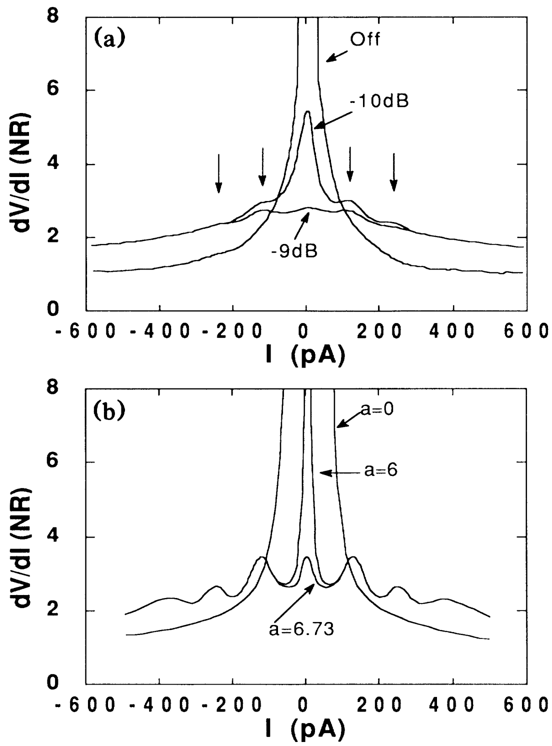

FIG. 2. The dynamic resistance $R_d = dV/dI$ as a function of dc current I for the same array as in Fig. 1 at 50 mK for three values of the microwave power ($f = 0.75$ GHz): (a) experiment; (b) calculations. $a = A/(e/C)$ is the normalized amplitude of the microwave voltage applied to the array. The amplitude difference between the two pumped curves in (b) corresponds to 1 dB in power. R_d is normalized to the array resistance (NR_N). Arrows show nominal positions of the resistance peaks according to Eq. (1).

structure is smeared; in fact in this case it is visible only in the derivative. At millikelvin temperatures, the threshold is very well defined and the current below V_t is extremely low ($I < 50$ fA) [see Fig. 1(a)]. The resistance at zero bias is at least 100 GΩ, i.e., at least 4 orders of magnitude larger than at high bias. Note that V_t is definitely less than NV_Δ, where $V_\Delta = 2\Delta(T)/e \approx 0.35$ mV is the superconducting gap voltage.

Applied microwaves smear the current suppression range gradually, and eventually make the dc I-V curve linear. At power levels which make the zero-bias dynamic resistance of order of (although still higher than) the asymptotic resistance $R_N = 1/G$, the dc I-V curves exhibit periodic structure which can be seen as periodic peaks in the R_d vs I plots [Fig. 2(a)]. Most remarkable, the dc current positions of the peaks are not dependent (within the $\approx 15\%$ accuracy of their definition) on the power, but are linear functions of the microwave frequency f (Fig. 3),

$$I_n = nef, \text{ for } n = \pm 1, \pm 2, \dots, \quad (1)$$

at least within our experimental range (0.7–5 GHz). On the contrary, dc-voltage positions of the peaks do depend on the microwave power. An increase of the temperature broadens the peaks (and eventually smears them out). Below 0.1 K the peak structure does not change very much.

All our experimental data find a natural explanation within the framework of the "orthodox" theory of the correlated single-electron tunneling.[1,2,16] Figure 1(b) shows dc I-V curves of a uniform array (with identical junctions) as calculated from the theory[16] for two experimental values of T, the calculated value $C_0 = 1.2 \times 10^{-17}$ of the stray capacitance of the electrodes, and fitted values $R_N = 0.21$ MΩ per junction, $C = 2.4 \times 10^{-16}$ F, $G_L/G = 0.15$, and $2\Delta(0) = 0.38$ meV. Here G_L is the junction conductance below the superconducting gap in a single voltage-driven junction, i.e., a junction without charging effects. The resistance is obtained directly from the asymptotic slope of the experimental I-V curve, the capacitance is consistent with the junction area, while the energy gap and subgap conductances are consistent with the literature data for similar junctions. One can see that the agreement of the overall shapes of the I-V curves can be characterized as perfect. The calcu-

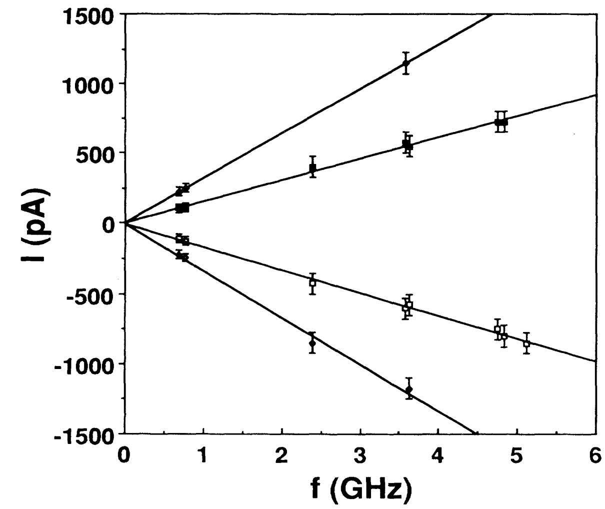

FIG. 3. dc current positions of the $n = \pm 1$ and $n = \pm 2$ resistance peaks for two arrays $N = 19$ and $N = 15$. The lines are least-squares fits to the data. They have slopes of 1.52×10^{-19} A/Hz, -1.63×10^{-19} A/Hz, 3.20×10^{-19} A/Hz, and -3.33×10^{-19} A/Hz, i.e., a good agreement with the values e and $2e$ given by the theory of time-correlated tunneling.

lated value 2.4 mV of the threshold voltage V_t is also close to the experimental value 2.0 mV; however, the exact shape of the low-current part just above V_t of the experimental I-V curve differs from that obtained in the calculations which used a crude assumption of constant G_L at $|V| < V_\Delta$ (all other results are not sensitive to this feature).

Note that according to theory the current suppression below V_t (the so-called Coulomb blockade of tunneling) is entirely due to charging effects and not related to the superconductivity of the electrodes. The superconductivity, however, does affect the general shape of the dc I-V curve at $T < T_c$, making the current relatively low for all voltages below $V_t + NV_\Delta \approx 8$ mV.

Figure 2(b) shows R_d vs I curves calculated with the same parameters as above for three values of the microwave amplitude. Theoretically, each resonance peak (1) is due to phase locking of the SET oscillations by the nth harmonic of the external microwave signal. In the ideal case ($T \to 0$, $C_0/C \to 0$, $GR_Q \to 0$, $N \to \infty$) the peaks would be infinitely high and would correspond to horizontal voltage steps in the I-V curve, similar to the Shapiro current steps in the Josephson effect. In reality, the peaks are broadened by quantum, thermal, and shot noise; in our case of relatively small N, the last contribution dominates. The similarity of the measured and calculated curves is obvious, but in the experiment the peaks have some additional broadening due to the ac signal used to record the derivative (≈ 5 pA rms) and possible noise reaching the junction.

In spite of the very reasonable agreement of the data with the theory of correlated single-electron tunneling, we should examine whether alternative explanations of the observed peaks are possible. First of all, qualitatively similar peaks (with even n) could arise due to the phase locking of Cooper-pair tunneling (the so-called Bloch oscillations[2,14]). In the absence of a quantitative theory of Bloch oscillations in junction arrays, one may argue that the peak with $n = 1$ could be due to phase locking of the second harmonic of these oscillations. Nevertheless, it seems hardly possible to explain in this way why the peak with $n = 1$ is the largest one (at higher frequencies of our range this peak alone was visible). Moreover, the Josephson coupling energy of our junctions can be estimated as 3×10^{-24} J at most, so that its ratio to the elementary charging energy $E_c = e^2/2C \approx 50 \times 10^{-24}$ J is less than $(GR_Q)^{1/2} \approx 0.15$. According to the theory of a *single* current-biased junction[2] (it should be qualitatively applicable to the long arrays as well[15]) the Bloch oscillations should be completely suppressed by the Zener tunneling in this case.

Somewhat naively one could ask whether the observed peaks could be either classical Shapiro steps (separated by the dc voltage intervals $\Delta V = hf/2e$) smoothed by noise or single-electron photon-assisted tunneling steps (with $\Delta V = hf/e$). These interpretations, however, should be excluded by the huge discrepancy between the theoretical and experimental values of ΔV (a few μV and several mV, respectively), as well as the mentioned fact of the microwave-power dependence of the voltage intervals.

In conclusion, we believe that our data and their analysis leave no doubt that the periodic peaks of the differential resistance as a function of dc current are due to the phase locking of the SET oscillations (with a noise-broadened spectral line) by the microwave signal. Thus the time correlation of the single-electron tunneling events has been observed conclusively.

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