Direct experimental observation of discrete correlated single-electron tunneling

L. S. Kuz'min and K. K. Likharev
M. V. Lomonosov State University, Moscow

(Submitted 6 March 1987)
Pis'ma Zh. Eksp. Teor. Fiz. **45**, No. 8, 389–390 (25 April 1987)

Periodic oscillations observed on the current-voltage characteristics of granular tunnel junctions can be described well by a theory of a correlated tunneling of single electrons. This effect can be utilized to develop an ultrasensitive (subelectron) electrometry.

The recent prediction[1] of coherent "single-electron" oscillations with a frequency $f = I/e$, which should be excited by a direct current I in a small-area tunnel junction at a low temperature, has rekindled interested in the effect that underlies these oscillations: a correlated discrete tunneling of single electrons. This effect has previously been observed[2,3] only indirectly, from its effect on the collective characteristics of systems of a large number of metal granules. In the present letter we report a direct observation of correlated tunneling in a system of two junctions formed by a single metal granule of submicron dimensions.

Figure 1 is a schematic sectional view of the thin-film structures, with an area of $20 \times 20\ \mu\text{m}^2$, which we studied. Indium granules with an average diameter on the order of 100 nm are formed between two solid films of lead alloys in such a way that they are separated from the lower film by an In_2O_3 tunnel barrier with a conductivity of about 10^6 S/cm. The granules are separated from the upper film by the same barrier and also by an insulating layer of SiO, about 50 nm thick, which completely covers most of the granules. Consequently, about half of the samples are essentially nonconducting (have a resistance $R \gtrsim 10^7\ \Omega$), while the others have a resistance $R \gtrsim 10^5\ \Omega$, which corresponds to conduction through a small number (1–10^2) of granules. At liquid-helium temperatures such samples exhibit the characteristic suppression of the tunnel current (i.e., a maximum of the derivative $R_d \equiv dV/dI$) in the range $|V| < V_\Sigma$, with a width $2V_\Sigma$ significantly greater than the value $2V_g \cong 2\Delta_{\text{Pb}}(T)/e$, associated with the superconductivity of solid films. Furthermore, as the current is increased, the $I(V)$ curves tend toward essentially linear asymptotes, which are shifted precisely $2V_t = 2V_\Sigma - 2V_g$ (Fig. 2a). This effect, which has been seen previously,[2] can be explained in a natural way[1–5] in terms of a "blockade" of the tunneling that arises because of electrostatic effects. In accordance with Ref. 2, the $I(V)$ and $R_d(V)$ curves for most of the samples are monotonic.

For a few of the samples, however, these curves exhibit large oscillations ($\Delta R_d / R_d$ up to $\pm 10\%$) with a period ΔV which remains strictly constant for a given sample and with an amplitude which falls off slowly with increasing V (Fig. 2a). At liquid-helium temperatures, this effect is completely reproducible. After the sample is heated to room temperature and then recooled, however, the oscillation phase fre-

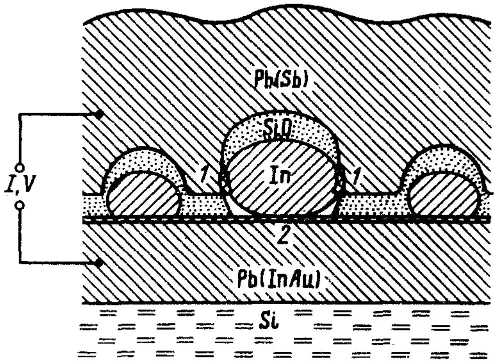

FIG. 1. Sectional diagram of the test structures.

quently shifts with respect to the origin (while the period ΔV is exactly conserved).

This is precisely the behavior that is predicted[4,5] by the theory of discrete single-electron tunneling in the case in which the conductivity of the system is dominated by tunneling across two junctions (1 and 2 in Fig. 1), which connect electrodes through a single granule. Each oscillation period corresponds to a change in the average charge of the granule by an amount e; the size of the period in the voltage is $\Delta V = 3/C_1$,

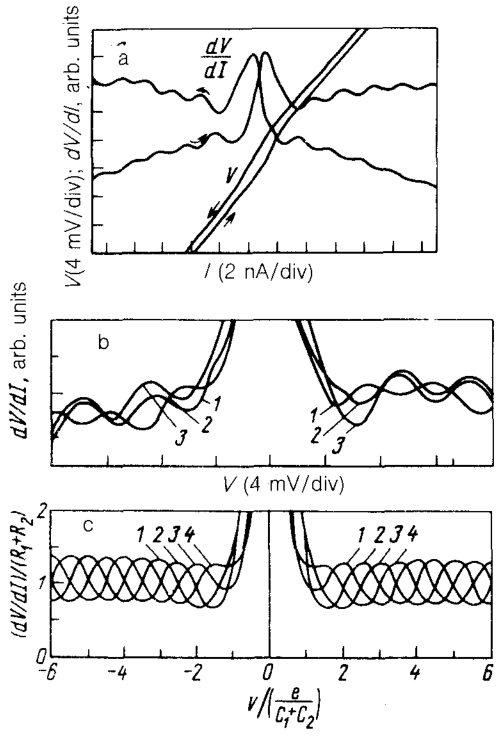

FIG. 2. The voltage V and the derivative dV/dI versus the current I or versus V. a: Experimental (Sample M147-V). The average slope of the $R_d(I)$ curves and their shift upon the reversal of measurement direction are of instrumental origin. b: Three experiments with the same sample at $T = 4.2$ K, alternated with cycles of heating the sample to $T \simeq 300$ K. c: Theoretical predictions. 1—$Q_0/e = 0$; 2—0.25; 3—0.5; 4—0.75 $(C_2/C_1 = 1)$.

where C_1 is the capacitance of the junction with the lower conductance ($G_1 \ll G_2$). According to the theory, the times at which an electron tunnels across junctions 1 and 2 are mutually correlated, although successive events of tunneling across one of the junctions may be uncorrelated. Figure 2c shows the functional dependence $R_d(V)$ calculated from the theory of Ref. 5 for the experimental parameter values $C_1 = e/\Delta V = 3.17 \times 10^{-7}\ F$, $T = 4.2$ K, and $\Delta(T) = 1.2$ meV, with the one adjustable parameter C_2/C_1, for several values of the parameter Q_0, which is the fractional part of the net electric charge of a granule. A comparison of Figs. 2b and 2c leaves hardly any room for doubting the validity of the interpretation offered above.

Furthermore, our structures can be used in a study of subelectron changes in Q_0, which apparently involve a movement in the tunnel barriers of isolated impurities with net charges over a distance on the order of interatomic distances.[3] Preliminary studies showed that at liquid-helium temperatures Q_0 remains constant within at most $3 \cdot 10^{-2}e$ for tens of minutes during the applicaiton of voltages of tens of millivolts. At room temperature, however Q_0 may vary rapidly, by an amount on the order of e, as a result of the application of the voltage of only a fraction of a millivolt. Both of these results are extremely favorable for possible practical applications of this effect.[5]

We wish to thank A. G. Odintsov for assistance in synthesizing the samples.

[1] A. V. Averin and K. K. Likharev, Zh. Eksp. Teor. Fiz. **90**, 733 (1986) [Sov. Phys. JETP **63**, 427 (1986)].
[2] H. R. Zeller and I. Giaever, Phys. Rev. **181**, 789 (1969).
[3] J. Lambe and R. C. Jaklevic, Phys. Rev. Lett. **22**, 1371 (1969).
[4] I. O. Kulin and R. I. Shekhter, Zh. Eksp. Teor. Fiz. **68**, 623 (1975) [Sov. Phys. JETP **41**, 308 (1975)].
[5] K. K. Likharev, Preprint No. 2/1986, Physics Faculty, Moscow State University.

Translated by Dave Parsons