Coulomb blockade effects at room temperature in thin-film nanoconstrictions fabricated by a novel technique

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A technique was developed to fabricate and probe nanosize tunneling structures in thin metallic films. Using oblique evaporation through conventional undercut electron-beam lithographic masks, as the sample resistance was measured *in situ*, we defined constrictions with widths and lengths of about 10 nm in thin granular palladium films. The tunneling conductivity through a network of metallic grains was studied. Single electron tunneling transistor effects were registered. An electrostatic gate voltage at room temperature could clearly modulate Coulomb blockade offsets of the order of 0.1 V in the current–voltage curves. © *1998 American Institute of Physics.* [S0003-6951(98)02250-5]

The electrical properties of nanostructures with ever-smaller size have been studied intensively during the past decade. New quantum effects[1] and novel types of electronic devices, like single electron transistors,[2] are being searched in structures that are beyond the capability of conventional electron-beam lithography. Therefore, considerable effort is put in novel fabrication techniques involving, e.g., self-assembled nanostructures,[3–5] molecules,[6] and scanning tunneling microscope (STM) lithography.[7] However, the technological problem of putting a small object in a tunneling gap is considerable and the subject of recent publications.[8,9] The technique used in Refs. 10 and 11 works well, but mainly for Al particles. In this letter, we describe a simple procedure to produce metallic nanoparticles in a nanogap between self-aligned electrodes. Granular Pd film was deposited through a conventional electron-beam mask under an angle. Electrical measurements could be performed during the fabrication and allowed to control the width of a nanoconstriction. Single electron tunneling (SET) transistor effects were routinely observed at room temperature in such a structure.

Thin granular AuPd films were used to fabricate SET transistors,[12,13] where Coulomb blockade $I-V$ curves were observed and current was modulated by an electrostatic gate at 77 K in a planar nanostructure. We have chosen Pd as the evaporated material because its high melting temperature gives hope that diffusion processes at room temperature will not modify the granular structure of the film. The onset of (activated) conductivity occurs at a film thickness of about 1–2 nm (at an evaporation rate 0.5 Å/s), and, hence, the grains in a Pd film are small. The background pressure during the deposition was 3×10^{-7} mbar.

To overcome the resolution limit of electron-beam lithography, we used an oblique evaporation through a lift-off mask, as developed by Dolan.[14] This technique allowed the device linewidth to be reduced as compared to the opening in the resist. Figure 1 shows the mask shape and demonstrates the reduction of a bridge size by deposition onto a tilted silicon substrate. A standard double-layer (PMMA)-copolymer system was used, with a thickness of the top PMMA layer of $\delta = 150$ nm. Assuming a rectangular shape of the resist walls, we expect the gap in the mask, with a width $W \approx 100$ nm, to be closed completely at the angle arctan(W/δ), about 35° to the substrate normal in our geometry. Since our electron-beam patterning is irreproducible at the level of 10 nm, this angle is different from experiment to experiment even with the nominally same mask.

Therefore, sample conductivity during film deposition was the only guide in making a decision whether the tilt of the substrate should be decreased in order to open the gap. The lift-off geometry suits perfectly well for such *in situ* measurements of the fabricated device. Due to an undercut in the copolymer layer, there is no electrical contact between the device and the masked-off part of the film, which covers

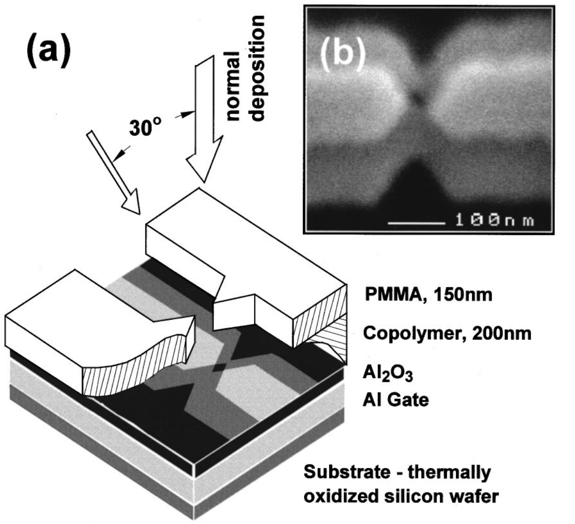

FIG. 1. (a) Schematic view of the mask for constriction. (b) Test deposition to determine the mask geometry. A scanning electron microscopy image of two overlapping 25 nm thick Au films: one is the result of deposition perpendicular to the substrate, demonstrating the initial mask shape; the other is defined by the angle deposition, showing the reduced width of constriction.

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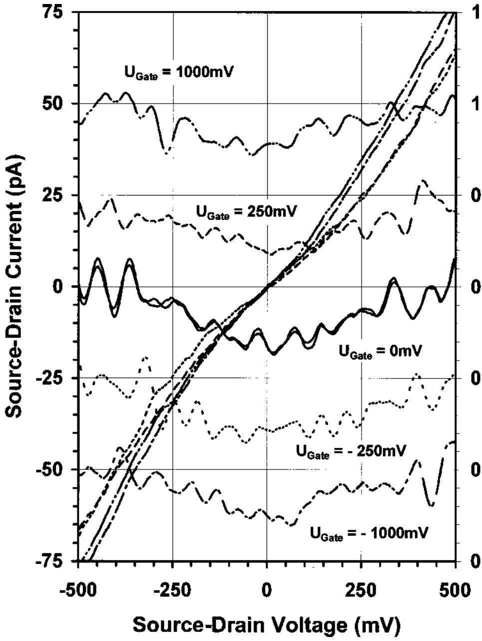

FIG. 2. Set of current–voltage characteristics $I-V$ and their derivatives dI/dV taken at different gate voltages U_{Gate}. Similarly marked curves for current–voltage characteristics and derivatives correspond to the same gate voltages. Derivative curves reveal structure interpreted as ''Coulomb staircases'' with a period of about 80 mV. The dI/dV for $U_{\mathrm{Gate}}=-1000$ mV is in correct scale while the other ones are successively shifted upwards by 0.2 nS. Two curves dI/dV for $U_{\mathrm{Gate}}=0$ are shown for two different $I-V$ measurements at this gate voltage, taken shortly one after another. The $I-V$ curves for $U_{\mathrm{Gate}}=0$ are omitted to make the picture more transparent.

the top layer of the resist. The electrometric circuit allowed *in situ* measurements of the sample resistance as high as $5\times10^{10}\ \Omega$.

To ascertain that the gap in the structure is closed, we started evaporation with a tilt angle of about 45°. About 4 nm of Pd was evaporated at this angle to form voltage and current contacts (the contact of the structure to large-scale gold pads was thus established). The tilting angle was then decreased stepwise (with a step of about 5°), widening the gap by 10 nm in every step. After each step in the tilt, we evaporated 2 nm of material while measuring the sample conductivity in order to check if the gap remained. (After 3–4 iterations, low-resistance palladium contacts were formed everywhere except in the very gap region.) Once a finite sample conductance was reached, the film thickness could be increased incrementally at the fixed tilt angle, so that in one experiment a set of different samples could be measured with a width of the constriction not exceeding the nominal value of 10 nm. Each sample consisted, therefore, of two low-resistance electrodes connected through a nanobridge made of a granular film. A third electrode, made of oxidized aluminum (air, 20 min in an oven at 135 °C), was prepared in advance on the substrate right under the constriction and served as an electrostatic gate (see also Fig. 1). A separate electrometric circuit checked the leakage current through the gate insulation. The voltage on the gate was varied within ±2 V, limited by the oxide breakdown at about 3 V. Measurements were performed at room temperature only, as an ordinary high-vacuum environment would not permit repeated depositions without absorbed gases if the substrate was cold.

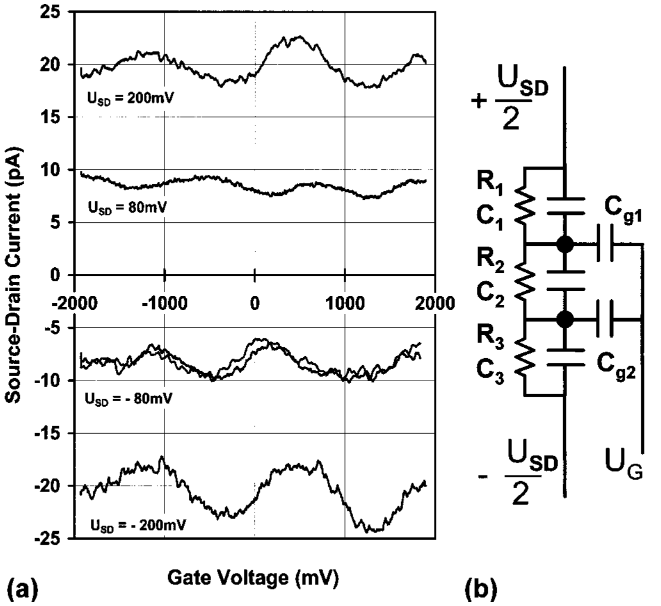

FIG. 3. (a) Current modulation by the electrostatic gate at room temperature. The periodicity is about 2 V. Traces at $U_{\mathrm{SD}}=-80$ mV were taken twice, the second 12 min later. (b) A model circuit reproducing the experimental data. $R_1=2.0\times10^{8}\ \Omega$; $C_1=2.8\times10^{-19}$ F; $R_2=3.0\times10^{6}\ \Omega$; $C_2=2.8\times10^{-19}$ F; $R_3=5.2\times10^{9}\ \Omega$; $C_3=1.6\times10^{-18}$ F; $C_{g1}=8\times10^{-20}$ F; and $C_{g2}=9\times10^{-20}$ F.

Clear signs of Coulomb blockade effects were observed in samples in the resistance range $10^{7}-10^{10}\ \Omega$. Figure 2 shows a set of current–voltage characteristics taken for a 2.5 nm thick film at different gate voltages. The current modulation by the electrostatic gate is shown in Fig. 3(a). It has essentially the same period of about 2 V at bias voltages up to 200 mV. This corresponds to a gate capacitance of the order of 9×10^{-20} F. Traces taken a few minutes after each other demonstrate a decent short-term sample stability. The recording time for each trace was about 1 min.

The observed effects are of truly mesoscopic nature. The transport through the described constriction is determined by electron tunneling through a network of islands within a square 10 by 10 nm^2. At a film thickness of 2–3 nm, we may expect the islands in the film to be of about the same size (2–3 nm), so a maximum of 10–30 islands may participate in the conductivity of the film. However, the actual number of islands that determines the conductivity path through the constriction is less than that number. A percolation problem has to be solved, where the tunneling resistance between any couple of islands depends exponentially upon the randomly distributed tunneling gaps between them. Following Ref. 15, we may expect that the conductance of such a structure is determined by the optimum, low-resistive tunneling path through a single chain of islands.[16] For our particular geometry, such a chain would consist of about three to four islands and its resistance is dominated by a single weak link, i.e., by the gap with the highest tunneling resistance.

Coulomb blockade effects at room temperature do not change the described qualitative picture very much since the charging energy of our particles is just a few times more than the thermal energy and much lower than typical heights of tunneling barriers. It means that Coulomb blockade effects do not change percolation paths and in the first approximation lead to an additional resistance of the selected chain, arising from the Coulomb effects on each island.[15] Since such a resistance depends exponentially on the charging energy of the island at finite temperature, one island with the smallest capacitance would probably dominate the additional resistance in such a short array. Basically, two cases can be imagined, depending upon a mutual position of the tunneling gap with the highest resistance and the smallest island. The first case is when the island is situated next to this important tunneling gap. It would give a pronounced current modulation by the electrostatic gate, but statistically it is not favorable and has been observed just a few times. In this letter we present the more typical case when the high-resistive tunneling gap and the smallest grain are not next to each other.

In this case, we can still see the traces of the current modulation caused by grains next to the high-resistive tunneling gap (weak Coulomb staircases and weak nonlinearity at low biases), but the main contribution to the current modulation comes from the transistor built around the smallest island. A double-grain model of such a system is shown in Fig. 3(b). It gives a rather good fit between experimental electrical characteristics and Monte Carlo simulations based on the ''orthodox'' theory.[2] Such a simple double-grain system already captures the main features observed in the experiment.[17] The precise fit, however, would be beyond the accuracy of the standard model neglecting the effects like tunneling barrier suppression at high biases,[6] the possible effect of other grains, etc. The parameters of the model are close to those observed for grains of similar size in STM experiments.[5] The relatively small capacitance of $(8-9)\times 10^{-20}$ F between grains and gate can be attributed to a screening by adjacent grains, which are better coupled to the source and drain electrodes. Presumably, such a screening would be reduced in even narrower constrictions.

We have studied[18] more than 20 samples with different length-to-width ratios. Similar results were obtained in all structures that were nominally 10 nm wide and had different lengths of constrictions. Gate effects were found for all samples in the resistance range of $10^7-10^{10}\ \Omega$ (film thickness range 1–3 nm). The motion of the offset charges limited the long-term sample stability, resulting in phase shifts of the observed gate dependencies. Low-resistance samples were less stable, probably because diffusion becomes more important in structures with smaller tunneling gaps between the grains. Wider structures, in the same resistance range, showed linear $I-V$ curves without any gate dependence. Thus, the width of the structure is vital for the observed effects. Note that the described technique allows a further reduction of the width.

To summarize, we have implemented a procedure for fabricating nanometer-sized constrictions in thin films using conventional electron-beam masks. With this technique we created and studied mesoscopic objects at room temperature. The conductance through such a constriction in a granular film was determined by a limited network of nanometer-sized grains and was controlled by the charge on a single grain. As a result, single electron transistor effects in such small networks of grains survive even at room temperature.

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[17] Deduced from this fit the charging energies of the grains are 41 and 125 meV. This means that at room temperature (25 meV) our system is in an ''intermediate'' temperature regime, where ''stochastic'' Coulomb blockade, intrinsic to the double-grain system at low temperatures, is replaced by ordinary periodic Coulomb blockade oscillations, in agreement with the paper by I. M. Rusin, V. Chandrasekhar, E. I. Levin, and L. I. Glazman, Phys. Rev. B **45**, 13469 (1992).

[18] All the measurements were performed on the samples kept in vacuum. The sample resistance changed (normally increased) and gate effects disappeared when the sample was exposed to air. However, dry oxygen (10 mbar) did not affect our samples.