Room Temperature SET Effects in Nano-constricted Granular Films

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Abstract

A technique was developed to fabricate and probe nanosize tunneling structures in thin metallic films. Using oblique evaporation through conventional electron-beam lithographic masks as the sample resistance was measured in situ, we defined constrictions with widths 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. Coulomb blockades up to about 0.1 V were observed, and they could clearly be modulated by an electrostatic gate voltage at room temperature.

The electrical properties of nanostructures with ever smaller size have been studied intensively during the past decade in a search for new quantum effects and novel types of electronic devices. The scientific interest has moved towards objects with sizes of a few nanometers. Such objects cannot be prepared by conventional electron beam lithography with a resolution being limited to ≈10-20 nm. Therefore, considerable effort is put in novel fabrication techniques involving, e.g., self-assembled nanostructures[1,2,3,4], molecules[5], and STM lithography[6]. A difficult task is to connect these naturally grown objects to large-scale contacts that provide electrical measurements. In our work, naturally grown palladium clusters and connecting leads are prepared simultaneously, in one cycle of granular film deposition. Single electron effects[7] are observed in a 10 nm wide granular strip at room temperature.

Thin granular AuPd film was used to fabricate SET transistors[8,9], where Coulomb blockade I-V curves were observed and current was modulated by an electrostatic gate at 77K in a planar nanostructure. We have chosen Pd as the evaporated material because its high melting temperature gives a hope that diffusion processes at room temperature will not destroy 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 of 0.5 Å/sec), and, hence, the grains in a Pd film are small. One nanometer of SiO underlayer was used to promote conductivity at low thickness.

The lateral size of the evaporation source is important in the fabrication of devices with features as small as a few nanometers. In our experiment a home-built effusion cell was used with a diameter of 1 mm; for a distance between the source and the substrate of 20 cm this gives a 'geometrical' blur of sample features of about 1 nm. 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[10], in combination with in situ control of sample conductance. Dolan's 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 $W\approx100$ nm to be closed completely at the angle arctan(W/δ), about 35 degrees 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 nominally the same mask. Therefore, sample conductivity during film deposition was the only guide to make 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

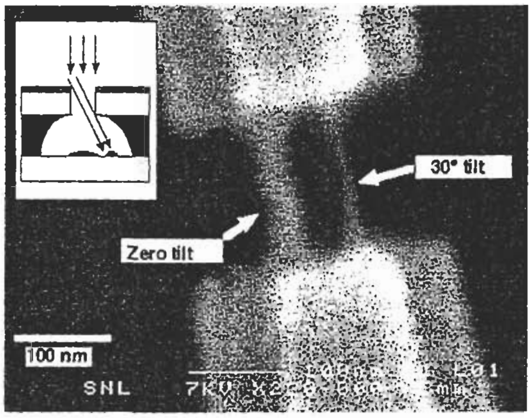

Fig.1 Test deposition showing the mask geometry. A SEM image of two overlapping 25 nm thick Au films: one is the result of deposition perpendicular to the substrate, demonstrating the initial mask shape; the other is defined by the angle deposition, showing the reduced width of constriction.

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 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 make sure that the gap in the structure is closed, we started evaporation with a tilt angle of about 45 degrees. 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 contacts was thus established). The tilting angle was then decreased stepwise (with a step of about 5^0), 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. 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, was prepared in advance on the substrate right under the constriction and served as an electrostatic gate. 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.

Clear signs of Coulomb blockade effects were observed in samples in the resistance range 10^8-$10^{10}\,\Omega$. Fig.2(a) shows a set of I-V curves taken for a 1,1 nm thick film at different gate voltages. Signs of "Coulomb staircases" with a period of about 100 mV can be seen. The current modulation by the electrostatic gate at different source-drain voltages for the same device is shown in Fig.2(b).

Samples made in the described manner must have at least one 10 nm wide constriction, which determines the device resistance. The transport through such a constriction is determined by electron tunneling through a network of islands. At a film thickness of 1-2 nm, we may expect the islands in the film to be of about the size of 2-3 nm, so a "cross-section" of this constriction consists maximum of 3-4 islands. The tunneling resistance between any couple of islands depends exponentially upon the randomly distributed tunneling gaps between them. Taking these facts into account, we may expect that one optimum, low resistive tunneling path through a single chain of islands determines the conductance of such a structure. The resistance of such a chain is, in turn, dominated by a single weak link, i.e. by the place with the highest tunneling resistance. The concept of a granular material as a "virtually" nonconducting medium with a few one-dimensional conducting paths was developed in Ref.[11], where it was shown that the formation of such chains is favored in granular materials.

The island adjacent to the most resistive tunneling gap may act as the central island in a SET geometry. It would influence the electron transport through the chain even at room temperature, as its size is in the range of 2-3 nm, in agreement with our observation. Coulomb staircases are expected in an extremely inhomogeneous structure. From the period of the Coulomb staircase we can estimate the total capacitance of the grain to 1.6×10^{-18} F. The relatively small capacitance of 0.8×10^{-19} F from the grain to the 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. The complicated shape of the

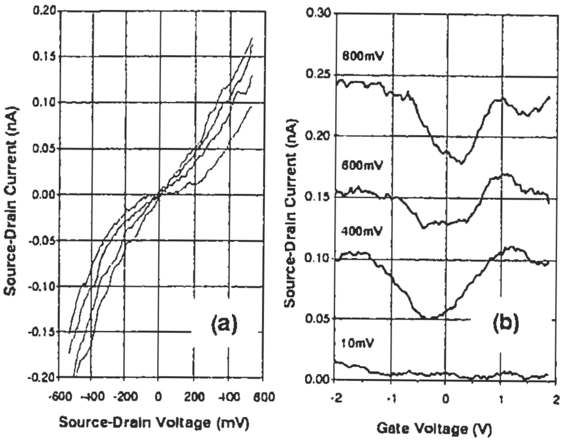

Fig.2 (a) A set of current-voltage characteristics taken at different gate voltages. (b) Current modulation of the device by the electrostatic gate. Source-drain voltages are indicated.

current modulation curves can be attributed to the presence of a second weak link in a chain. We have studied samples with different widths. Similar results were obtained in all structures that were nominally 10 nm wide. Gate effects were found for all samples in the resistance range of 10^8-10^{10} Ω (film thickness range 1-3 nm). Low resistance samples were less stable, probably because diffusion becomes more important in structures with smaller tunneling gaps between the grains. However, in the G Ω resistance range sample stability was fairly good.

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. Single electron transistor effects in such a small networks of grains were observed at room temperature.

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References

1. R.P. Andres, T. Bein, M. Dorogi, Science, **272**, 1323 (1996)
2. E. S. Soldatov, V. V. Khanin, A. S. Trifonov, D. E. Presnov, S.A. Yakovenko, G. B. Khomutov, G.P. Gubin, V. V. Kolesov, JETP Lett., **64**, 556 (1996)
3. K.-H. Park, J. S. Ha, W. S. Yun M. Shin, K.-W. Park, E.-H. Lee, Appl. Phys. Lett., **71**, 1469 (1997)
4. A. Bezryadin C. Dekker, G. Schmid, Appl. Phys. Lett., **71**, 1273 (1997)
5. D. Porath, O. Millo, J. Appl. Phys. **81**, 2241 (1997)
6. K. Matsumoto, M. Ishii, K. Segawa Y. Oka, B. J. Vartanian, J. S. Harris, Appl. Phys. Lett., **68**, 34 (1996)
7. D.V.Averin, K.K.Likharev, in Mesoscopic phenomena in solids, edited by B.L. Altshuler, P.A.Lee, R.A.Webb, (North-Holland, New-York, 1991), Chapt.6.
8. W.Chen, H.Ahmed, J. Vac. Sci.Tecnol. **B13**(6), 2883 (1995)
9. W.Chen, H.Ahmed, K.Nakazoto, Appl. Phys. Lett. **66**(24), 3383 (1995)
10. G. J. Dolan, Appl. Phys. Lett., **31**, 337 (1977)
11. R.I. Shekhter, I.O. Kulik, Sov.J.Low Temp.Phys., **1**, 215 (1975)

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