

Fabrication of nanoscale gaps in integrated circuits

Roman Krahne,^{a)} Amir Yacoby, Hadas Shtrikman, and Israel Bar-Joseph
Department of Condensed Matter Physics, Weizmann Institute of Science, 76100 Rehovot, Israel

Tali Dadosh and Joseph Sperling
Department of Organic Chemistry, Weizmann Institute of Science, 76100 Rehovot, Israel

(Received 16 April 2002; accepted for publication 24 May 2002)

Nanosize objects such as metal clusters present an ideal system for the study of quantum phenomena and for the construction of practical quantum devices. Integrating these small objects in a macroscopic circuit is, however, a difficult task. So far, nanoparticles have been contacted and addressed by highly sophisticated techniques not suitable for large-scale integration in macroscopic circuits. We present an optical lithography method that allows for the fabrication of a network of electrodes separated by gaps of controlled nanometer size. The main idea is to control the gap size with subnanometer precision using a structure grown by molecular-beam epitaxy. © 2002 American Institute of Physics. [DOI: 10.1063/1.1495080]

There is growing interest in the study of the electrical properties of nanosized objects. In these small scale structures, the size quantization and Coulomb energies are considerably enhanced and can exceed the thermal energy kT at room temperature. Therefore, such systems are ideal for studying quantum phenomena and for constructing practical quantum devices. The single-electron transistor (SET) has emerged as an important experimental platform in both aspects. It consists of a nanosize metal island connected through two tunnel barriers to the leads.¹ The SET also presents most of the problems that characterize device physics at this length scale: fabrication of macroscopic leads with a nanometer-size gap, definition of a nanosize island, positioning it between the leads, and controlling the tunnel barriers with high precision. The fabrication of the leads has proven to be a particularly challenging task. It requires nanoscale resolution for defining the gap, and the ability to integrate the device in a highly connected and complex macroscopic circuit. A variety of solutions to this issue have been suggested, of which the majority are appropriate for contacting only a single device. These include using the tip of a scanning tunneling microscope for contacting,² electromigration,³ breaking thin wires,⁴ or metal deposition on electron-beam defined structures.^{5–7} Other techniques attempted to use a conducting DNA-based network⁸ to solve the connectivity issue. Recently logic circuits with carbon nanotube have been reported.⁹

In this letter we present a method that allows us to fabricate a network of electrodes that are separated by gaps of controlled nanometer size using conventional optical lithography. The gap size is determined by the thickness of a molecular beam epitaxy (MBE) grown layer embedded in a sandwich structure. This layer is selectively etched and after metal evaporation a gap is formed exactly where the layer was removed. Controlled positioning of the nanoparticles between the electrodes is done by electrostatic trapping.⁷ We show that the device exhibits clear Coulomb blockade (CB)

characteristics in the transport measurements and can function as a SET.

The sample fabrication is illustrated in Figs. 1(a)–1(d). A quantum-well (QW) structure is grown on a GaAs substrate by MBE. The thickness of the two undoped AlGaAs layers above and below the QW are 200 and 100 nm, respectively. The thickness of the embedded GaAs layer is varied between 5 to 10 nm in various samples. In the first step, 200-nm-high mesa structures are defined by optical lithography and a standard wet-etching process such that the GaAs layer is exposed on the side of the mesa [Fig. 1(a)]. Next, the pattern of the electrodes is defined by optical lithography. On the exposed areas a few tens of nanometers of the GaAs layer (from the side) are removed by selective wet-etching¹⁰ using citric acid and H₂O₂ (5:1), as shown in Fig. 1(b). The selectivity of the etching between GaAs and AlGaAs is nominally 100:1, hence the width of the etched layer is the QW width. The electrodes are fabricated by thermally evaporating a thin film (5–15 nm) of PdAu from a direction perpendicular to the plane of the wafer surface such that a gap is formed exactly where the GaAs was removed [Fig. 1(c)]. The size of the gap is determined by the crystal structure (which can be controlled with subnanometer precision), surface roughness of the etched AlGaAs/GaAs interface due to selective etching (less than one nanometer for short etching times), and metal evaporation which can be controlled on the nanometer scale. We manage to fabricate gaps which are less than 5 nm wide and estimate the minimum gap size that can be achieved by our method to be about one to two nanometers. An important advantage of this method is that since the mesa structure and the electrode pattern are defined by optical lithography, it allows the simultaneous fabrication of many electrodes separated by nanosize gaps on the wafer surface [see illustration in Fig. 1(f)].

To position the nanoparticles between the electrodes we use the method of electrostatic trapping:⁷ the electrodes are immersed in an aqueous or organic solution of Au clusters. The Au clusters can be chemically fabricated with controlled size in the range of 1 to 100 nm diameter.¹¹ By applying a voltage between the electrodes a dipole moment is induced

^{a)}Electronic mail: roman.krahne@weizmann.ac.il

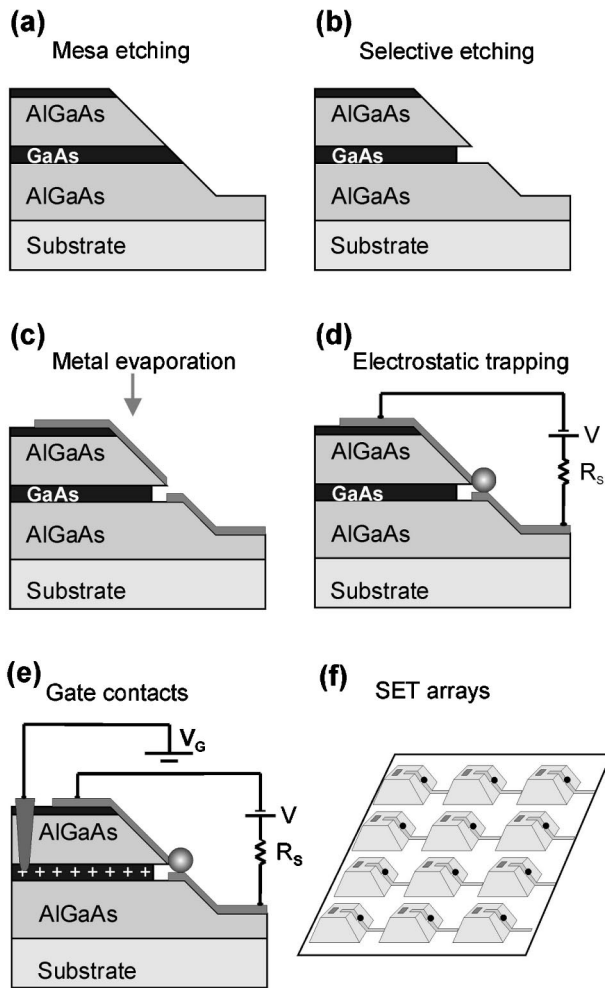


FIG. 1. (a) Sketched description of the fabrication process: a mesa structure is defined by optical lithography and wet etching, (b) selective wet etching removes some tens of nanometers of the GaAs layer, (c) after metal evaporation two electrodes are separated by a gap of a few nanometers, (d) the nanocluster is positioned by electrostatic trapping (see Ref. 7), (e) a heavily doped GaAs layer could be used as a gate, and (f) the process allows for the fabrication of a network of devices with gaps of controlled nanometer size.

in the Au cluster and it is attracted to the point of strongest electric field, i.e., the gap, by dipole-dipole interaction. A series resistance, $R_S = 1 \text{ G}\Omega$, is integrated in the circuit such that when trapping occurs the voltage drops on that resistor and the device cannot trap more clusters. In this way only a single cluster is trapped in the gap. The setup is sketched in Fig. 1(d). For trapping, we apply an AC voltage of 8 V at 50 kHz for a few seconds. AC voltage is used to increase the impedance of the liquid solvent and also to be able to trap charged particles. After trapping, the liquid is blown off with N_2 .

In order to form tunnel barriers between the electrodes and the clusters, the electrodes can be covered with thiol molecules before the trapping.^{3,5} However, we find that if the cluster diameter is only slightly larger than the gap, a tunnel barrier can exist even for uncoated electrodes. This tunnel barrier could arise from organic molecules on the cluster surface that stabilize the Au clusters, or from contamination on the electrodes formed during the trapping process.¹² We note, however, that the strength of this barrier is reasonably reproducible as evidenced by transport measurements. The geometry sketched in Fig. 1(d) suggests that the contact of

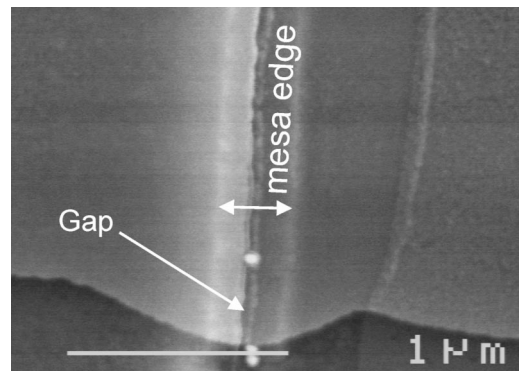


FIG. 2. SEM image of the metal electrodes separated by a gap slightly larger than 10 nm and a single trapped Au cluster with 60 nm diameter. Bright regions are covered with PdAu.

the trapped cluster to the lower electrode is better than to the upper one. This asymmetry in the contacts should lead to different tunnel resistances, which is indeed the case as we will see later.

A scanning electron microscope (SEM) image of a single trapped cluster between two metal electrodes is displayed in Fig. 2. In the center of the image we see the mesa edge, with the mesa plateau to the left, and the etched groove separating the electrodes in the vertical direction. The trapped cluster is seen below the center of the image. Two more clusters, which do not bridge the gap, can be observed at the right side of the scale bar. The gap size is slightly larger than 10 nm and the width of the electrodes is a few microns each. To enhance the visibility of the trapped cluster, clusters having a relatively large diameter of 60 nm are displayed.

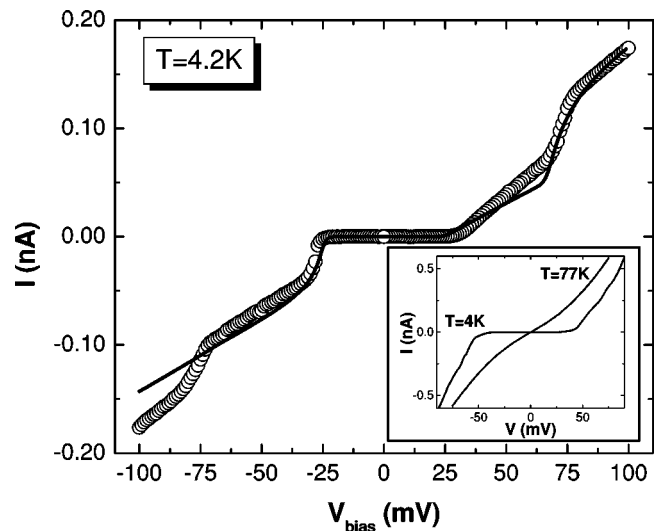


FIG. 3. Experimental and theoretical $I-V$ curves of trapped metal clusters. Large circles show experimental data measured on a Au cluster with 10 nm diameter at 4.2 K. The asymmetry with respect to positive or negative V_{bias} , i.e., a linear onset at +25 mV and a step like onset at -25 mV, results from an asymmetry in tunnel resistances R_1 and R_2 to the electrodes and a residual fractional charge Q_0 on the trapped cluster. A theoretical $I-V$ curve is depicted by the solid line with fractional charge $Q_0 = 0.24e$, $R_1 = 30 \text{ M}\Omega$, $R_2 = 400 \text{ M}\Omega$, $C_1 = C_2 = 5 \times 10^{-19} \text{ F}$, and a stray capacitance to the electrodes of $C_g = 2.5 \times 10^{-18} \text{ F}$. The inset shows experimental $I-V$ curves for different temperatures of another sample where a Au cluster with 10 nm diameter was trapped.

Figure 3 shows an I - V curve measured on a trapped Au clusters with 10 nm diameter at 4.2 K. A clear CB behavior can be observed up to $V_{\text{bias}} \approx \pm 25$ mV. For larger voltages, $|V_{\text{bias}}| > 25$ mV, we observe a strong asymmetry in the I - V curve with respect to positive and negative voltage. For positive bias voltage we find a linear rise in current in the range of $25 \text{ mV} < V_{\text{bias}} < 65 \text{ mV}$, whereas for negative bias we observe a step-like feature at $V_{\text{bias}} = -25$ mV, which then saturates in a linear slope. This asymmetric behavior has been discussed by Hanna and Tinkham¹² assuming different tunnel resistances to the electrodes, i.e., $R_2 \gg R_1$. They distinguished four different cases in a $C_1/C_2 - Q_0$ phase diagram, each with a qualitatively different onset in conduction. Here, C_1 and C_2 are the capacitances of the trapped cluster to the electrodes and Q_0 is the residual fractional charge on the cluster. The solid line displayed in Fig. 3 corresponds to the case of $C_1/C_2 \approx 1$ and $Q_0 = 0.24e$. In order to fit our data,^{13,14} we used $R_1 = 30 \text{ M}\Omega$, $R_2 = 400 \text{ M}\Omega$ (which satisfies very well the assumption of $R_2 \gg R_1$,) and for the capacitances $C_1 = C_2 \approx 5 \times 10^{-19} \text{ F}$, and a stray capacitance to the electrodes of $C_g = 2.5 \times 10^{-18} \text{ F}$. The inset in Fig. 3 shows transport measurements of another sample at different temperatures, where a Au cluster with a diameter of 10 nm was trapped. At $T = 4.2 \text{ K}$ we find the CB region to be slightly larger than in the previous sample. Clearly, also at higher temperatures ($T = 77 \text{ K}$) a nonlinear behavior in the I - V curve can be observed. Measuring the I - V curve of a certain device at different times after the cooling reveals the abrupt occurrence of changes in the characteristics of the conduction onset and slight changes in the size of the Coulomb blockade region. This behavior was also observed by Klein and coworkers⁵ and was attributed to changes in the local charge distribution in the vicinity of the cluster. Repeated warming and cooling of the device leads to similar effects.

An important question is how to apply a gate voltage to the trapped cluster in order to get true transistor functionality. This is not trivial and to the best of our knowledge, so far no gated device using metal clusters of nanometer size has been realized. The major problem is the screening of the gate voltage by the comparatively large electrodes in close vicinity to the small cluster. Our design provides a method to position a gate electrode only a few tens of nanometers away from the cluster. As illustrated in Fig. 1(e), the GaAs layer, which has been used for the selective etching, can also be heavily doped during the MBE growth and then used as a gate. The great advantage of this method is that due to the mesa design, each cluster can be gated individually. This would al-

low large scale integration of SETs with true transistor functionality on semiconductor surfaces by optical lithography [Fig. 1(f)], especially if the series resistor R_S is also integrated in the electric circuit on the wafer. This would enable simultaneous trapping on a large number of devices. Another approach to position a gate electrode in very close proximity to the cluster is to grow the lower AlGaAs layer only a few tens of nanometers thick and to use a n -doped GaAs substrate. In this design the substrate could serve as a gate; however, this setup would not allow the individual gating of each cluster. Experiments to realize a gated structure are in progress and will be reported elsewhere.

In conclusion, we presented a fabrication method to define compatible contacts to nanosize objects by optical lithography and selective etching. In transport measurements at low temperatures, we demonstrated Coulomb blockade behavior. In order to operate the nanoclusters as SETs, we proposed a gate design that allows large scale integration of such devices on semiconductor surfaces.

We want to thank D. Mahalu for helpful discussions and her assistance in the early stages of this work. The research of one of the authors (R.K.) was supported by the "Fritz Thyssen Stiftung" and the European Commission under Contract No. HPRI-CT-1999-00069. This work was partially supported by a grant from the Israel Science Foundation to one of the authors (J.S.).

¹T. A. Fulton and G. J. Dolan, Phys. Rev. Lett. **59**, 109 (1987).

²D. Porath, Y. Levi, M. Tarabiah, and O. Millo, Phys. Rev. B **56**, 9829 (1997).

³H. Park, A. K. L. Lim, A. P. Alivisatos, J. Park, and P. L. Euen, Appl. Phys. Lett. **75**, 301 (1999).

⁴M. A. Reed, C. Zhou, C. J. Muller, T. P. Burgin, and J. M. Tour, Science **278**, 278 (1997).

⁵D. L. Klein, P. L. McEuen, J. E. Bowen Katari, R. Roth, and A. P. Alivisatos, Appl. Phys. Lett. **68**, 2574 (1996).

⁶D. Porath, A. Bezryadin, S. de Vries, and C. Dekker, Nature (London) **403**, 635 (2000).

⁷A. Bezryadin, C. Dekker, and G. Schmid, Appl. Phys. Lett. **71**, 1273 (1997).

⁸E. Braun, Y. Eichen, U. Sivan, and G. Ben-Yoseph, Nature (London) **391**, 775 (1998).

⁹A. Bachtold, P. Hadley, T. Nakanishi, and C. Dekker, Science **294**, 1317 (2001).

¹⁰G. C. de Salvo, W. F. Tseng, and J. Comas, J. Electrochem. Soc. **139**, 831 (1992).

¹¹G. Schmid, *Clusters and Colloids* (VCH, New York, 1994).

¹²A. E. Hanna and M. Tinkham, Phys. Rev. B **44**, 5919 (1991).

¹³A. Korotkov and Y. Nazarov, Physica B **173**, 217 (1991).

¹⁴A. Korotkov, URL: <http://qt.tn.tudelft.nl/CHARGE/>