Multilevel Atomic-Scale Transistors Based on Metallic Quantum Point Contacts

By *Fangqing Xie, Robert Maul, Christian Obermair, Wolfgang Wenzel,**
Gerd Schön, and *Thomas Schimmel**

Atomic scale transistors[1–3] based on metallic quantum point contacts were demonstrated recently. They allow controlled binary switching of an electrical current between a conducting "on state" and a non conducting "off state" by means of an independent gate electrode. The devices that operate reproducibly at room temperature open fascinating perspectives towards quantum electronics and logics on the atomic scale. Even an integrated circuit consisting of atomic scale transistors[3] as well as a nanoelectromechanical atomic switch[4] were shown.

Here, we demonstrate a multilevel atomic quantum transistor that allows gate controlled switching between *different* quantized conducting states. Multilevel logic and storage devices on the atomic scale are of great interest as they will allow more efficient data storage and processing with a smaller number of logical gates. Our experiments are combined with detailed computer simulations that provide a detailed understanding of the multilevel switching process. The results provide a basis for the future development of ultra small devices for multilevel logics on the atomic scale.

Metallic quantum point contacts exhibit two striking features, namely their atomic scale dimension and electronic quantum transport, both which have motivated extensive experimental and theoretical work by many groups in recent years.[1–12] Experimentally, three different approaches are available for the fabrication of metallic quantum point contacts: mechanically controlled deformation of thin metallic junctions,[4–6] electro chemical fabrication techniques,[1–3,7–11] and high resolution lithography.[5,12] In metallic point contacts the conductance depends on both the atomic configuration within the junction[13] and the chemical valence of the metals.[2,6,14,15] Two terminal conductance switching devices based on quantum point contacts were developed with an STM like setup[16] and with electro chemical methods.[9,17] Actively controllable devices, such as atomic scale transistors or relays with three terminals were fabricated using silver quantum point contacts in an electro chemical cell.[1–3,10] Even a single atom transistor was demon strated, opening and closing an electrical circuit by the reproducible opening and closing of a single atom contact, which was actively controlled by an independent gate electrode.[1] Controlled bistable switching was performed between a quan tized, electrically conducting "on state" exhibiting a conductance of $G_0 = 2e^2/h$ ($\sim 1/12.9\,\mathrm{k\Omega}$) or preselectable multiples of this value and an insulating "off state". While reproducible switching in these cases was always performed by opening and closing a quantum point contact, i.e., by switching between a quantized conducting state and a non conducting state, it was not clear if this kind of gate electrode controlled switching is also possible between *two different* conducting states of one and the same contact. Such kind of switching would involve two different stable contact configurations on the atomic scale, between which reversible switching would occur even without ever opening the contact.

Our experiments show that such a controlled switching between *two* conducting "on states" can be implemented by manipulating the atomic configuration within the junction applying an electrochemical cycling technique.

Figure 1 shows a schematic diagram of our experimental setup. A silver point contact is deposited electrochemically in a narrow gap between two gold electrodes on a glass substrate (see [1,3] for details). Figure 2a gives a first demonstration of the operation of an atomic transistor switching not between zero and a conductance level G_1, but between *two different* quantized non zero conductance levels G_1 and G_2. In order to achieve such switching between *two* different conducting levels, the following, modified procedure was performed on an atomic scale silver quantum point contact: After formation of an atomic scale contact by electrochemical deposition from $AgNO_3$ (1 mM) + HNO_3 (0.1 M) in bi distilled water, a cycling process was performed. As soon as the conductance exceeds a preset "target" value (upper threshold) near G_2, we stop the deposition and reverse the electrochemical control potential to dissolve the junction again. After the conductance drops below a lower

[*] Dr. W. Wenzel, Prof. T. Schimmel, Prof. G. Schön
Institute of Nanotechnology and Center for Functional Nanostruc tures (CFN)
Forschungszentrum Karlsruhe, Karlsruhe Institute of Technology (KIT)
76021 Karlsruhe (Germany)
E mail: wolfgang.wenzel@kit.edu
Prof. T. Schimmel, Dr. F. Q. Xie, Dr. C. Obermair
Institute of Applied Physics and Center for Functional Nanostruc tures (CFN)
University of Karlsruhe, Karlsruhe Institute of Technology (KIT)
76131 Karlsruhe (Germany)
E mail: thomas schimmel@kit.edu
R. Maul
Institute of Nanotechnology, Forschungszentrum Karlsruhe,
Karlsruhe Institute of Technology (KIT), 76021 Karlsruhe, Germany
Prof. G. Schön
Institut für Theoretische Festkörperphysik and Center for Functional Nanostructures (CFN)
University of Karlsruhe, Karlsruhe Institute of Technology (KIT)
76131 Karlsruhe (Germany)

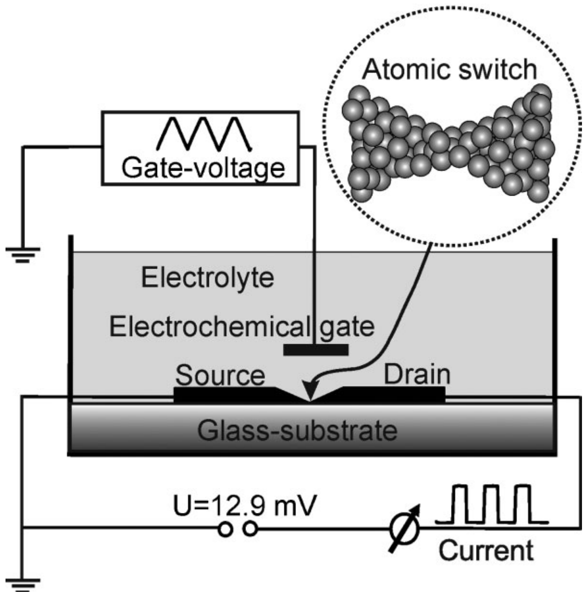

Figure 1. Schematic of the experimental setup. Within a narrow gap between two gold electrodes on a glass substrate, a silver point contact is deposited electrochemically. A multilevel atomic scale quantum con ductance switch is fabricated by applying a procedure that involves repeated computer controlled electrochemical cycling.

threshold (near G_1) we repeat the deposition/dissolution cycle. After a number of switching cycles, frequently, a new bistability develops at the point contacts. The contact changes its switching behavior and starts reproducible bistable switching between the two well defined quantized conductance levels. Now, a controlled change of the electrochemical potential U_G (the control potential or "gate" potential) leads to a controlled switching of the conductance of the quantum point contact between the two quantized conducting states G_1 and G_2.

The upper diagram of Figure 2a gives the control potential (blue) as a function of time, while the lower diagram shows the resulting, simultaneously recorded conductance versus time curve (red), respectively. The two states exhibit conductance levels of $G_1 = 3.0\ G_0$ and $G_2 = 5.0\ G_0$, respectively. Sharp transitions are observed between the two conductance levels. Figure 2b gives a further example of an interlevel quantum transistor, in this case switching between the conductance levels of $G_1 = 1.0\ G_0$ and $G_2 = 3.0\ G_0$, respectively. The observed reproducible switching can be explained by the formation of two highly stable atomic configurations of the contact area corre sponding to two different quantized "on state" conductance values, between which the contact is transformed or "switched" reversibly by applying the corresponding control potential.

Figure 2c gives an example for a transition from a switching sequence between zero and *one* finite conductance value to the switching sequence between *two* different finite values. Three distinct, stable configurations of the contact are identified at zero conductance, a lower quantized conductance level at 1.0 G_0 and a higher quantized conductance level at 3.0 G_0 from the experimental data. In both cases the upper levels are the same. Before the time indicated by the arrow, the conductance is switched reversibly between zero and 3 G_0. After the time

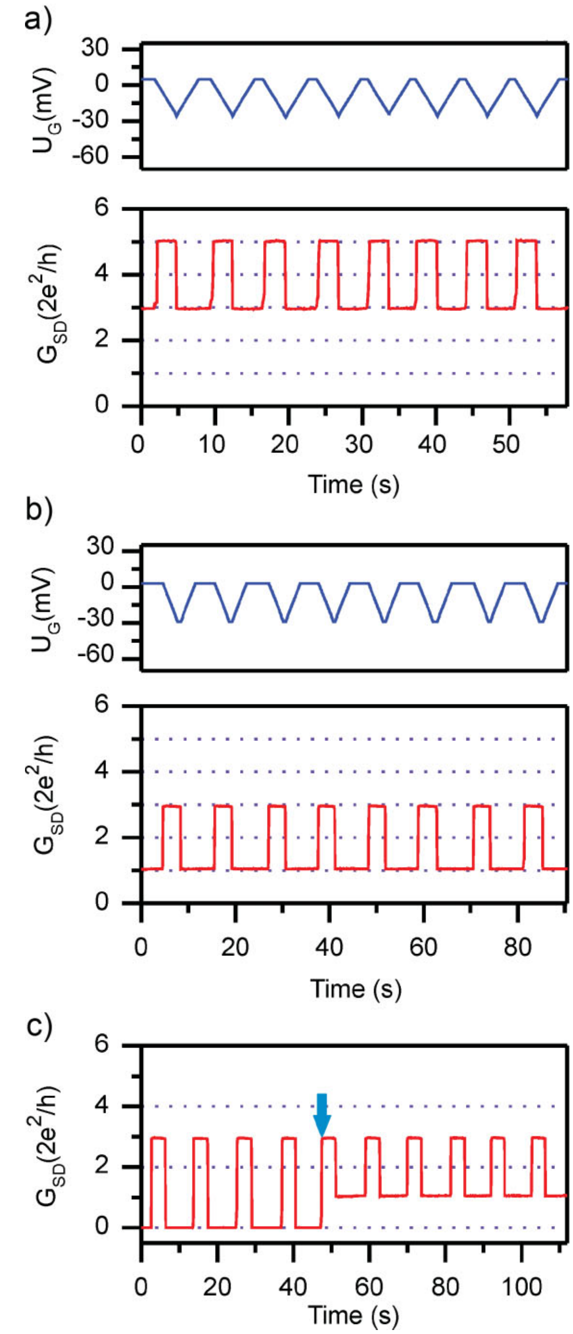

Figure 2. a,b) Experimental demonstration of the operation of an interlevel transistor based on an atomic scale silver quantum point contact. A controlled change of the electrochemical gate potential U_G leads to controlled switching of the conductance of the quantum point contact G_{SD} between two quantized conducting states. The upper diagram displays the control potential (blue curve) applied to a "gate" electrode while the lower shows the corresponding conductance switching of the point contact (red curve). The two states between which the switching occurs in (a) exhibit conductance levels of 3 G_0 (G_0 $2e^2/h$) and 5 G_0, respectively. b) Example of an interlevel quantum transistor switching between the conductance levels of 1 G_0 and 3 G_0. c) Experimental demonstration of a multilevel atomic scale transistor switching between an "off state" and two different "on states".

indicated by the arrow in Figure 2c the contact does not open completely any more but the conductance remains at a level of 1 G_0 in the lower conductance level, subsequently switching reproducibly between the two conducting states of 1 G_0 and 3 G_0. This observation excludes a switching mechanism by superposition of two independent, parallel point contacts. It rather indicates true multilevel switching between different configurations of *one and the same* point contact.

In order to explain the multilevel conductance switching in the experimental data described above, we combine atomic structure computer simulations of opening/closing processes in silver nanojunctions with zero bias conductance calculations. We generate non idealized silver electrode geometries by simulation of the deposition process: Starting from distant Ag(111) layers we evolve individual atoms in a material specific potential for silver.[18] By depositing one ion at a time,[19,20] we generate junctions with a predefined integer conductance quantum as previously described.[3] Figure 3a (left) and (right) shows two final, representative silver nano junctions consisting of 508 and 561 Ag atoms with 3 and 5 atoms in the minimal cross section (marked red), respectively (see *simulation methods* for details).

We then simulate many switching cycles for each junction. Experimental modification of the electrochemical potential modulates the interfacial tension of the embedded silver electrodes[21–23] which results in a mechanical strain on the junction. We simulate the opening/closing cycle of a junction by evolving the atoms of a "central" cluster under the influence of the electrochemical pressure. While the electrodes gradually move apart or closer together, all atoms of the central cluster relax in a quasi adiabatic path between the open and the closed conformation. The silver nano junctions in Figure 3a allow for bistable conductance switching between 0 and 3 G_0 (left) or 0 and 5 G_0 (right). In our simulations, we find a reproducible bistable electrode reconstruction of the central cluster of atoms, allowing for the bistable switching between predefined conductance values.

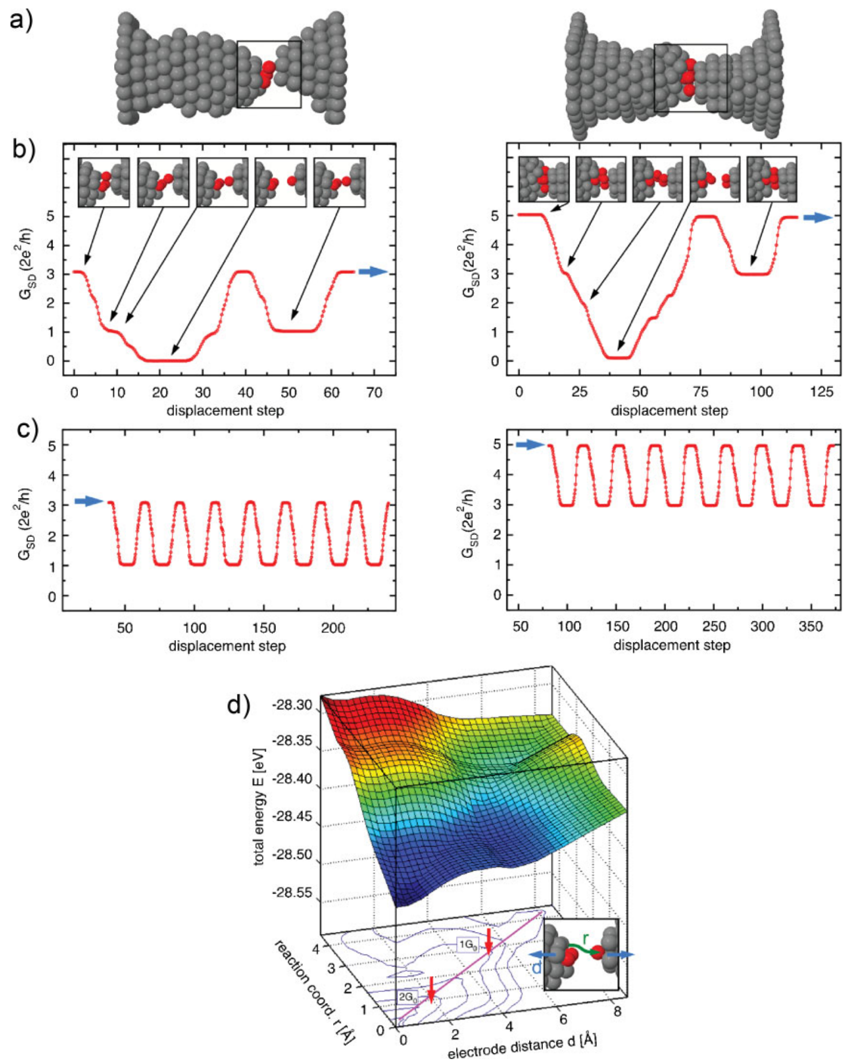

Figure 3. Computer simulations of multilevel switching conformations switching between 1 G_0 and 3 G_0 (left) and 3 G_0 and 5 G_0 (right): a) Initial nanojunction. The bridging silver atoms of the minimal cross sections are marked in red. b) Conductance during two switching cycles with the corresponding tip geometries shown as insets. c) Subsequent switching cycles follow the sequences shown in (b), demonstrating repeated interlevel switching. The simulation verifies the reproducible bistability of the silver contacts, in perfect agreement with the experimental observations. d) Relation between structure, total energy and conductance of a multilevel point contact: (top) potential energy surface as a function of the electrode distance d and the reaction coordinate *r* (inset), (bottom) independently computed contour plot of the conductance projected to the bottom of the diagram. The regions indicated by red arrows on the bottom surface indicate parallel valleys on the energy and conductance surfaces. As long as the switching process alternates between points on these two valleys on the energy surface reproducible switching will be observed.

The calculation of conductance for each electrode displacement step is shown in Figure 3b for these geometries. The leftmost conductance minima (at step 18 for the left junction, at step 37 for the right junction) are related to a complete rupture of the contact yielding 0 G_0 for both conformations. In this case the amplitude of the electrode displacement is 8.55 Å (11.4 Å) for the left (right) silver contact geometry. Using approximate experimental values, we decrease the electrode displacement to 2.85 Å (8.30 Å) for the left (right) electrode in subsequent switching cycles. The reduction of the displacement amplitude results in long term reproducible, bistable switching between conductance levels of 1 G_0 and 3 G_0 (left contact) and of 3 G_0 and 5 G_0 (right contact).

Close inspection of the intermediate geometries of the junction explains this surprising result, which does not occur for every junction: Some bistable junctions exhibit conductance plateaus, which are characterized not by one, but by a whole ensemble of structurally related conformations. Detailed analysis of this ensemble reveals the mechanism of the multilevel switching which was observed in the experiments described above: For the junction on the left of Figure 3a, a single silver atom rolls over a finite displacement range over the two other bridging atoms to its left (see insets of Fig. 3b, left) before finally disconnecting. Choosing the correct displacement amplitude, this induces multilevel switching, because not only the terminal geometries, but also the formation/dissociation pathways are conserved in the switching process. When we repeat the switching cycle for multiple times for both junctions (Fig. 3c) we find a lock in effect with a conductance variation below 0.11 G_0. This lock in effect is also in agreement with our experimental observations.

To explain the correlation between atomic structure, energy and conductance we calculate the potential energy surface (PES) and the zero bias conductance for junction geometries generated by independently varying the electrode electrode distance d and a reaction coordinate r (see inset in Fig. 3d). This reaction coordinate of one specific bridging atom is chosen to correlate linearly with d along the observed reaction path ($r = 0.56$ d). The inset shows a schematic presentation of the path of the bridging silver atom during the opening process characterized by the reaction coordinate r. We find the global energy minimum at the closed and unperturbed state of the contact (Fig. 3d). We find parallel valleys on both the conductance and energy surface, which explain the stability of the conductance plateaus at 1 G_0 and 2 G_0 during the switching process.

To conclude, we have demonstrated multilevel metallic quantum point contact transistors. These devices can be switched reversibly between different quantized (non zero) conductance levels by fabrication of bi and multistable configurations on the atomic scale. The devices reproducibly operate at room temperature, the source drain conductance being switched by a control voltage applied to an independent, third gate electrode. Due to the combined electrochemical fabrication and annealing process, well ordered, bistable contact configurations are achieved, exhibiting conductance quantization at integer multi ples of the conductance quantum G_0. Computer simulations allow a detailed understanding of the multilevel switching process based on bi and multistable atomic scale reconstruction of the contact configuration. The results allow to configure model systems for studying conductance quantization and atomic scale reconfigurations in predefined, switchable systems. At the same time they open intriguing perspectives for multilevel logics and quantum electronics at the atomic scale.

Experimental

Simulation Methods: The start conformation of the deposition simula tions consists of two parallel Ag(111) surfaces 38.8 Å apart, with a Ag Ag distance of 2.88 Å. A Metropolis Monte Carlo method is used to deposit individual silver ions under the influence of the electrostatic field, calculated by solving the Poisson equation for the source charges of the present electrode conformation and a material specific Gupta potential. A deposition simulation for a single ion comprises 10000 steps of a random spatial displacement drawn from a uniform distribution of at most 0.1 Å in each direction. The deposition algorithm is stopped, if a predefined number of non overlapping pathways connect the left and right electrode.

During the opening/closing process the silver clusters are displaced in steps of 0.15 Å. For each cluster displacement we perform 10000 simulated annealing steps, where the fictitious temperature is reduced from 300 K to 3 K. The initial switching cycle is considered successful, if all atoms return to their original positions to within 0.28 Å.

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