Origin of yield problems of single electron devices based on evaporated granular films

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An evaporated nanometer scale granular film provides a simple system for studying Coulomb blockade effects. This technique has often been used during the last few decades. However with respect to potential devices, specific problems continue to obstruct broader application. It is virtually impossible to observe Coulomb blockade in one–dimensional structures, and even for wide two–dimensional systems the yield is frustratingly low. We study these problems using a comprehensive theoretical framework that enables us to model both the growth aspects, and the electrical characteristics. In particular, we study how the morphology of the islands influences their electrical properties. Explanations for the observed behavior are put forward. © *1999 American Institute of Physics.* [S0003-6951(99)02137-3]

The electronic conduction of nanometer scale granular films has been studied for a long time.[1] In fact, discussion of Coulomb blockade[2] started with these systems.[3] It was soon realized that the basic requirement of the observation of the Coulomb blockade phenomenon is a double junction system of sufficiently high resistances ($R \gg h/e^2$) with an island of sufficiently low capacitance [$C \ll e^2/(k_B T)$] in between. Until the development of two-angle shadow evaporation[4,5] granular systems dominated this field. More recently, however, interest has turned from exploration to application, and from stochastic granular structures to lithographically defined devices. This includes a combination of both, such as lithographically defined one-dimensional (1D) granular films[6] and the postprocessing of nanometer size clusters with high accuracy.[7]

One reason for this change of approach is the fabrication yield problem of granular structures. 1D systems of nanometer size clusters have been achieved,[6] but Coulomb blockade has not yet been observed in these systems. The situation is more optimistic for two-dimensional (2D) structures, but low yield poses a problem here as well. Postprocessing offers a solution, but, unfortunately, to date it is not feasible for large scale applications.

In this letter we study yield problems from a simulation point of view. We concentrate on two aspects: namely the lack of Coulomb blockade in 1D films and the growth conditions in 2D systems. For simplicity, we restrict this consideration to the case of gold films on a silicon dioxide substrate. However, the framework used is generic and is able to cover different material systems if the parameters are adapted accordingly.

A computer simulation package has been developed and is used to model all relevant aspects of granular structures, from the actual growth of the samples to the computation of the capacitances and tunneling resistances between the grains and, finally, the full current–voltage ($I-V$) curves. Electrical characterization can be performed while the growth process takes place thus giving *in situ* control of the deposition.

The simulation package consists of two fundamental parts: the simulation of the deposition, island formation, and island growth, and the program used to calculate the $I-V$ characteristics of granular devices of given geometry. Both parts employ Monte Carlo algorithms, however in very different contexts.

As for the first aspect, the growth process is simulated with a deposition–diffusion algorithm.[8] It includes the atomic deposition at random positions, 2D diffusion on the substrate, island formation by congregation of free atoms, the growth of existing islands, and the coalescence of islands. The three-dimensional nature of the films is accounted for by assuming a hemispherical shape of all the islands. The presence of metallic contacts prior to the deposition can also be accounted for.

The growth model has proved to reproduce well the morphology of the experimental structures.[9,10] Additional processes such as diffusion of deposited atoms into the substrate and (subsequent) Ostwald ripening of islands turned out to be not relevant and therefore they are neglected. Moreover, surface tension of the forming islands is found to be considerable,[9] thus favoring the gold dimer as a stable ar-

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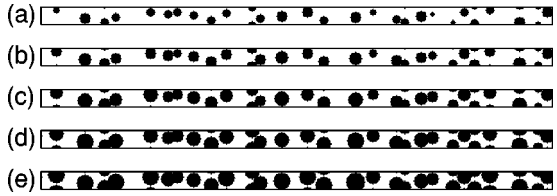

FIG. 1. Snapshots of a 1D granular film during deposition for coverages: (a) 20%, (b) 30%, (c) 40%, (d) 50%, and (e) 60%. Note that periodic boundary conditions apply in the vertical direction.

rangement of two gold atoms, leading to hemispherical shape of larger islands, and preventing reevaporation of atoms once they are captured by an island as well as migration of whole islands.

The amorphous silicon oxide surface is simulated by a square lattice of lattice constant 2.57 Å for the diffusion process.[11] Another parameter that influences the growth process is the diffusion/deposition ratio, i.e., the number of diffusion steps that one atom is allowed to perform on the grid before a new atom is deposited. It influences the size distribution of the generated film. In accordance with experimental results[9] we use 1.8×10^9 for this parameter.

The outcome from the growth simulation is the position and size of the individual islands. These purely geometric data are forwarded to the electrical characterization. This second part of the simulation is described extensively elsewhere.[12] Eventually, it produces the $I-V$ characteristics according to the ''orthodox theory''[13] from the geometric input data.

We have applied the modeling procedure both to 1D and 2D arrays. Both these types of structures can be fabricated experimentally.[6] As for the 1D systems, a series of snapshots at different surface coverages of the growth process are shown in Figs. 1(a)–1(e). Notice that most of the islands are formed within the very early stages of the growth process after which the growth is dominated by the enlargement of the existing islands rather than by the formation of new ones.

In Fig. 2 we evaluate the growth data of Fig. 1 with respect to the interdot distance d. This is a very sensitive quantity since tunneling has been observed via gaps less than 2 nm only, even under favorable, tunnel-barrier-height reducing dielectric films.[9,10] In Fig. 2 the probability density is shown for different values of the substrate coverage. Negative d indicates coalesced islands. Remarkably, the width of the distribution hardly changes and the long tail towards

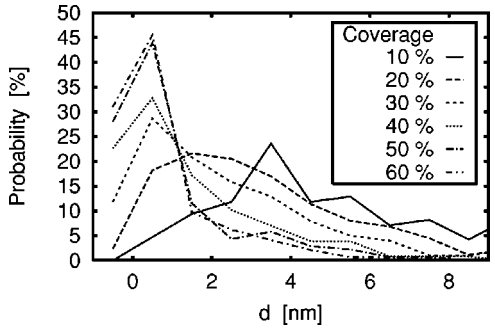

FIG. 2. Change of the interdot separation distribution with increasing coverage for a 1D system. The histograms are shown for coverage from 10% to 60%. Negative d means coalesced islands.

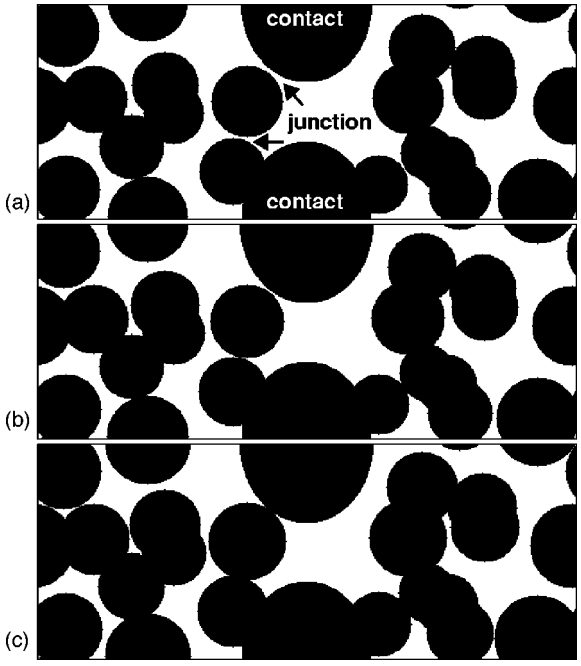

FIG. 3. Snapshots of a 2D granular film during deposition for coverages: (a) 60%, (b) 64%, (c) 70%. The evolution of a double junction is visible in the vicinity of the lead contacts. Note that periodic boundary conditions apply in the vertical direction.

large d results in a surprisingly high probability of finding wide tunneling gaps. At 60% coverage, for instance, where coalescence is predominant, the probability of finding a prohibitive tunneling gap of 3 nm or more is still 10%. It follows that 1D chains containing more than ten islands have an extremely low chance of being conducting, and correspondingly even lower chance of showing the particular condition for Coulomb blockade. This corresponds well to experimental findings.[6] Therefore, while the fabrication of 1D granular systems remains an impressive achievement in technological terms, there is doubt that such systems will be feasible for making single electron devices.

Our analysis has also been extended to 2D systems. In this case, two electrical contacts are placed on the grid prior to the deposition process. The growth process is altered in the vicinity of the contacts. In particular, the contacts act as sinks for the deposited atoms, and the region around the contacts remains depleted of islands. This feature is apparent in experiments.[14] In order to obtain a Coulomb blockade system, it is necessary that a tunneling path between the two contacts is established. This might involve several islands but it is necessary that all the inter-island separations are small enough so that tunneling can occur. In practice this means that the islands must be separated by less than 2 nm.[6]

We have performed numerous simulations of the growth process and we have found that the conditions for Coulomb blockade devices are relatively unlikely to occur. The yield is about 10%. In Figs. 3(a)–3(c) we show a system that complies with the above requirements. The various figures are snapshots taken during the growth process at coverages ranging from 60% to 70% of the surface. As can be seen in this case there is an island [Fig. 3(a)] that is placed in between the contacts and separated by a small distance from the electrodes. In this case tunneling from the contacts to the

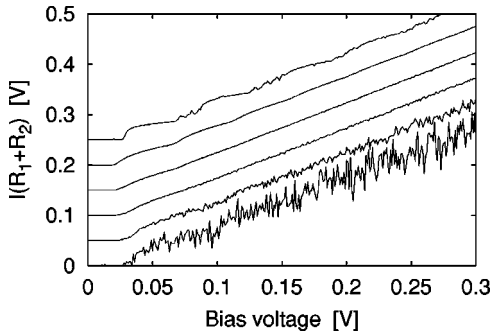

FIG. 4. $I-V$ characteristics of the 2D system shown in Fig. 3 for coverages from 60% (bottom) to 70% (top). The curves are scaled and offset by 50 mV per curve for comparison and clarity.

island is allowed, the island becomes chargeable, and Coulomb blockade effects can be observed.

The associated $I-V$ characteristics are shown in Fig. 4. The curves are scaled and offset for comparison and clarity, respectively. The high noise level of the low current curves is a result of the Monte Carlo simulation. A clear Coulomb gap is visible in all curves. This gap is accompanied by a Coulomb staircase. As is well known from the ''orthodox theory,'' [13] a Coulomb staircase occurs only when the tunnel junctions are asymmetric. Due to inherent lack of control over the island position during the deposition process, most systems containing isolated islands display this asymmetry. The probability of finding a symmetric system is very low in comparison. This is a situation encountered in numerous experiments.

In conclusion, we have comprehensively studied the simulation of granular metallic films. This simulation allows us to model both the growth process and the electrical characteristics of these structures. The latter can be evaluated *in situ*.

We have applied the procedure both to 1D granular chains and to 2D granular films. An explanation of the absence of Coulomb blockade in 1D structures has been proposed. In 2D systems it has been found that the probability of obtaining a device operating in the Coulomb blockade regime is rather low, about 10%. This is due to the fact that a tunneling path between the electrodes must be available with all the islands separated by a distance of no more than 2 nm. Even lower is the probability of finding a symmetric double junction, displaying Coulomb blockade, but no Coulomb staircase. We attribute to this the fact that the staircase is widely observed as a signature of charging effects in experimental double junctions.

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[1] W. F. G. Swann, Philos. Mag. **28**, 467 (1914).
[2] H. Grabert and M. H. Devoret, *Single Charge Tunneling: Coulomb Blockade Phenomena in Nanostructures*, NATO ASI Series B: Physics, Vol. 294 (Plenum, New York, 1992).
[3] C. J. Gorter, Physica (Amsterdam) **17**, 777 (1951).
[4] J. Niemeyer, PTB Mitt. **84**, 251 (1974).
[5] T. A. Fulton and G. J. Dolan, Phys. Rev. Lett. **59**, 109 (1987).
[6] A. S. Cordan, A. Goltzené, Y. Hervé, M. Mejias, C. Vieu, and H. Launois, J. Appl. Phys. **84**, 3756 (1998).
[7] T. Junno, S.-B. Carlsson, H. Xu, L. Montelius, and L. Samuelson, Appl. Phys. Lett. **72**, 548 (1998).
[8] M. Boero, P. A. Mulheran, and J. C. Inkson, Microelectron. Eng. **42**, 515 (1998).
[9] M. Meijas, PhD thesis, Universite Louis Pasteur Strasbourg, 1998 (in French).
[10] M. Meijas, C. Lebreton, C. Vieu, A. Pepin, F. Carcenac, H. Launois, and M. Boero, Microelectron. Eng. **42**, 563 (1998).
[11] C. Vieu (private communication).
[12] H.-O. Müller, K. Katayama, and H. Mizuta, J. Appl. Phys. **84**, 5603 (1998).
[13] D. V. Averin and K. K. Likharev, in *Mesoscopic Phenomena in Solids, Modern Problems in Condensed Matter Sciences (30)*, edited by B. L. Altshuler, P. A. Lee, and R. A. Webb (Elsevier, Amsterdam, 1991), pp. 173–271.
[14] B. Abeles, P. Sheng, M. D. Coutts, and Y. Arie, Adv. Phys. **24**, 407 (1975).