Features of Polycrystalline Electron Transport of Colloidal Quantum Dots InSb and PbS

Nikolay D. Zhukov, Ildar T. Yagudin

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Authors' credentials:
Nikolay D. Zhukov Candidate of Physic and Mathematic, Director of the Enterprise Ref-Volga-Svet , LLC, Saratov, RF
(ndzhukov@rambler.ru)

Ildar T. Yagudin, Research Scientist, Scientific Production Enterprise "Volga", LLC, Saratov, RF
ORCID 0000-0001-9013-3832
(invoker9000@gmail.com)

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Abstract: On samples, in the multigranular layer structure of colloidal quantum dots (QDs) in narrow-gap InSb and PbS semiconductors, electrons are transported in intergranular nanogaps during current flow by thermal emission from shell-free QDs, or by tunnel emission from core-shell QDs. Electrons are injected into QDs by tunnel emission and *via* their passage through the shell. For shell-free QDs, the current is limited by space charge, similar to the Coulomb blockade.

Keywords: semiconductor (SC), quantum dots (QDs), polycrystalline structure (PCS), nanoparticles, atomic force microscope (AFM), current-voltage characteristics (CVC).

Introduction

Investigating the physics of processes in a polycrystalline structure (PCS) relates to the field of disordered structures, such as, for example, semiconductor (SC) polycrystals. Numerous published sources on polycrystalline materials classify them as densely packed structures with nearly zero gaps between crystallite grains [1]. For those, physical models of the contact phenomena are employed. The specific features of the PCS under our consideration are relatively large gaps between the grains, at which electrons experience emission from the grain into the gap and injection from the gap into the grain. We studied these processes on separate grains of the substrate surface using the method of scanning tunneling microscopy [2, 3].

In our study [4], the mechanisms of the current in the PCS nanoparticles of the most promising semiconductors (Si, GaAs, InAs, InSb) were investigated. A detailed analysis of the current-voltage characteristics enabled us to establish that their behavior is determined by the mechanism of intergranular tunnel emission from the near-surface electronic states of submicron particles. The parameters of the emission process were identified, and the formula for the dependence of current I on voltage U was obtained.

PCS, based on quantum dots (QDs) of narrow-gap semiconductors, for which intra- and intergranular quantum size effects can occur, imposing a heavy impact on electron transport, are of particular interest [2].

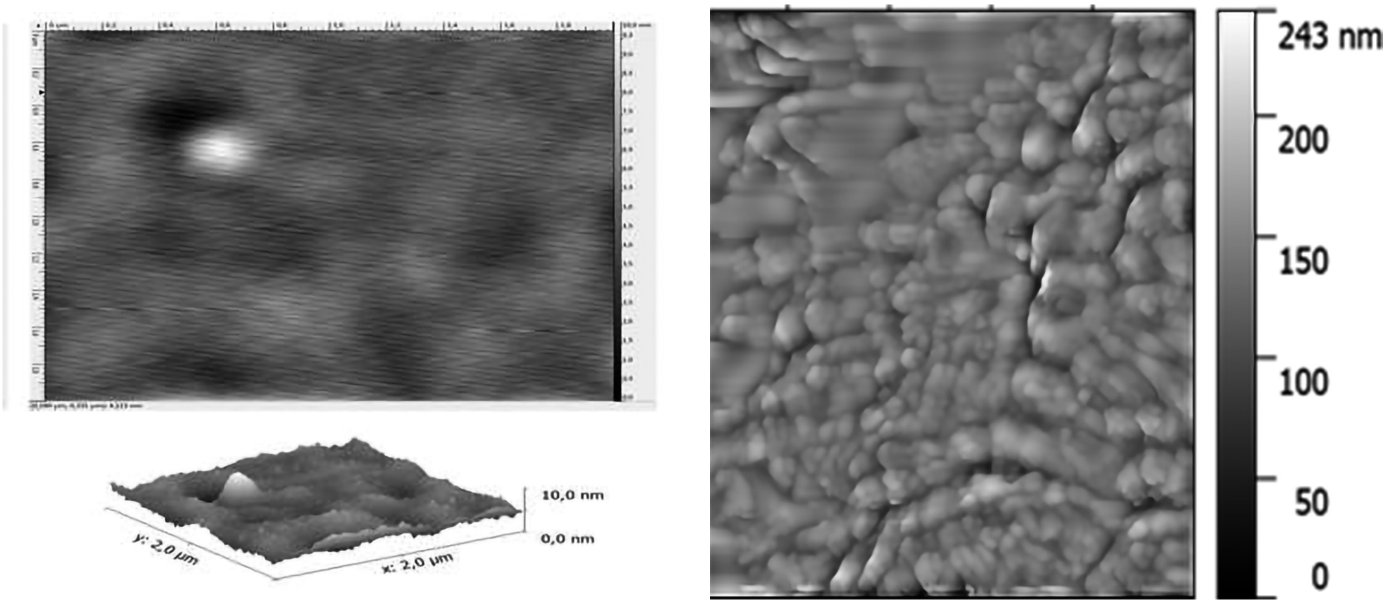


Figure 1. 3D-AFM- display of surface fragments in the PbS polycrystalline layer

Materials and Methods

In our study, colloidal quantum dots were investigated: both shell-free QDs (InSb) and core-shell QDs (InSb/CdS, InSb/InP, PbS/CdS).

PCS samples were prepared by QD deposition from alcohol-based solutions, followed by deposition on a glass substrate with comb aluminum film electrodes *via* self-organization of ensembles on the substrate surface under controlled evaporation of the solvent [5]. Figure 1 presents a typical fragmented image of the layer surface obtained with a 3D atomic force microscope (AFM). The coatings had a layered structure of tightly packed clusters, with irregularities between those not exceeding 10 nm, while at some places, they experienced the mutual tight contact.

The study of the current mechanisms was carried out on the basis of measuring the current-voltage characteristics (CVC). For measuring CVC of the PCS, we used a non-standard generator of unipolar low-frequency pulse sawtooth voltage up to 300 V, pulse duration of 500 µs, and a duty cycle of 128. The parameters of the circuit elements and the regimens were selected, taking into account the absence of impact of the reactive components on the CVC. The transients were monitored by oscillograms. The methodology was described in detail in our earlier publication [4]. Studies of the CVC of individual nanoparticles were performed on a STM SOLVER NANO scanning tunneling microscope by multiple repeated measurements and their averaging. Experimental CVC were processed using Microsoft Excel, and only those, for which an analytical approximation of the curves has given maximum values of reliability coefficients of 0.99 and above, were used for the analysis.

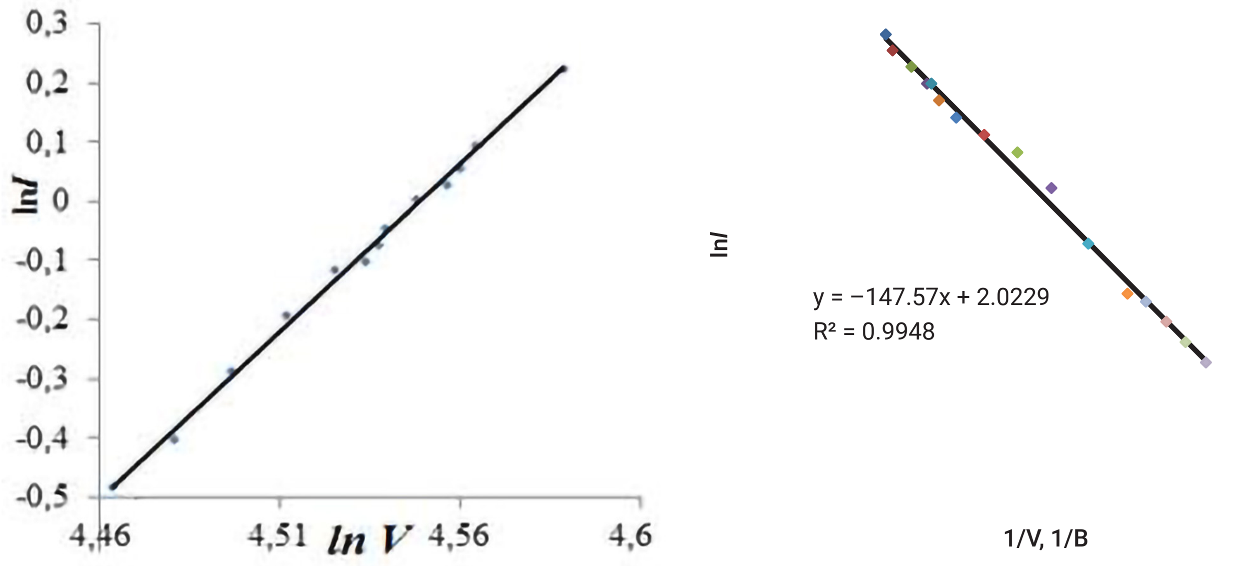
Results

To explain the obtained results and to select the electron transport model, we carried out the STM study of individual nanoparticles on a conductive substrate.

Analysis of all CVC (current I as a function of voltage V) and the choice of models were carried out on the basis of conventional knowledge on electron transport through the heterointerface into a semiconductor: thermal emission model – $I \sim \exp[B_e V]$, tunnel emission model – $I \sim \exp[B_t/V]$, and current-limited-by-space-charge model $I \sim V^C$ ($2 < C < 3$).

Figures 2 and 3 show, as examples, typical CVC of the PCS and STM in characteristic coordinates.

Table summarizes the main measurement results. Approximately 20 measurements were made on each sample. Random sets were taken from about five measurements, the average approximation for each parameter was computed. The maximum deviations from the mean were ± (10-15) %.



a b

Figure 2. Current-voltage characteristics of PCS: a – InSb, **b** – InSb/InP

TABLE.
Data Summary of the Measurement Results of the Current-Voltage Characteristics

CVC parameters	STM, minus on conductive substrate			STM, plus on conductive substrate			PCS		PCS PbS
	InSb	InSb/CdS	InSb/InP	InSb	InSb/CdS	InSb/InP	InSb	InSb/InP	
B_e	3.43		15.75						1.2
B_t	2.23	−6.98	−6.63		−4.72	−3.3/−3.9*		−140	−9.1
C				2.67			4.9		2.25

From the obtained data, it is possible to build a model for the intergranular transport of the electrons.

As the Table data imply, during emission from quantum dots (-V on the sample), for QDs-InSb, two sections of the CVC were observed: in the low-voltage region (up to 1.2 V) – thermal emission with B_e≈3.43; whereas in the high-voltage region (above 1.2 V) – tunnel emission with the parameter B_t~2.23. For QDs-InSb/CdS and InSb/InP, the tunnel emission mechanism prevailed at B_t~7. The parameter ratio equal approximately to 3, is explained by the fact that the decisive value in the formula for the parameter B_t is the square root of the effective mass [2]. The ratio in this case is approximately 3 (m_{InSb}~0.015m_0; m_{CdS}~0.2 m_0; m_{InP}~0.1m_0; *www.xumuk.ru*). Such result may suggest that emission occurs from the shell layer, rather than from the QD core.

During emission from the probe (+V on the sample), the CVC for QDs-InSb/CdS and InSb/InP is determined by tunneling and the transit of electrons through the shell into the QD core. The *Bt* parameter in this case has a value intermediate between the core (InSb) and the shell (CdS and InP). For the QDs-InSb/InP sample, at voltages above 1.5 V, the Fowler-Nordheim dependence was observed (in the Table, the parameter is marked '3.9*'); that is, the CVC was determined by the emission from the probe. This may indicate a facilitated nature of tunneling through the InP shell, which has better size quantization properties than the CdS shell.

For QDs-InSb, a characteristic power dependence was observed, by which we could assume the presence of the space-charge limited current (SCLC) *sensu* Coulomb of QDs (Table, *C*~2.67). This effect is

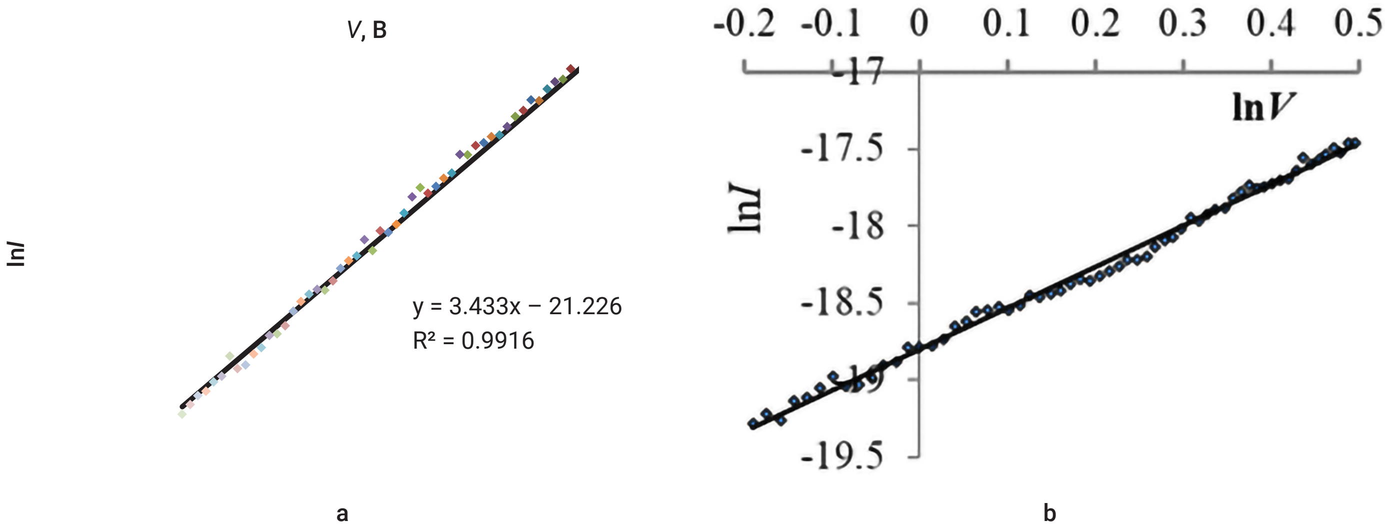


Figure 3. Current-voltage characteristics of STM emission from QD-InSb (a) and from the probe (b)

similar to the Coulomb blockade and is a direct proof of the quantum dimension of nanoparticles.

The CVC of the PCS for QDs-InSb imply the presence of SCLC, but with a higher value of the parameter C (Table, C~4.9). The CVC for QDs-InSb/InP had a clearly tunneling nature with a large value of the parameter B_t~140. Based on the model of the current flow through sequentially located QDs with 'jumps' through many intergranular nanogaps [4], and using the B_t parameter ratio of (−140:2.23), it can be assumed that, in the electric current lines, there are on average approximately 50-60 gaps, that is, the layer consists of about 100-200 QD (1–2 μm).

Conclusion

Hence, the structure of the polycrystalline layer of quantum dots for the flow of the current and electron transport has intergranular nanogaps, in which the electrons, when emitted from a semiconductor, penetrate the barriers by means of the thermal emission method for shell-free QDs, or by the tunnel emission method for core-shell QDs.

When a quantum dot is injected into a semiconductor, electrons undergo the tunnel emission, along with the transit through the shell, and space charge limitation similar to the Coulomb blockade.

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