

Nanostructures with Quantum Dot Clusters: Long Photocarrier Lifetime

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Major restrictions of semiconductor optoelectronic devices operating at room temperatures are caused by short photoelectron lifetime, which strongly reduces the photoresponse and puts strong limitations on manipulations with photoelectrons. Here we present results of modeling novel optoelectronic materials, based on structures with correlated dot clusters. The main distinctive characteristic of these quantum-dot structures are collective potential barriers around dot clusters. The barriers provide an effective control of photoelectron capture due to separation of highly mobile electron states transferring the photocurrent from the localized electron states in quantum dots. The novel nanostructured materials combine manageable photoelectron lifetime, high mobility, and quantum tuning of localized and conducting states. Thus, these structures have strong potential to overcome the limitations of traditional quantum dot and quantum-well structures. Besides manageable photoelectron kinetics, the advanced quantum-dot structures will also provide high coupling to radiation, low generation-recombination noise, and high scalability.

Keywords: Photodetectors, Carrier Lifetime, Quantum Dot Clusters, Photoconductive Gain.

1. INTRODUCTION

Many optoelectronic devices are based on the phenomenon of photoconductivity in which a material becomes more electrically conductive due to photocarriers created by electromagnetic radiation. Photocarriers contribute to the electric current until they recombine or are trapped by impurities and defects. Long photocarrier lifetime would substantially improve operation of semiconductor devices, such as electromagnetic sensors and solar cells. New nanostructured materials that provide long photocarrier lifetime at room temperatures would significantly increase the commercial market for infrared and terahertz detectors and find numerous applications in public safety, industry, and healthcare.

Quantum-dot (QD) nanostructures were considered as a promising candidate for improving the room-temperature optoelectronic devices due to expected controllable intradot kinetics related to electron transition between discrete QD levels. These expectations were based on the “phonon bottleneck” concept, which assumes that the phonon-assisted bound-to-bound transitions are prohibited, unless the energy between two discrete levels matches to the phonon energy.¹ According to this concept, the intrinsic

electron relaxation in quasi-1D nano-objects, such as QDs, was anticipated to be significantly slower than in 2D and 3D structures. However, the phonon bottleneck model completely ignores modification of electron states due to interaction effects, e.g., due to a finite width of electron energy levels. It is not surprising that the experimentally measured phonon-mediated electron relaxation turned out to be much faster than it is expected in the phonon bottleneck concept.^{2–4} Recent investigations⁵ unambiguously demonstrated that the actual intra-dot kinetics is completely opposite to what can be expected for weakly interacting electrons and phonons. In reality, strong coupling between electrons and longitudinal optical (LO) phonons leads to formation of the polaron states, which decay due to the interaction of LO phonons with acoustical phonons. Such kinetics results in strong energy and temperature dependences of the electron relaxation. At helium temperature, long relaxation time (~ 1.5 ns) was observed for the level separation of 14 meV (3.4 THz).⁵ However, the relaxation time decreases to ~ 2 ps for the 30 meV transition. The relaxation time also drastically decreases, if temperature increases. For example, for 14 meV transition, the relaxation time was measured to reduce from 1.5 ns at 10 K to 560 ps at 30 K, and further to 260 ps at 50 K. At room temperatures the polaron decay time is observed in the range of 2–30 ps, depending on the electron energy.⁵ Thus, after

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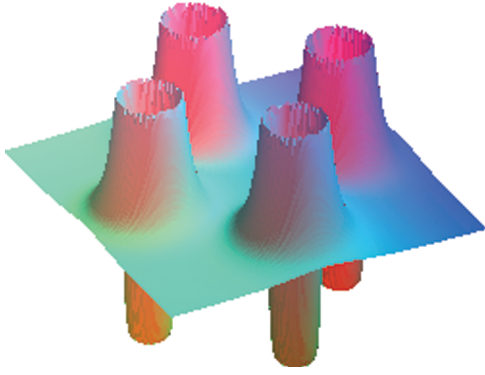


Fig. 1. Quantum-dot structure with local potential barriers around dots.

numerous experiments with various QD structures, no true phonon bottleneck has been found. Thus, the intra-dot electron relaxation at room temperatures turns out to be very fast and practically unmanageable.

Recent advances in nanotechnology lead to new fascinating possibilities for controlling inter-dot kinetics of photoelectrons by means of potential barriers in specially engineered QD structures.⁶ Potential barriers are always created, when electrons populating the dots are taken from the specific areas located relatively far from the dots. For example, Figure 1 shows local potential barriers around single dots. To effectively suppress electron capture process, the barrier height should be at least two–three times larger than kT . Therefore, at room temperatures, the local barriers should be ~ 0.1 eV and, as simple evaluations show, quantum dots should comprise at least ten electrons. This requires relatively large dots and high level of doping. Collective potential barriers created by groups of dots (clusters, rows, planes) are more effective to separate localized states in these dots from the conducting channels. For example, Figure 2 shows potential barriers around dot cluster. In this work we investigate the photoelectron kinetics in structures with correlated dot clusters, which allows for creating significant potential barriers. Changing the electron occupation of quantum dots one can manage the potential barriers around dots and control the photoelectron capture processes. Manageable kinetics opens various

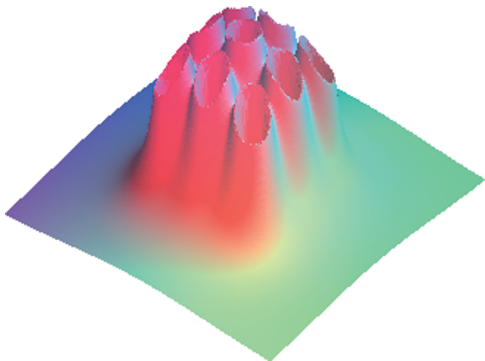


Fig. 2. The collective potential barriers around a cluster with 9 dots.

possibilities for developing adaptive sensors on the base of these structures.

2. PHOTOELECTRON KINETICS IN VCDC STRUCTURES

In recent publications^{7–10} we developed a model and numerical tools for modeling of photoelectron kinetics in quantum-dot structures with potential barriers. The developed model takes into account the main features of QD structures with barrier-limited capture at room temperatures. Employing Monte-Carlo method, we consider three basic scattering mechanisms, which includes electron scattering on acoustic, polar optical, and intervalley phonons. Our modeling considers electrons which may populate in Γ –, L –, and X – valleys and takes into account redistribution of carriers between valleys. The applied electric field provides strongly non-equilibrium electron distributions, but, at the same time, the increase of electron energy is not enough to initiate the avalanche processes or to modify significantly the potential barriers. We consider the carrier capture process as a specific inelastic scattering process, which is confined in the dot volume and in which a carrier is transferred from a conducting state above the potential barrier to a bound state below the barrier. We assume that from a bound state a carrier will relax to the deep quantum-dot states faster than it could return back to the conducting state.

In the vertically-correlated-dot-clusters (VCDC) structures presented in Figure 3 positions of the dot clusters are correlated in the vertical direction, which is the direction of photocurrent. The collective barriers around the dot clusters are formed by the carriers in the dots, and in this way create sufficient barriers compared to local barriers, which require a large number of carriers per dot. The barriers separate the localized electron states in quantum dots and the conducting channels in the matrix and therefore could suppress the electron capture into dots. Once the electrons are excited by the radiation, electrons drift in the

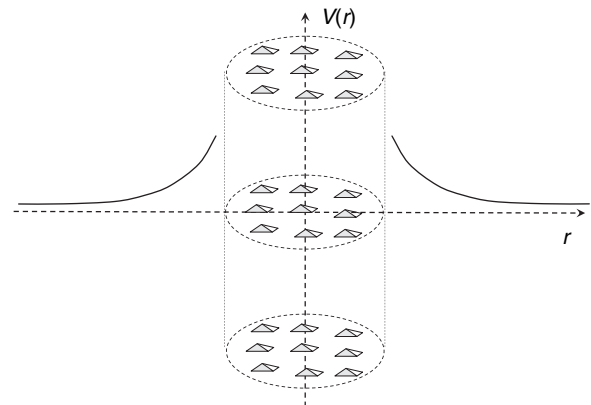


Fig. 3. The potential profile in the structure with vertically-correlated-dot-clusters (VCDC).

areas between dot clusters through high-mobility conducting channels, which are away from the photo-absorption areas. If the radius of the cluster, b , exceeds the distance between dot planes, c , the potential barrier around clusters has a logarithmic form $V_m = e^2 N n / (2 \pi \epsilon \epsilon_0 c) \ln(w/b)$, where N is the number of QDs in the cluster, n is the average occupation in the dot, and $2w$ is the distance between the centers of two neighboring clusters. In the following simulations we take $N = 9$, $b = 75$, and $c = 40$. The distance between the dots is about 55 nm. Since the barrier height is determined by the dot concentration and dot occupation, the capture time has an exponential dependence on these factors. Besides, the capture processes are weakly sensitive to geometrical parameters w and b due to logarithmic dependence.

In order to increase the absorption strength of the radiation, a large occupation in QDs is desirable, which could be determined by the doping concentration and applied bias. However, a large number of occupation is hard to implement, since it requires large quantum dots due to the discreteness of localized states in QDs. Figure 4(a) shows the exponential dependence of the capture time on the dot occupation n , which can be explained by the linear dependence of potential barrier height on the dot occupation. The charge in QD clusters provides large potential barriers around clusters, which effectively prevent the photoelectron capture into the dots and increase the capture time. Figure 4(b) presents the dependence of capture time on dot concentration. As seen, the capture time decreases with

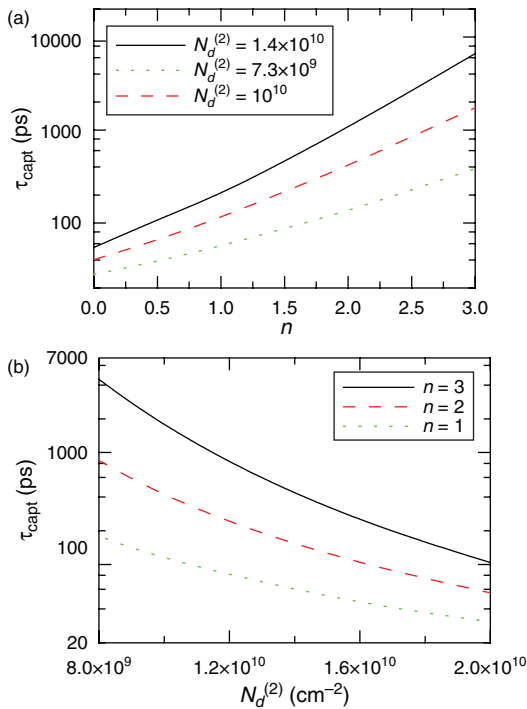


Fig. 4. (a) Capture time as a function of dot occupation, n , for three values of the QD concentration in a QD plane, $N_d^{(2)}$; (b) Capture time as a function of dot concentration, $N_d^{(2)}$.

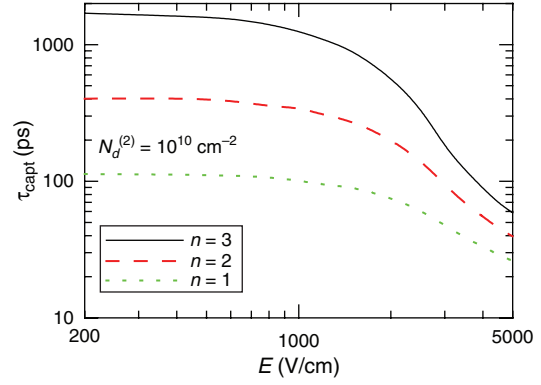


Fig. 5. Capture time as a function of electric field in VCDC structures.

increase of the dot concentration. This is because at fixed occupation, the potential barrier height increases slightly with the decreasing of dot concentration due to logarithmic dependence on w . In other words, smaller dot concentration, which means less trapped centres, provides less capture probability.

Usually a large applied bias is used to drive the photocurrent. Figure 5 represents the capture time as a function of electric field for three values of occupation. At small electric field, capture time is almost a constant, where the transit time decreases dramatically. When the electric field is up to a characteristic value, which is of the order of 10^3 V/cm, capture time starts to reduce significantly. This is because electron heating increases average energy of photoelectrons, which increases the probability of electrons to be captured.

Figure 6 presents the exponential dependence of the capture time on the inverse of average electron energy, which demonstrates that capture time is proportional to $\exp(1/\bar{\epsilon})$. Thus, the carrier capture in the electric field can be described by the equation from our previous publications,⁶⁻⁸ where the thermal energy kT is replaced by a factor of $\sim 2\bar{\epsilon}/3$. We may conclude that the effect of the electric field on carrier capture is well described by the model of electron heating. At low electric fields, electrons

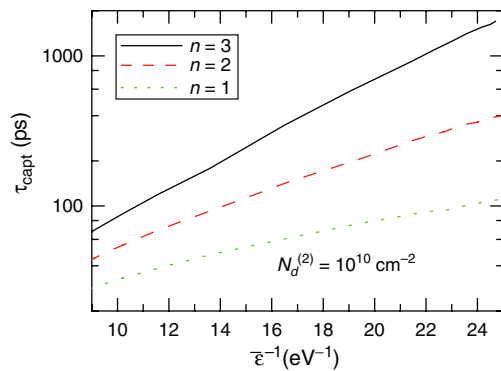


Fig. 6. Capture time as a function of the inverse electron energy attained in the electric field in VCDC structures.

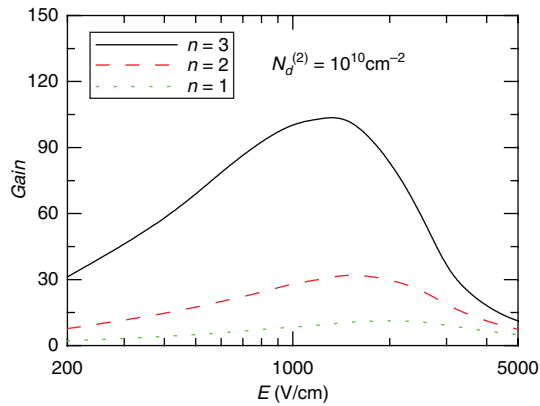


Fig. 7. Photoconductive gain as a function of electric field in a device with length of $1\ \mu\text{m}$.

are weakly heated. At higher electric fields, average electron energy becomes comparable with the potential barrier and therefore the electrons can be captured by quantum dots with much higher probability.

Photoconductive gain is defined as the ratio of the carrier lifetime to carrier transit time. In Figure 7 we present the photoconductive gain as a function of the electric field for a device with the length of $1\ \mu\text{m}$. As seen, the gain approaches a maximum value at electric field of the order of $10^3\ \text{V/cm}$, which is also the characteristic field for the dependences shown in Figure 5. This nonmonotonic dependence on the electric field may be explained in the following way. At small electric fields, where capture time is almost constant, the gain increases due to a decrease of the transit time. When the electric field increases up to a characteristic value, transit time almost saturates and capture time reduces substantially and, therefore, the gain decreases substantially.

3. SUMMARY

Using Monte-Carlo simulations, we investigated photoelectron kinetics in nanostructured materials with

vertically correlated dot clusters. We calculated the photoelectron capture time, responsivity, and photoconductive gain as functions of doping of a QD structure, its geometry, and electric field applied. Our main result is that the photoelectron kinetics is very sensitive to collective potential barriers around dot clusters and the capture processes can be substantially suppressed by a proper choice of the geometry of a QD structure and modulation doping. Detailed analysis shows that effects of the electric field on capture processes may be understood in terms of electron heating. The structures with vertically-correlated-dot-clusters have a strong potential for development of adaptive infrared photodetector with tunable operating time and sensitivity.

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