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Dependence of photosignal decay in colloidal HgTe quantum dot based photoresistors on preparation conditions

V. O. Yakovlev¹, T. V. Koroleva¹, K. T. Khakimov¹, O. A. Saptsova¹, A. A. Koronnov¹,
O. V. Vershinina¹, A. G. Medvedev¹, V. P. Ponomarenko^{1,2}, and V. S. Popov^{1,2*}

¹ Moscow Institute of Physics and Technology, Moscow Region, Dolgoprudny, 141701 Russia

² RD&P Center ORION, JSC, Moscow, 111538 Russia

* E-mail: popov.vs@mipt.ru

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This paper examines the influence of colloidal quantum dot synthesis conditions on the number and types of surface states, which in turn determine the response time of photosensors based on them. It is shown that replacing ethanedithiol with β -mercaptoethanol improves the response time of HgTe CQD-based photoresistors by three orders of magnitude.

Keywords: photosensor, colloidal quantum dots, CQDs, surface states, mercury telluride.

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Introduction

Photosensorics based on colloidal quantum dots (CQD) is one of the most dynamically developing areas of infrared photoelectronics [1, 2]. Photosensitive elements of resistive and barrier types based on colloidal quantum dots are used both for single photosensitive elements and the formation of matrix photoreceiving devices for the short- and medium-wave IR-ranges [3]. The sensitive layers in such photosensors are based on consolidated layers made by a liquid method from sols of semiconductor nanocrystals, mainly lead sulfide and mercury telluride. With this approach, the transfer of carriers in the resulting polycrystalline nanostructured layers proceeds primarily by a hopping mechanism. The surface of nanocrystals in layers formed by chemical methods contains a large number of different functional groups and uncompensated bonds, resulting in the appearance of surface states. The issues of response speed in photosensors based on CQDs play an important role due to the large surface area of quantum-confined materials and a more difficult-to-control environment surrounding the nanocrystals during the formation of consolidated films. The study of the effect of

surface conditions on the sensitivity and response speed of photosensors during the targeted modification of CQD surfaces and their layers is of great importance for the development of methods for the formation of photosensitive layers. The results of studies of the effect of traps presented in the literature are usually focused on the study of attenuation curves in photoresistors [4, 5], photoluminescence, [6, 7] or pump-probe spectroscopy [8] when varying ligands in PbS CQDs, at the same time, sufficient attention has not been paid to other CQDs, and the influence of the synthesis conditions of the nanocrystals themselves has not been considered in the literature.

The purpose of this work is to study the changes in surface states when varying the methods of CQD synthesis and post-synthetic treatment of the HgTe CQD surface by studying the decay curves of the photoelectric signal in photoresistors based on HgTe CQDs.

1. HgTe CQDs synthesis and the fabrication of layers based on them

Synthesis of HgTe CQDs

The HgTe CQD synthesis was carried out according to the published method [9] using the

complex compound $\text{NH}_4[\text{HgCl}_2(\text{SCN})]$ as a mercury precursor.

Photoresistive structures were manufactured in two stages. The first stage is substrate activation, and the second stage is the successive layer-by-layer formation of the photoresistive structure. Both stages involve using the same solution for activator and ligand exchange: a solution of ethanedithiol (EDT) in hydrochloric acid and isopropanol (HCl:EDT:IPA, 1:1:20 by vol.) or a β -mercaptoethanol (β -ME) solution in hydrochloric acid and isopropanol (HCl: β -ME:IPA, 1:1:20 by vol.).

Methods

Fourier-transform IR spectra were obtained using a Spectrum 100 FTIR spectrometer (Perkin-Elmer, USA) equipped with an attenuated total reflectance (ATR) accessory. High-resolution images were obtained using a JEM-2100 transmission electron microscope (JEOL, Japan).

2. Method for studying photoelectric signal decay curves

The study of photocurrent decay processes is one of the classic tools for studying surface states in photoconductive layers of semiconductors [9]. Depending on the type of trap and the energy level created by it, the trapped carriers return after some time to the valence band or conduction band.

Since the photocurrent is proportional to the number of charge carriers [10], the expression for the current decay over time is described by the following expression [11]:

$$I_{\Phi} = I_{\Phi 0} e^{-\frac{t}{\tau}},$$

where $I_{\Phi 0}$ is the photocurrent at saturation, and τ is the lifetime of the carrier.

The signal is divided by the amplitude of the photocurrent to analyze the multistage relaxation. The threshold is determined as follows:

$$I_{\text{thr}} = I_{\text{min}} + \gamma(I_{\text{max}} - I_{\text{min}}),$$

where I_{min} and I_{max} is the minimum and maximum values of the photocurrent in the selected interval.

Coefficient γ sets the relative position of the threshold level within the full dynamic range of the photovoltage and thus defines the boundary

between different stages of the relaxation process. In the method used, the value is selected in the following form γ :

$$\gamma = \frac{1}{e^n},$$

where e is the base of natural logarithms, and the exponent factor n is a whole or a fractional number set by the user depending on the expected number and extent of relaxation components. When $n = 1$, the threshold corresponds to the level $1/e$ of the full amplitude range, which is a characteristic value for single-exponential decay and is naturally associated with the determination of the time constant. As the exponent factor increases, the threshold value consistently shifts to the area of lower photocurrents, which allows the identification of slower relaxation stages.

This method of setting thresholds ensures a physically reasonable signal division into sections, each of which corresponds to a certain time range and, respectively, the dominant relaxation mechanism. As a result, each selected area can be analyzed independently using its own exponential approximation, which increases the accuracy of determining the time constants and amplitudes of individual components, while reducing the mutual effect of fast and slow processes when processing the whole signal together.

A running-median smoothing is used to reduce the noise impact and single outliers. At the same time, a combined mode is also possible in which the initial stage of the time series is preserved without filtration and the subsequent part is subject to smoothing. This allows one to preserve the initial changes in photovoltage and simultaneously increase the analysis stability of the slow relaxation area.

The approximation of the relaxation part of the photoresponse is carried out using a model of the sum of decaying exponential components. In general, the model is written as follows:

$$I_{\text{model}}(t) = I_{\tau} + \sum_{k=1}^N A_k \exp\left(-\frac{t - t_s}{\tau_k}\right),$$

Where I_d is the dark current, A_k is the amplitude of individual relaxation components, τ_k represents the time constants corresponding to them, N is the number of exponential summands and t_s is the

effective time shift of the relaxation beginning. At the same time, it is assumed that $A_k \geq 0$, which ensures a physically reasonable interpretation of the results.

The results of processing are the numerical values τ of sections of the photovoltage attenuation curve, as well as a set of time dependences including the initial signal and approximating curves.

3. Discussion of the results

In this work, HgTe colloidal quantum dots with an exciton absorption peak in the range from 2.13 to 3.16 μm were synthesized (Fig. 1).

According to the transmission electron microscopy data, HgTe nanocrystals were synthesized in all cases (Fig. 2). Photoresistor layers based on HgTe CQDs had a thickness of about 80–90 nm according to atomic-force microscopy data.

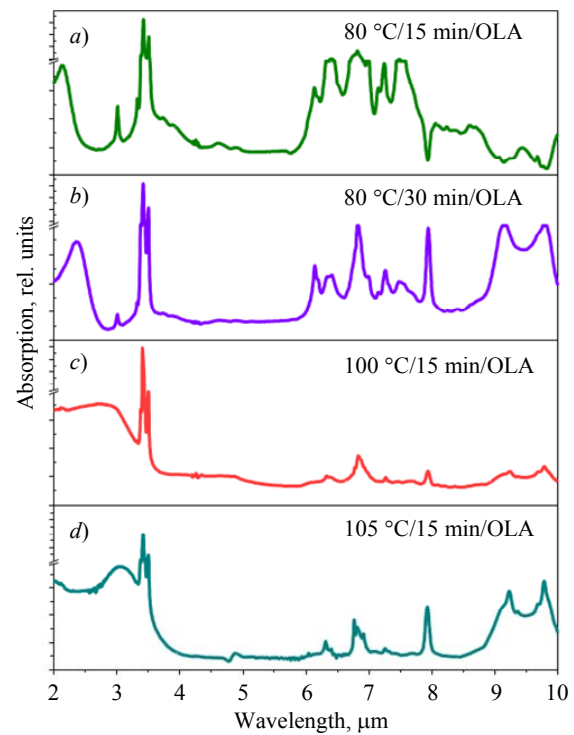


Fig. 1. Fourier-transform IR spectra of HgTe CQDs synthesized under different modes (OLA: oleylamine)

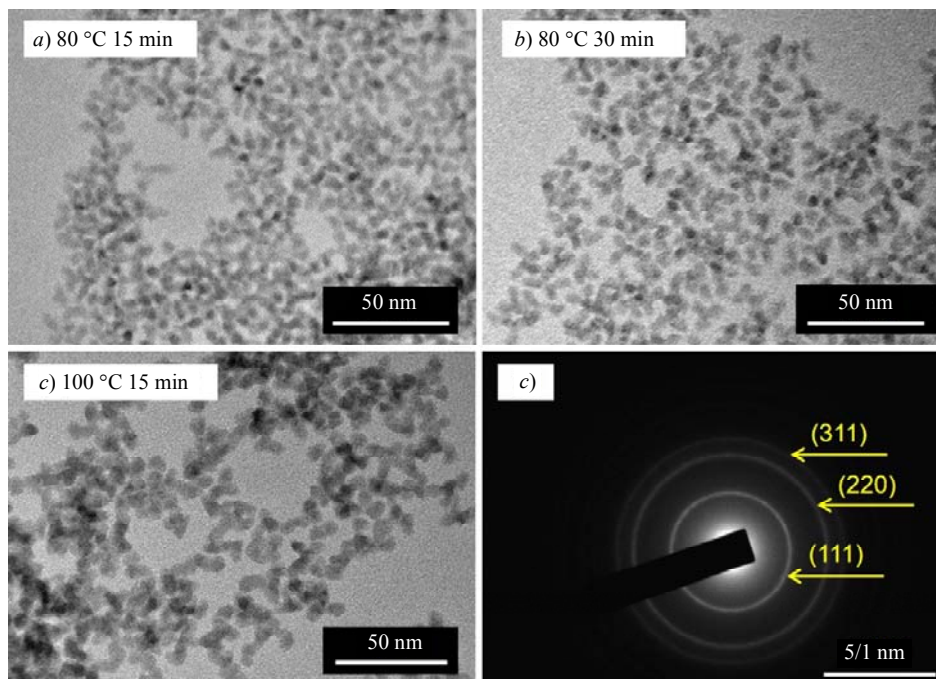


Fig. 2. (a–c) Transmission electron microscopy of HgTe CQDs obtained under different conditions; (d) selected-area electron diffraction pattern

The study of the decay curves showed that the number and type of surface states vary depending on the synthesis conditions of the CQDs and the type of ligand used for the surface modification of the photosensitive layers (Fig. 3).

Since the lifetime characterizes the energy of the surface state (namely, deeper traps correspond to longer lifetimes) [12], it can be concluded that depending on the quantum dot and the used ligand, surface states that are significantly different in terms of energy appear on its surface.

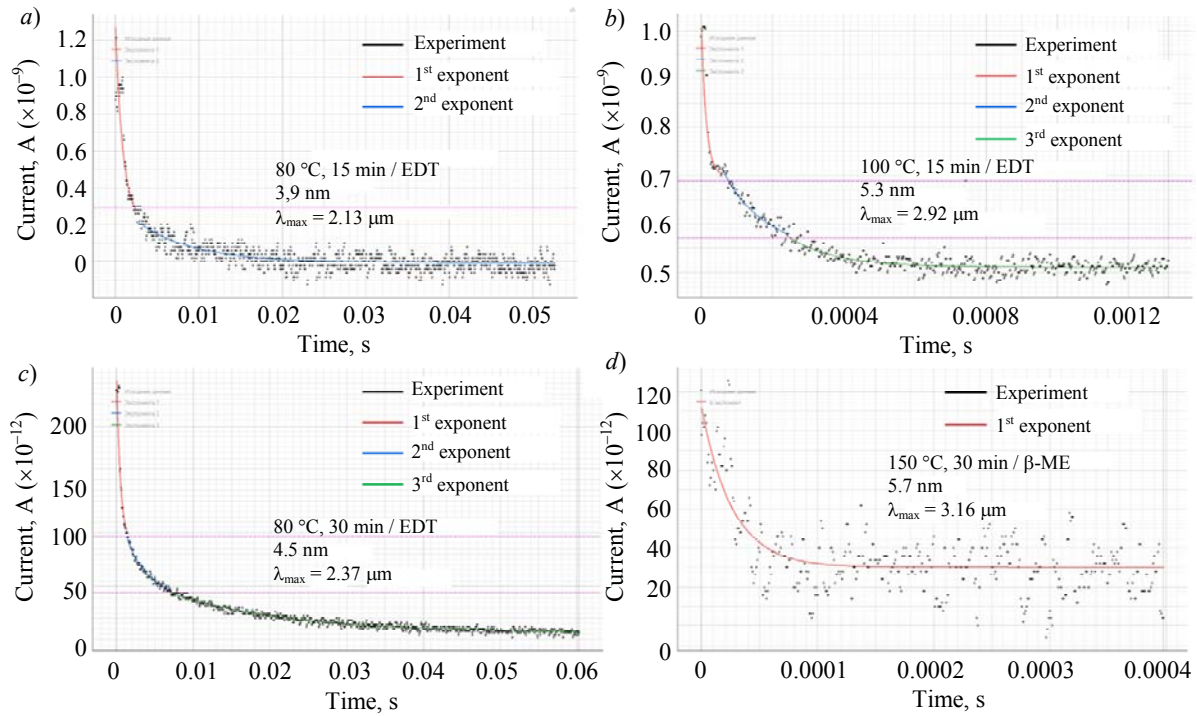


Fig. 3. Decay curves of the photosignals of photoresistors based on HgTe CQDs obtained under different conditions (temperature and synthesis time: a) 80 °C, 15 min, b) 100 °C, 15 min, c) 80 °C, 30 min) and using different ligands during surface modification: b) ethanedithiol (EDT) and d) – β -mercaptoethanol (-ME)

For HgTe CQDs with subsequent exchange by ethanedithiol, when the CQD size changes from 3.9 nm to 4.5 nm, the surface states are preserved, which are characterized by $\tau \sim 0.0002$ s and $\tau \sim 0.003$ s, while a new type of surface states is added with $\tau \sim 0.02$ s. When changing the CQD size from 4.5 nm to 5.3 nm, only those surface states are preserved that are characterized with $\tau \sim 0.02$ s, and simultaneously,

new surface states occur with $\tau \sim 0.08$ s and $\tau \sim 0.003$ s (Fig. 4).

The use of β -mercaptoethanol instead of ethanedithiol for ligand exchange at a close quantum dot size (5.3 and 5.7 nm) drastically alters the surface states: instead of three types with $\tau \sim 0.02$ s, $\tau \sim 0.08$ s and $\tau \sim 0.003$ s, only one type with $\tau \sim 0.00001$ s is observed (Fig. 4).

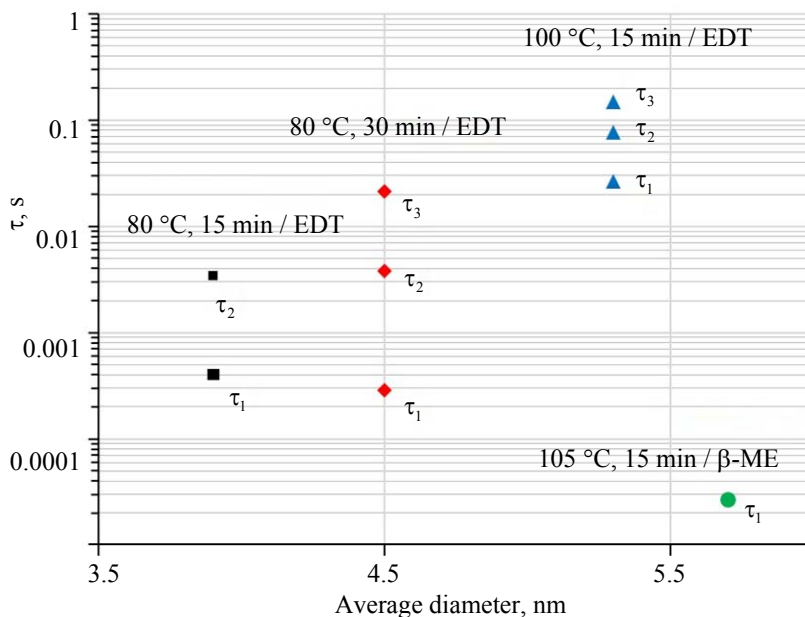


Fig. 4. τ values obtained during the processing of decay curves of the photoelectric signals of photoresistors based on HgTe CQDs obtained under different conditions (temperature and synthesis time: 80 °C, 15 min, 100 °C, 15 min, 80 °C, 30 min) and using different ligands during surface modification: ethanedithiol (EDT) and β -mercaptoethanol (-ME)

Conclusion

The analysis of the photoresponse decay of photoresistors based on HgTe CQDs, obtained while varying the synthesis parameters and the size of the nanocrystals, shows the existence of different types of surface states. The use of β -mercaptoethanol as a ligand exchange allows one to reduce the number of surface states from three types to one. This surface state is characterized by

the shortest lifetime found in this paper, equal to 10 μ s, thus ensuring a fast response speed of the photosensor.

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About authors

Yakovlev Viktor Olegovich, Junior Researcher, Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9). E-mail: iakovlev.vo@mipt.ru SPIN code: 3324-6931, AuthorID: 1290912, ORCID 0009-0009-3507-082X

Koroleva Taisiya Viktorovna, Junior Researcher, Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9). E-mail: koroleva.tv@mipt.ru SPIN code: 6991-6170, AuthorID: 1292280, ORCID 0000-0003-0792-1948

Khakimov Karim Timurovich, Junior Researcher, Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9). E-mail: khakimov.kt@mipt.ru SPIN code: 8812-6012, AuthorID: 1209525, ORCID 0000-0001-8133-6128

Saptsova Olga Alexandrovna, Junior Researcher, Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9). E-mail: saptsova.aa@mipt.ru SPIN code: 3710-5884, AuthorID: 1323937, ORCID 0009-0008-5445-4629

Koronnov Alexey Alexeyevich, Candidate of Technical Sciences, senior research scientist Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9). E-mail: koronnov.aa@mipt.ru SPIN code: 9854-4380, AuthorID: 116157, ORCID 0000-0001-6046-3400

Vershinina Olesya Valerievna, Junior Research Assistant, graduate student, Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9). E-mail: seraia.ov@phystech.edu

Medvedev Alexander Gennadyevich, Candidate of Chemical Sciences, Deputy Head of the Laboratory, Senior Researcher, Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9). E-mail: medvedev.ag@mipt.ru SPIN code: 3699-6664, AuthorID: 822022, ORCID 0000-0002-6762-004X

Ponomarenko Vladimir Pavlovich, Doctor of Physical and Mathematical Sciences, Chief Designer, Head of Department, Professor Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9); RD&P Center ORION, JSC (111538, Russia, Moscow, Kosinskaya st., 9). E-mail: 2392325@gmail.com SPIN code: 6882-8721, AuthorID: 171358

Popov Viktor Sergeevich, Candidate of Chemical Sciences, Head of Laboratory, Leading Researcher, Associate Professor, Moscow Institute of Physics and Technology (141701, Russia, Moscow Region, Dolgoprudny, Institutsky Per., 9); RD&P Center ORION, JSC (111538, Russia, Moscow, Kosinskaya st., 9). E-mail: popov.vs@mipt.ru SPIN code: 1128-2332, AuthorID: 625948, ORCID 0000-0002-0962-9775