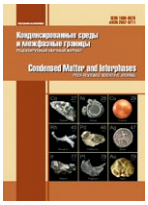

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Photoelectric response in sandwich structures based on condensed layers of Ag_2S quantum dots passivated with thioglycolic acid

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Abstract

The study is aimed at developing a technique for forming a structure with a Schottky barrier in the form of a multilayer Al-Ag_2S-ITO sandwich structure, which includes a condensate of colloidal Ag_2S quantum dots passivated with thioglycolic acid molecules (Ag_2S/TGA QDs).

The spectral properties were studied using a USB2000+ spectrometer (Ocean Optics, USA) with a USB-DT light source (Ocean Optics, USA). Electrophysical and photoelectric properties of the structures were studied using a Keysight B1500A semiconductor device analyzer (Keysight tech, USA). The study of the temperature dependences of the properties in the temperature range from 300 to 360 K was carried out in a Shielded room (Faraday cage) placed in a muffle furnace. It was found that the conductivity of the Al-Ag_2S-ITO structure is mostly governed by the Schottky barrier at the Al-condensed Ag_2S QDs film junction.

At the junction between the condensed Ag_2S QDs film and Al, signs of the formation of a rectifying contact were found.

Under the action of the optical radiation with a wavelength of 650 nm and less, which corresponds to the most probable exciton transition in the UV-Vis absorption of Ag_2S/TGA QDs, an increase in the current was found for the negative branch of the J-V curve.

Keywords: Silver sulfide, Activation energy, Charge carrier mobility, Conduction mechanisms, Schottky barrier

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1. Introduction

In recent years, optical and transport properties of nanostructures based on semiconductor colloid quantum dots (QDs) have been of particular interest. Due to the existence of QDs in the form of ensembles in colloid solutions, it is possible to obtain thin condensate layers required for the implementation of practical applications using various methods, such as spin coating, dip coating, the Langmuir–Blodgett method, etc. [1–4]. The issue of transport properties comes to the fore in such condensates and their connection with energetic and electrophysical properties of the components (substrates, electrodes, etc.) as well as optical properties of QDs condensates [5, 6].

Optical, electrophysical, and recombination properties in addition to the fabricability of QDs condensates are important for the development of new photodetectors [2] and other photonics devices. The advantages of the use of QDs in photovoltaic devices are related to the ability of fine adjustment of energy levels of the element, modification of the surface in order to increase the efficiency of light absorption, including near-infrared light range [7–9]. The correct choice of the surface ligand of QDs has a positive effect on the transport properties of condensates [6, 7, 10]. To date, it has been established that the use of shorter ligands provides the formation of concentrated layers of QDs condensates, in which the effectiveness of charge diffusion between neighbouring QDs is significantly facilitated. Thiocarboxylic acids are highly promising and can be effectively used to solve these issues [11]. However, there are few studies of the electrophysical photovoltaic properties of QDs passivated with this ligand [12]. Condensates of Ag_2S QDs, in their turn, are of particular interest as model photosensitive media. There are known approaches and technologies for the efficient passivation of interfaces with thiocarboxylic acids for them [13–26].

The possibilities of creating new thin-film systems based on condensates of Ag_2S QDs were mentioned in certain works [27–29]. For example, the addition of silver sulphide as an isolating layer in a Schottky diode improved its characteristics and increased the rate of rectification [29]. However, currently there are no available systematic studies of the conductive properties, photoresponse, and photoprocess patterns that determine them in systems based on Ag_2S QDs condensates passivated with short-chain molecules of thiocarboxylic acids.

This work partially fills this gap and is dedicated to the analysis of the electrophysical properties and photoresponse in multilayer structures of the Al-Ag_2S-ITO type, which include a condensate of colloidal Ag_2S quantum dots passivated with thioglycolic acid molecules (abbreviated as Ag_2S/TGA QDs) as a photosensitive element.

2. Experimental

2.1. Samples

Electric parameters and photoresponse were studied on Al-Ag_2S QDs-ITO sandwich structures [30]. The initial colloid solution Ag_2S/TGA QDs in the volume of 0.4 ml was applied to the conductive indium tin oxide (ITO) or aluminium substrates obtained using magnetron sputtering. Surface resistance of the substrates did not exceed 20 Ohm/sq, and geometric parameters were 10×10 mm.

The samples of colloid Ag_2S/TGA QDs were synthesised as part of water colloid synthesis. It was based on mixing aqueous solutions of $AgNO_3$ and TGA with a molar ratio of 1:1 with the aqueous solution of Na_2S the concentration of which corresponded to the molar ratio of $AgNO_3$:TGA:Na_2S which was 1:1:0.33. High purity reagents by Sigma-Aldrich were used for the synthesis.

2.2 Methods of experimental studies

Morphology of Ag_2S/TGA QDs was studied using Libra 120 (CarlZeiss, Germany) and JEOL 2000FX (JEOL Ltd., Japan) transmission electron microscope with high resolution. The surface morphology of thin films of Ag_2S/TGA QDs condensates was evaluated using reflection and transmission microscopy on a MII-4M (LOMO, Russia) micro interferometer.

The absorption properties were studied on a USB2000+ spectrometer (Ocean Optics, USA) with a USB-DT light source (Ocean Optics, USA).

The electrophysical and photoelectric properties of the studied structures were measured using a B1500A semiconductor analyser (Keysight tech, USA). The temperature dependences of

these properties in the range from 300 to 360 K were investigated in a screened chamber (Faraday cage) placed in a muffle furnace. As for the source of monochromatic radiation, we used an incandescent lamp together with an MDR-41 monochromator with a diffraction grating of 1200 mm^{-1}. The operational spectral range was 500–1300 nm, and the power of radiation on the sample was 200 μW.

3. Results and discussion

3.1. Structural and optical properties of samples

An analysis of TEM images demonstrated the formation of ensembles of individual Ag_2S/TGA QDs with an average size of 2.5 nm and a dispersion of 20% (Fig. 1a). The studies of TEM imaging with high resolution showed the formation of Ag_2S nanocrystals in a monoclinic lattice (space group $P2_1/c$) (Fig. 1b). The thickness of films determined by the interferometric method for Ag_2S/TGA QDs was 280–300 nm (Fig. 1c).

The optical absorption band of the initial colloidal solutions of Ag_2S/TGA QDs caused by the most probable exciton transition had a maximum in the region of 1.9 eV (650 nm). This value of energy exceeded the width of the band gap for Ag_2S crystals with a monoclinic crystal structure equal to 1.0–1.1 eV (Fig. 1d) [31]. This specific feature is the expression of the size effect. Using the Kayanuma formula [32]:

$$\Delta E = \frac{\hbar^2\pi^2}{2\mu R^2} - 1.8\frac{e^2}{\varepsilon R} - 0.248\frac{\mu e^4}{2\varepsilon^2\hbar^2},$$

where ε = 5.95 is the dielectric constant of bulk Ag_2S [33], R is the radius of Ag_2S QDs in cm, $\Delta E = E_{exc} - E_{mass}$ is the value of a quantum size effect in absorption spectra in erg, $\mu = \frac{m_e \cdot m_h}{m_e + m_h}$, $m_e = 0.42m_0$ and $m_h = 0.81m_0$ are the effective mass

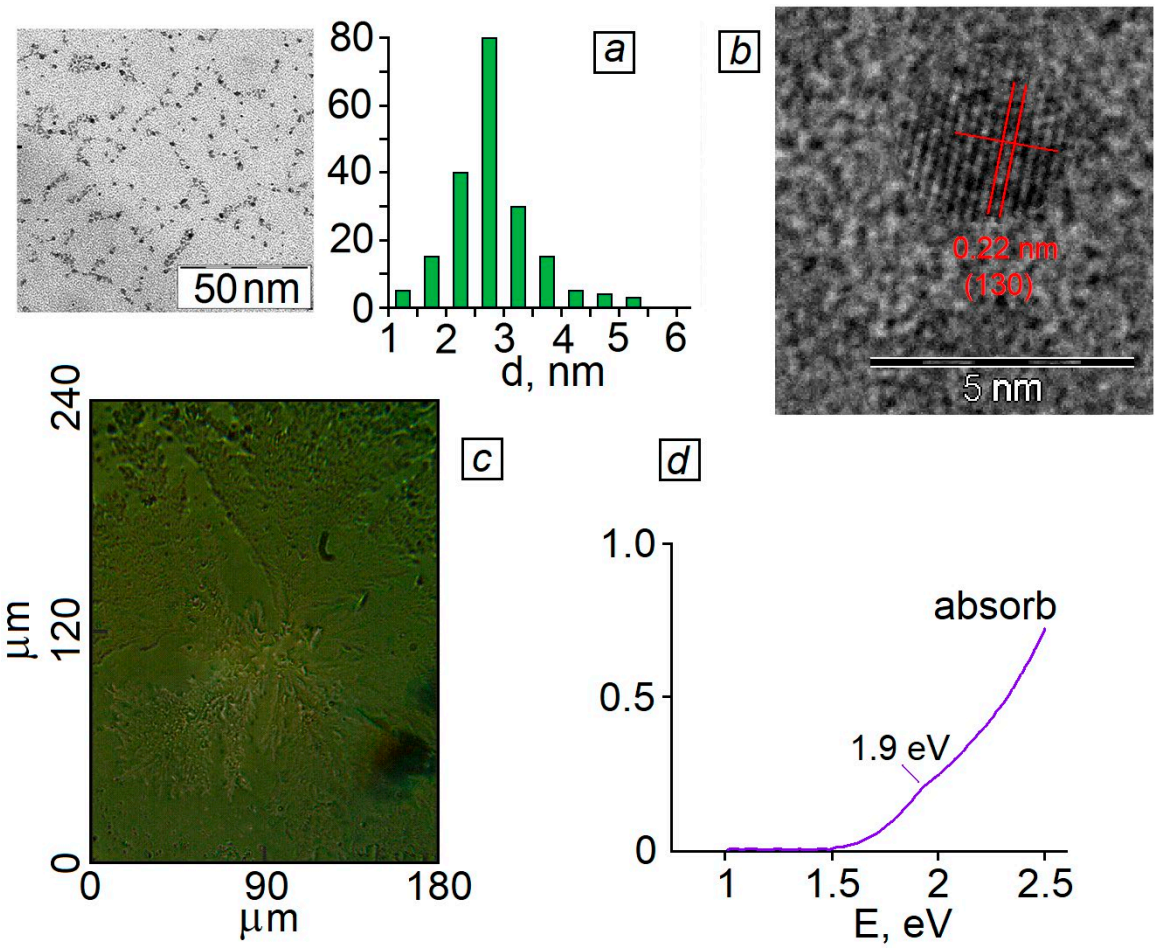

Fig. 1. TEM image, size distribution histogram of Ag_2S/TGA QDs (a). High resolution TEM (b). Optical photograph of an Ag_2S/TGA QD film (c). Optical absorption spectra of Ag_2S/TGA QDs (d)

of the electron and the hole, and the size of nanocrystals was 1.9 nm.

3.2. Electrophysical properties

Figure 2 presents the logarithmic dependence of the current on the applied voltage upon reversed bias for the studied sandwich structures Al-Ag_2S QDs-ITO. In Fig. 3 the current/voltage diagram (J-V curve) was asymmetric in relation to the zero field, which indicated the barrier structure. It should be noted that the currents were of the same magnitude, both with forward and reversed bias.

Based on the estimated value of the work function of electrons in the components of the investigated sandwich structure (4.2–4.8 eV for ITO [34, 35]; 4.1–4.9 eV was the estimated work function for the Ag_2S QDs film [31, 36], 4.25 eV was the electron work function for Al [37]) and their ratio, it can be expected that an ohmic contact will be formed in the sandwich structure at the ITO-Ag_2S QDs film interface and a Schottky barrier at the Ag_2S QDs-Al film interface.

We also formed an Al-Ag_2S QDs-Al sandwich structure to confirm the dominant role of Schottky barrier at the contact with Al-Ag_2S QDs in the formation of conductivity of the Al-Ag_2S QDs-ITO sandwich structure. A J-V curve for this structure also demonstrated the formation of a barrier, and the currents in both directions were similar while the J-V curve was symmetric in relation to the zero field. This indicated that the conductivity of the Al-Ag_2S QDs-ITO structure was mainly determined by Schottky barrier emerging at the Al-Ag_2S QDs interface. Thus, a rectifying contact was formed at the Al-Ag_2S QDs film interface due to lower work function of Al [38, 39]. J-V curves of the studied films were exponential, therefore, the conductivity mechanism was apparently implemented according to Schottky emission [40,41].

To determine the conductivity mechanism, the J-V curve were replotted on a logarithmic scale [42], which allowed determining the dominant conductivity mechanism. Experimental dependencies of current on the applied voltage (Fig. 3) could be described in three regions by the power law $I \sim U^m$[43], where m is the value of the slope for each region of the applied voltage. The value m showed the kinetics of charge carriers and the type of conductivity [42].

The resistive conductivity mechanism is typically [42] observed for the lowest voltage applied to the investigated structure, when the concentration of charge carriers injected into the QDs film is significantly lower than the concentration of intrinsic carriers. In this case, the coefficient m must be equal or close to one. Experimentally determined value m_1 for the region of voltages up to 0.2 V was equal to 0.2. As the voltage increased, the regions from 0.2 to 0.7 V could also be identified, where the value m_2 was 2.3, and the range of voltages more than 0.7 V, where m_3 was 5.49. The region of

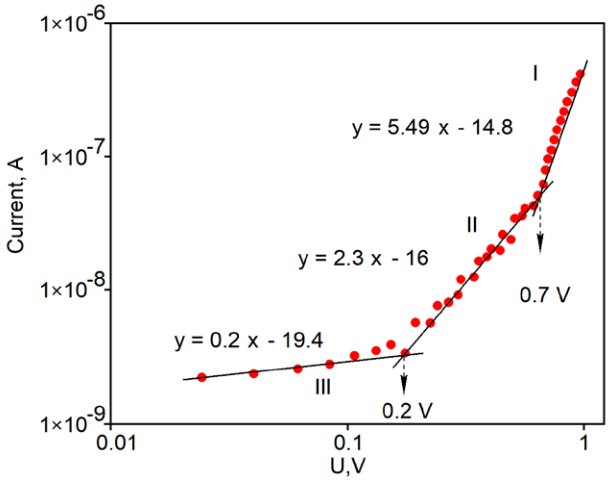

Fig. 2. Logarithmic dependence of the current on the applied voltage at a negative bias at T = 300 K for thin-film sandwich structures based on Ag_2S/TGA QDs

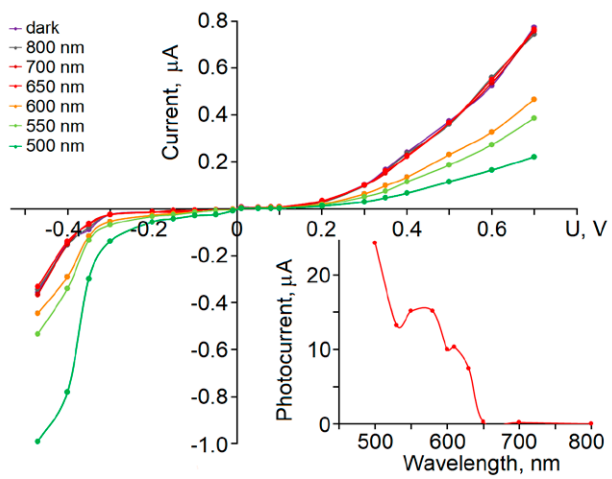

Fig. 3. *J-V* curve for sandwich structures based on Ag_2S/TGA QDs. The inset shows the photosensitivity spectrum of sandwich structures based on Ag_2S/TGA QDs

low voltages (<0.2 V) showed slightly strange behaviour, which was apparently due to the heterogeneous structure of the Ag_2S QDs film and the contribution of several processes to the conductivity. These processes will be described in detail in a different work.

A current mode with a space charge limited conduction (SCLC) was observed in the range of applied voltages from 0.2 to 0.7 V. The third range corresponded to the charge transfer due to the trap charge limited conduction mechanism (TCLC) [45]. Localised states of recombination luminescence centres seemed to participate in the formation of this third section of J-V curve (Fig. 1). Shallow localised states that we previously discovered for similar samples of colloidal Ag_2S QDs using thermally stimulated luminescence can also have a considerable effect [46].

3.3. Photoelectric response

Figure 3 presents the J-V curves for Al-Ag_2S QDs-ITO sandwich structures both in the absence of radiation and in the presence of illumination for the film structure with monochromatic radiation. It can be seen that there are no changes in the negative branch of J-V curve for the optical wavelength of more than 700 nm (1.77 eV). When comparing to the optical absorption spectra of Ag_2S QDs films, for which an exciton absorption peak was observed in the region of 1.9 eV (650 nm), and the long-wave absorption edge is approximately 700 nm, we can state that the absorption of light by Ag_2S QDs condensates is the primary act in the formation of the photoresponse of sandwich structures based on Ag_2S/TGA QDs. The form of the photocurrent spectrum measured for the negative shift (0.2 V) reiterated the long-wave edge of the absorption spectra of the Ag_2S/TGA QDs film. The forward J-V curve branch shows the changes that occurred under the influence of the radiation with a wavelength of less than 700 nm, which also proves the dominant role of the absorption process in the Ag_2S/TGA QDs film for the formation of a photoresponse.

4. Conclusions

As a result of studying the electrophysical properties of the Al- Ag_2S QDs-ITO sandwich structure, where the main working component was a condensate of colloidal Ag_2S QDs passivated with thioglycolic acid (TGA) molecules, we established new patterns that indicate the formation of photosensitive systems with a Schottky barrier. The obtained experimental data showed that the conductivity of such structures was determined by Schottky barrier at the Al-Ag_2S QDs interface. Spectral area of photosensitivity of these structures coincided with the region of absorption of Ag_2S QDs condensates. The obtained results indicate that model objects can be developed for photodiode structures based on condensates of colloid Ag_2S/TGA QDs.

Contribution of the authors

The authors contributed equally to this article.

Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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