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## Semiconductor Quantum Dots as Materials for Lasers Based on Them

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Quantum dots (QDs) today belong to the central research objects of many scientific groups. The study of the properties of structures of small size is important both for the further development of electronics, and for the improvement of existing semiconductor devices. At present, there are many methods of obtaining QDs obtaining under the laboratory conditions: the method of ultrasonic grinding, molecular beam epitaxy, pulsed laser ablation, as well as using such methods of chemical synthesis as organometal synthesis, synthesis in reverse micelles, electrothermal synthesis, sol-gel synthesis, synthesis using thiol stabilizers, synthesis in a non-aqueous medium. Nowadays, the most promising methods are based on the use of the phenomenon of self-organization. These are molecular-beam epitaxy and colloidal chemistry methods, and the latter are used for synthesis of nanosized crystals. The band gap width and the energy of luminescence peak are determined by the size of the particles.

**Keywords:** quantum dots; hydrothermal synthesis, solvothermal synthesis; semiconductor quantum dots; quantum-dimensional effects.

*Article acted received 17.04.2018; accepted for publication 15.06.2018.*

### Introduction

In recent years, the research interest in semiconductor heterostructures with null-dimensional stressed nanobjects (quantum dots (QDs)) has increased considerably. For a long time, low-level nanosystems form a new area of research in physics of the solid state. The study of the properties of structures of small size is important both for the further development of electronics, and for the improvement of existing semiconductor devices. Modern semiconductor technologies allow to create artificial space constraints for quasiparticles. In view of this, semiconductor nanodeterostructures, and especially QD, today belong to the central research objects of many scientific groups.

### I. Quantum dots: basic concepts

Quantum dots (QD) - is an isolated semiconductor crystal, limited by all three spatial coordinates, with dimensions smaller than the characteristic Borov radius of the exciton in this semiconductor. The potential of practical application of QD is related to the possibility of controlling the effective band gap width when changing linear dimensions.

Quantum-dimensional effects play a key role in the

optoelectronic properties of QD. The energy spectrum of QD (called "artificial and atoms") differs significantly from the spectrum of a bulk semiconductor. There are two basic approaches to describing the energy spectrum of QD - "top-down" and "bottom-up" [1] In a voluminous semiconductor, the charge carrier can be moved in all three directions of space. In QD, the charge carrier is spatially limited in all three coordinates, since the dimensions of the QD are commensurate with the wavelength of the electron oscillation. As a result, the charge carrier in a nanocrystal is in a three-dimensional potential well [2], which leads to the degeneracy of the continuous energy spectrum of a bulk semiconductor at discrete levels.

An important characteristic of semiconductors and semiconductor nanoparticles is the gap band gap ( $E_g$ ) between the upper energy level of the valence band (VB) and the lower level of the conduction band (CB). When a photon is absorbed by a semiconductor with energy exceeding the width of the band gap, the transition of the excited electron from the valence band of the crystal to the conduction band takes place. This process is, strictly speaking, the formation of two quasiparticles - an electron  $e^-$  with an effective mass  $m_e^*$  and a negative charge  $-e$  in the conduction band, characterizing the excitation of the electronic subsystem of the crystal, and the hole with an effective mass  $m_h^*$  and a positive charge

+e characterizes an ensemble of electrons of the valence band, from which one electron was removed. The coupling state of the electron-hole pair forms a hydrogen-like quasiparticle, an exciton, which is characterized by the Bory exciton radius  $a_B$  [3]

$$a_B = a_0 \frac{e' m_0}{m}, a_0 = \frac{4\pi \epsilon_0' \hbar^2}{m_0 e^2}, \quad (1)$$

where  $a_0$  - the first radius of Bohr is an atom of hydrogen,  $\epsilon_0'$  - dielectric has become,  $\epsilon'$  - dielectric to penetrate this semiconductor,  $m_0$  - mass of tranquility of the electron,  $\mu$  - effective mass of the exciton.

$$\frac{1}{m} = \frac{1}{m_e^*} + \frac{1}{m_h^*}, \quad (2)$$

where  $m_e^*$  - electron mass,  $m_h^*$  - mass hole. The Boris radius of the exciton is larger than the corresponding radius of the hydrogen atom and the period of the crystal lattice, since the effective masses of the electron and the hole are smaller than the electron resting mass, and  $\epsilon$  is much greater than 1. For most semiconductors,  $a_B$  lies within the range of 1-15 nm, which significantly exceeds parameters of the crystal lattice ( $a_L$ ) and allows to receive structures whose size is  $d_i$  in one, two or all three directions  $a_L < d_i < a_B$ . In the latter case (with a space restriction on all three coordinates) the resulting object is quasi-zero-dimensional and is called a quantum dot.

With the size of the particle of order  $a_B$  and less the exciton feels the spatial constraint, the electron-hole pair is localized inside the nanocrystals, and the exciton can be regarded as a "particle in a box" (a three-dimensional potential well). The energy of the exciton inside the nanoparticle is greater than the energy of the "free" exciton in the bulk material, which leads to an increase in the band gap width [4].

In a bulk semiconductor crystal, the total exciton energy consists of the width of the band gap  $E_g$ , which is reduced by the binding energy of the electron and the hole (exciton energy)  $R_y^*$  and the kinetic energy of the center of mass of the exciton. The last for a spherical nanoparticle with radius  $r \sim a_B$  corresponds to the exciton energy in a potential well and is  $n^2 \pi^2 \hbar^2 / 2\mu r^2$ . The Coulomb interaction of an electron and a hole gives the following expression for the exciton excitation energy of a spherical semiconductor nanoparticle:

$$E = E_{g \text{ bulk}} + \frac{n^2 \pi^2 \hbar^2}{2R^2 m} - 0,248 R_y^* - \frac{1,78 e^2}{e'R}, \quad (3)$$

$$R_y^* = \frac{e^2}{2e'a_B}$$

The last three plots (3) correspond to the energy of the localization of the exciton in the nanoparticle. Values  $n > 1$  correspond to the exciton transition in higher excited states. The last member of the expression takes into account the Coulomb interaction. The sum of the first two terms (3) represents the effective width of the bandgap in nanocrystals:

$$E_{g \text{ qd}} = E_{g \text{ bulk}} + \frac{n^2 \pi^2 \hbar^2}{2R^2 m}, \quad (4)$$

The electron-optical properties of quantum-dimensional structures depend on the size of the crystal in the direction in which the motion of charge carriers is limited. For anisotropic structures, the effective masses of an electron and a hole depend on the direction of motion, and in this case the expression (3) is complicated:

$$E = E_{g \text{ bulk}} + \frac{\pi^2 \hbar^2}{2} \left( \frac{n_x^2}{m^2 L_x^2} + \frac{n_y^2}{m^2 L_y^2} + \frac{n_z^2}{m^2 L_z^2} \right) - 0,248 R_y^* - \frac{1,78 e^2}{eR}, \quad (5)$$

where  $\mu_i$  - effective masses of the exciton,  $L_i$  - geometric dimensions of the structure along the corresponding directions ( $i = x, y, z$ ),  $n_i$  - integers [5].

From formula (4) it can be seen that as the size of the QD increases, the gap between the levels of the quantization of the electron and the hole increases, which leads to an increase in the energy of the optical transitions. In the first approximation, the effective width of the forbidden band of QD increases with decreasing particle size as  $R^{-2}$ . This is the main manifestation of the quantum-dimensional effect for semiconductors - the "blue shift", the hypochromic displacement of the edge of the absorption band and the position of the peak of luminescence. When the photon of the corresponding energy is excited, the electron passes from the VZMO to the NSMO. Such electronic transitions between levels of dimensional quantization are called excitonic transitions. The energy below the exciton maximum ( $n = 1$ , see formula (3)) in the absorption spectra of nanoparticles (near the longwave edge of absorption) is quite clearly detected and serves as a source of information as to the electronic structure of nanoparticles (that is, the width of the bandgap QD-Eg qd), and the statistical characteristic of this ensemble of low-frequency (ie, the distribution of particles by size) [6, 7]. The energy of the first exciton peak of the QD is determined by the composition of the width of the band gap  $E_g$  and the size of the QD [8, 9, 10].

Thus, using different materials and changing the size of the particles, the width of the band gap obtained by the CT can be varied in the exemplary range  $\sim 4.0$ - $0.5$  eV ( $\sim 300$ - $1500$  nm), covering the spectral range from the infrared to the UV region [11].

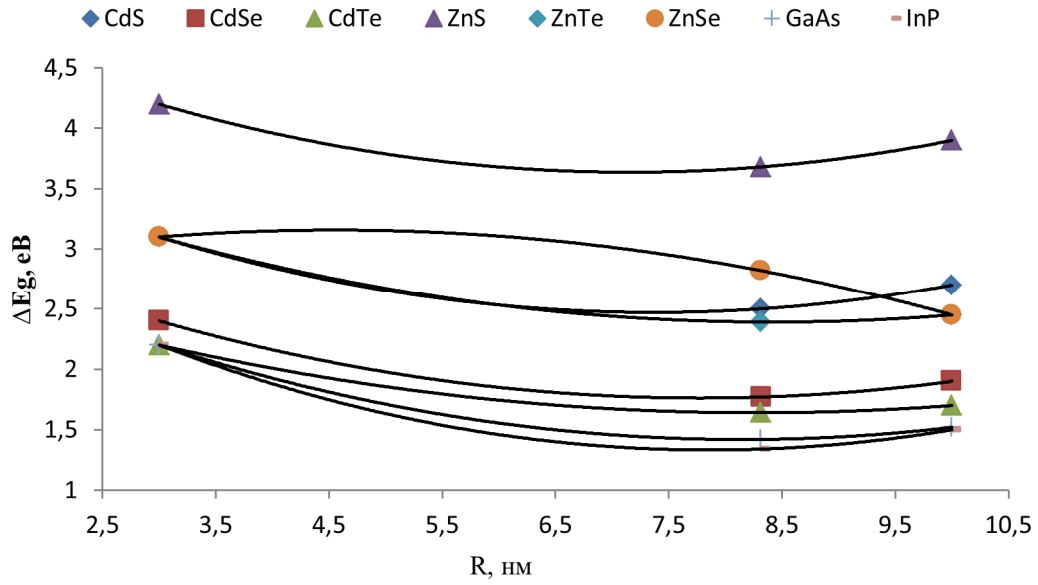
In fig. 1-2 shows the dependence of the band gap width on its size for different materials.

$$D = (1,61 \cdot 10^{-9}) \lambda^4 - (2,66 \cdot 10^{-6}) \lambda^3 + (1,62 \cdot 10^{-3}) \lambda^2 - (0,43) \lambda + 41,57,$$

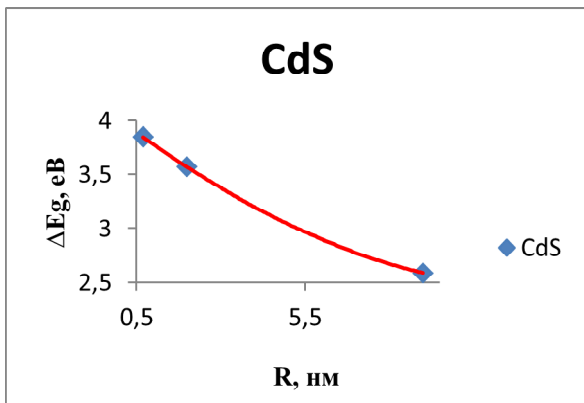
where  $D$  is a diameter of QD,  $\lambda$  is a position of the first exciton peak of absorption.

QDs attract attention due to its luminescence properties. Semiconductor nanocrystals possess a number of parameters that allows manipulation of the band gap characteristics and, consequently, spectral-luminescence properties. Such the parameters are the size, shape and composition of nanoparticles.

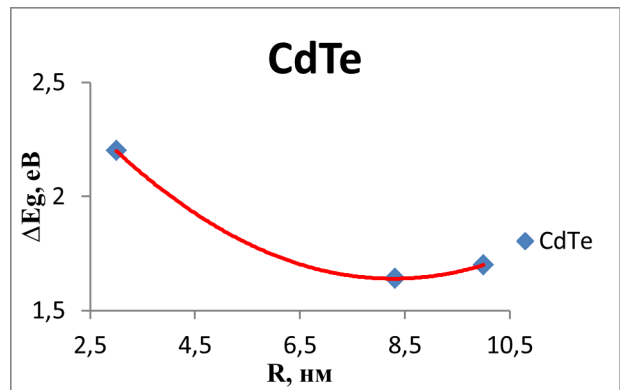
The dependence of the luminescence of quantum dots on their size is in a very wide range of the spectrum - from the near ultraviolet to infrared radiation.



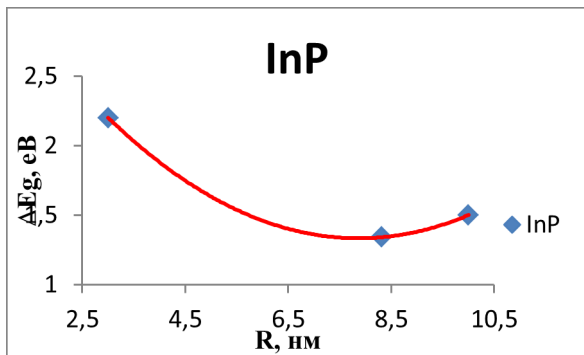
**Fig. 1.** Dependence of the band gap width of the quantum dots on its size CdS, CdSe, CdTe, ZnS, ZnTe, ZnSe, GaAs, InP.



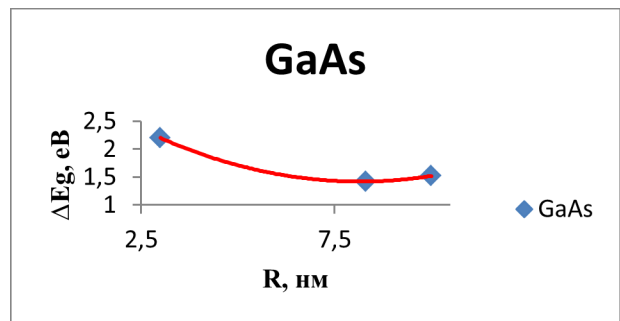
**Fig. 1, a.** Dependence of the band gap width of the quantum dots on its size CdS.



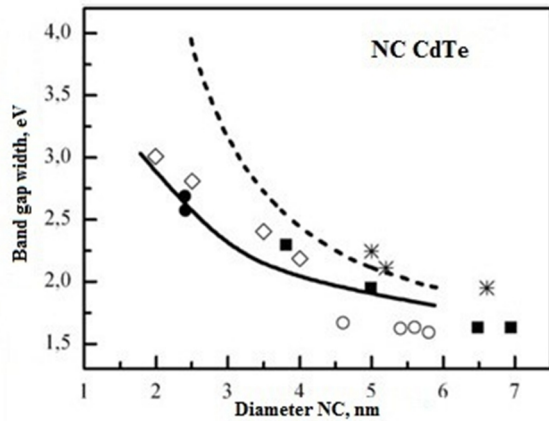
**Fig. 1, b.** Dependence of the band gap width of the quantum dots on its size CdTe.



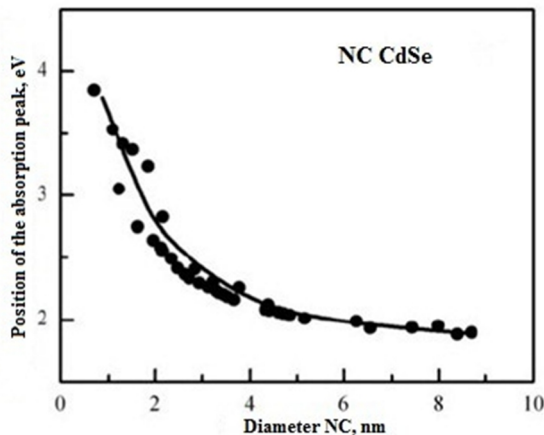
**Fig. 1, v.** Dependence of the band gap width of the quantum dots on its size InP.



**Fig.1. g.** Dependence of the band gap width of the quantum dots on its size GaAs.



**Fig. 2.** Change the band gap width of CdTe IP with the size. Experimental data are obtained at different temperatures: [12] ( $\diamond$ ) and [12] ( $\circ$ ) - 300 K; [13] ( $\circ$ ) -> 300 K; [14] ( $\bullet$ ) -77 K (higher values) and 300 K (lower values). An integral line is the theoretical values of calculations in the approximation of the strong coupling [15], the dashed line is in the approximation of effective masses.



**Fig. 3.** Dependence of the diameter of the CdSe quantum dots on the position of the first excitonic absorption peak, measured at  $T = 300$  K. Points are the synthesis of data from work [16]. The dimensions of the nanocrystal are determined from the data of the enlightening electron microscopy, with the exception of the smallest NC, the dimensions of which are determined by the XRD data. An empirical subordinate curve for the dependence of the dimensions (solid curve).

CTs have a great potential for practical application wherever variables are needed, optical properties rebuilt in wavelengths - in lasers, solar panels, LEDs [17, 18, 19].

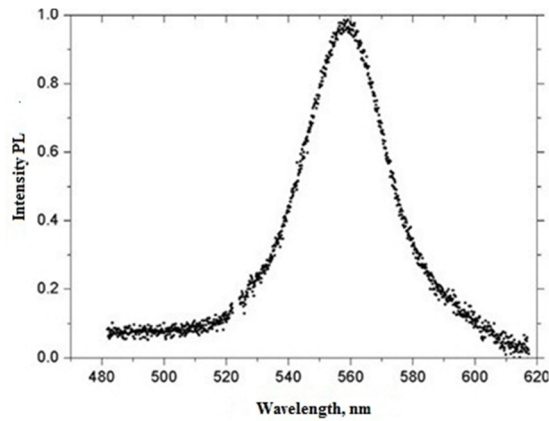
The term "colloidal" is introduced to distinguish between "classical" quantum dots grown on a substrate by the method of molecular beam epitaxy, from sols obtained by chemical methods in a solution.

Colloidal quantum dots significantly differ from the above systems because they are chemically synthesized using wet chemistry and there are free nanoparticles or nanocrystals grown in solution. Colloidal quantum dots

are a subgroup of a broader class of materials that can be synthesized at a nanoscale level using wet chemical methods. In the production of colloidal nanocrystals, the reaction chamber is a reactor that contains a mixture of compounds that controls the nucleation and growth. In the general synthesis of quantum dots in the solution, each type of atoms, (which are part of the nanocrystal) is introduced into the reactor in the form of a precursor. A precursor is a molecule or complex containing one or more types of atoms, which are necessary for the growth of nanocrystals. Once precursors are introduced into the jet chamber, they decompose and form new reactive units (monomers) that cause nucleation and growth of nanocrystals. The energy required for the decomposition of precursors provides a liquid in the reactor, or it is provided by thermal collisions, or by a chemical reaction between the liquid medium and the precursors, or a combination of these two mechanisms [20].

The colloidal method refers to the methods of grain growth and allows the large variation of the concentration of colloidal particles in the solution. Such colloidal nanosystems with particle sizes of 1 - 100 nm are considered to be classified into ultrafine systems [21]. In the organic solvent, solutions are introduced from the elements of the II and IV groups of the Mendeleev table, which settle on the solvent molecules. In carrying out such reactions, it is necessary to carefully observe a number of parameters, such as the pH level and the concentration of a number of organic compounds in the system, as it significantly affects the size of the particles in the colloid. As an example, the synthesis of colloidal solutions of CdSe quantum dots is carried out in two stages [22]. In a typical method, large CdSe nanocrystals (radius of about 2.5 nm) are synthesized by placing the solution from the active mixtures containing Cd and Se in a flask and heating in a vacuum at temperatures above 100°C. First, the growth of the grains of the new phase is in progress. Upon reaching a certain maximum temperature ( $\sim 200$  ° C), a weak supersaturation is created in the solution, and the CdSe nanocrystals become insoluble in the reaction mixture. The formation of new grains is discontinued and at this temperature quantum dots are slowly grown from these "nucleated crystals" and increase in size as the reaction proceeds. The size of the particles is monitored by recording, for example, the photoluminescence of the growing solution. After the desired particle size has been reached, the solution is cooled and washed several times to remove the by-products, while the synthesized CdSe particles should be insoluble in any solvent used. For the synthesis of smaller CdSe nanocrystals (radii from 1.0 to 1.5 nm), the growth solution of particles with the same ratio of reagents is cooled as soon as its temperature reaches the above maximum. After precipitation and washing of CdSe nanocrystals, they can be used as nuclei in the synthesis of CdSe / ZnS kernel / shell structures. Figure 4 shows the photoluminescence spectra of colloidal quantum dots CdSe / ZnSe.

It was the discovery of convenient methods for the chemical synthesis of ashes of monodisperse nanocrystals of chalcogenide semiconductors in the early 90's which served as a catalyst for the study of the properties of these objects, among which, first of all, the



**Fig. 4.** Photoluminescence spectra of colloidal QDs CdSe / ZnSe.

presence of discrete energy levels of carriers instead of bulk semiconductor zones, and their state because of quantum-dimensional effects strongly depends on the size of nanocrystals. Colloid QDs, thus, represent a huge theoretical interest, but they gained true popularity due to their practical properties. The absence of defects in the crystalline structure, together with the good passivation of the surface, increases the probability of radiative recombination of the carriers after photo-excitation, which leads to high quantum photoluminescence outputs (up to 80%). These properties have practical application in creating on their basis new generations of solar cells, lasers, sensors.

## II. Classification of quantum dots

Colloidal synthesis of quantum dots opens up wide opportunities in obtaining quantum dots based on various semiconductor materials and quantum dots with different

geometry (form). Most of the advantages of colloidal quantum dots is due to the presence of quantum-dimensional effects in them [23], because their size is much smaller than the de Broglie wavelength of the electron for the material that forms the core of the nanoparticle. The fundamental parameters of optical transitions in CT can be described using the approximation of an effective mass based on exciton effects [24]. It is important to synthesize quantum dots, complex semiconductors, which are characterized by different composition, size, and form.

### 2.1. Composition of QD (semiconductor material)

In the first place, quantum dots are of practical interest as luminescent materials. The main requirements for semiconductor materials on which QD are synthesized is the presence of a direct-band nature of the band spectrum, which provides effective luminescence, has an effective mass of charge carriers and manifests quantum-size effects in a rather wide range of sizes within the size of nanocrystals. Among the semiconductor materials used to obtain QD, it should be noted:

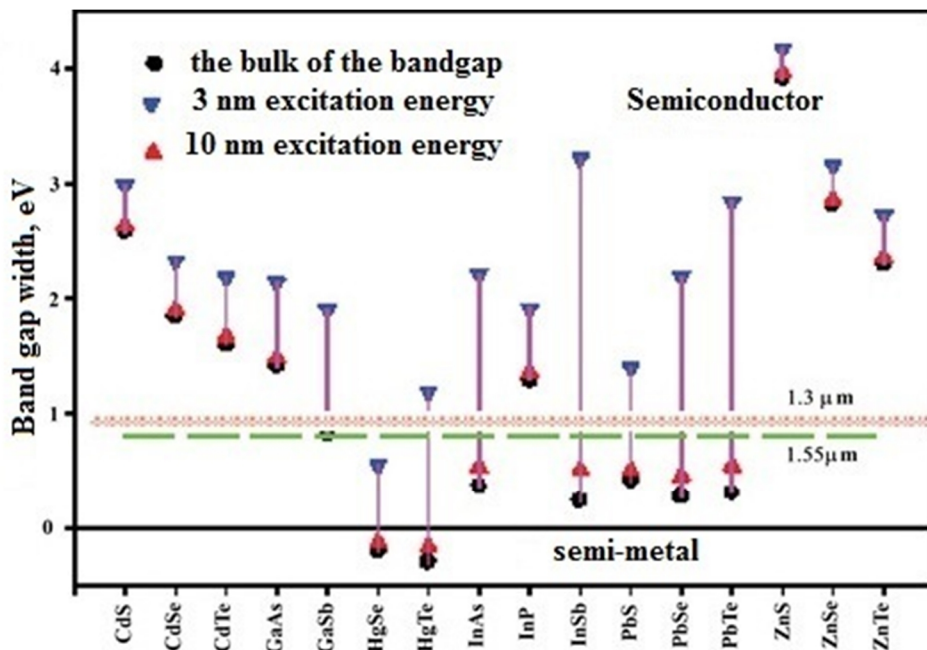
broadband semiconductors (oxides ZnO,  $TiO_2$ ) - ultraviolet range, midrange semiconductors ( $A^2B^6$ , for example, cadmium chalcogenides),  $A^3B^5$  - visible range and narrow-band semiconductors ( $A^2B^4$ , such as chalcogenides of lead) near infrared range.

Figure 5 shows the possibility of varying the effective band gap width for the most common semiconductor materials in the form of nanocrystals with the size of 3-10 nm.

### 2.2. The form of QD

In addition to the composition and size seriously affects the properties of QD geometric shape.

- Spherical QD (directly quantum dots) - this is a large part of quantum dots. At the present time, they have



**Fig. 5.** Ranges of effective width bandwidth change for quantum dots when changing from 3 to 10 nm.

the most practical application. Which are the most simple to manufacture. Modern experimental methods make it possible to create multilayer quantum dots (QDs) both closed and open type [25]. For spherically symmetric, multilayer QD heterostructures, taking into account the simple parabolic law of the dispersion of quasiparticles, rectangular spherically symmetric potentials and barriers by the S-matrix method, and the solutions of the corresponding Schrödinger equations [26], it is possible to determine the dependences of the energy spectra of the particles from the geometric size of the layers of the nanoheterostructure.

- Ellipsoidal QD (nanorods) - nanocrystals drawn in one direction. Coefficient of ellipticity 2-10. From a practical point of view, this class of quantum dots has application as a source of polarized radiation. For large coefficients of ellipticity  $> 50$ , this type of nanocrystals is often called threads (nanowires).

- Complex geometry QDs (for example, tetrapods) can be synthesized in quite different forms - cubic, asterisk, etc. From a practical point of view, tetrapods can be used as molecular switches. Currently, they are of a largely academic interest.

### 2.3. Multicomponent QD

Methods of colloidal chemistry allow to synthesize size multicomponent quantum dots from semiconductors with different characteristics, primarily with different band gap widths. This classification is largely analogous to that traditionally used in semiconductors.

InSb-based heterostructures derived from InAs and GaSb narrow-band substrates are promising for optoelectronic devices operating in the medium-infrared (IR) range of 2-5 microns. InSb QD-based heterostructures placed in the narrow-band matrix n-InAs were characterized by electroluminescence in the range of wavelengths of 3-4 microns in a wide range of temperatures up to room temperature. The observed luminescence was due to the interface radiated transitions of electrons from self-consistent quantum wells on the side of InAs matrix layers through a discrete heterojunction of the InSb / InAs type II at the level of quantization of holes in the InSb QD. However, nanoheterostructures in the InSb / InAs system only work in the long-wavelength part of the IR range (from 3 to 5  $\mu\text{m}$ ), whereas for penetration into the short-wave portion (from 2 to 3  $\mu\text{m}$ ), matrices with a larger band gap will be needed.

#### a) Alloyed QD

Typically, the number of impurities 1-10 atoms per quantum dot with an average number of atoms in the quantum dot is 300-1000. At the same time, the electronic structure of the quantum dot does not change, the interaction between the atom of the impurity and the excited state of the quantum dot is dipole and reduces to the transfer of excitation. The main alloying impurities are manganese, copper (luminescence in the visible range).

#### b) QD based on solid solutions

For quantum dots, the formation of solid solutions of semiconductors is possible if there is a mutual solubility of materials in bulk state. As in the case of bulk semiconductors, the formation of solid solutions leads to a modification of the energy spectrum - effective

characteristics are the superposition of values for individual semiconductors. This approach allows you to change the effective width of the bandgap for a fixed size - it gives one more way to control the characteristics of quantum dots. An example is the built-in layer of the solid solution of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  - ( $x \sim 0,1-0,4$ ) in a relatively thick ( $\sim 1 \mu\text{m}$ ) GaAs layer. In a QD, such a potential well restricts the motion of electrons and holes in all three directions. The width of the band gap of the  $\text{In}_x\text{Ga}_{1-x}\text{As}$  -  $E_g(x)$  is less than the band gap band GaAs 1.43 eV and decreases linearly with increasing  $x$  to 0.36 eV in InAs. On the verge of these materials, there is a heterojunction of the so-called "catching" type, in which the breaks in the conduction band  $\Delta E_c(x)$  and the valence band  $\Delta E_v(x)$  form spatially coupled potential wells for electrons and holes respectively [27].

#### c) QD based on heterojunctions

This approach is realized in QD-type core shell. Properties of nanocrystals can be controlled by coating them with a shell of another material. The shell material in such a "core-shell" structure may be a metal, a semiconductor, or an oxide. The shell material allows for improved properties of nanocrystals. For example, defects localized on the surface of semiconductor NCs can be transferred to the buffer layer of the shell material to obtain NC with a higher quantum emission. In the general case, it involves the formation of a contact of two parts from different semiconductors. By analogy with the classical theory of heterojunctions, two types of QD kernel shells can be distinguished (Fig.6)

Type 1. The width-wise semiconductor is a shell, narrow-band-core. the relationship between the band gap width is shown on (Fig. 6). A broadband semiconductor acts as a passivator of surface states and locates an electron-hole pair inside the core. This approach is used to increase the luminescence efficiency.

Type 2. The width of the forbidden semiconductor zones can be compared, but the edges of the zones are shifted relative to each other. Excited charge carriers are energetically advantageous in different parts of such a nanocrystal, for example, it is advantageous to have an electron in the shell, a hole in the nucleus (or vice versa). The kernel shell of type 2 demonstrates the spatial separation of charge carriers and may be interesting for the creation of systems with a large lifetime of photo-stimulated electron-hole pair. Another effect is to reduce the effective band gap width (band gap width is determined by the energy difference between the edge of the conduction band of the shell material and the edge of the valence band of the core material) and the shift of the maximum luminescence to the red area.

Different methods are used to obtain QD-type "core-shell". The key requirements for such techniques are as follows: the ideal method should only give nanoparticles of the "core-shell" type, and not a solid solution nanoparticle; The layer thickness of the shell should be uniform for monodisperse nanocrystals of the "core-shell" type.

The formation of "core-shell" heterostructures was first demonstrated in [28] on the example of CdSe / ZnS and ZnS / CdSe nanoparticles. Fig. 7 The nanoparticles are also obtained as a result of controlled deposition of molecules of the same type of semiconductor (shell) into



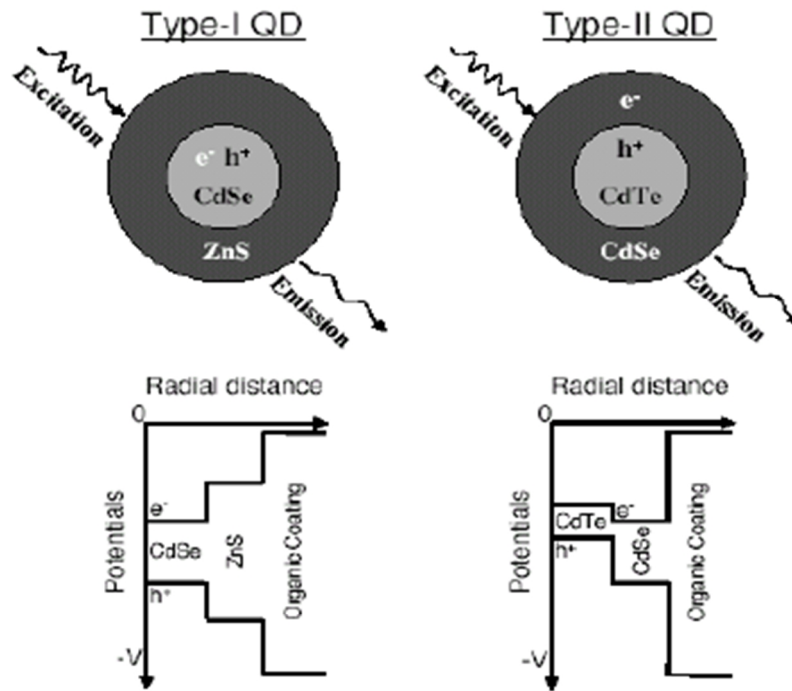


Fig. 6. Schematic structure and band diagram for quantum dots core shell from heterojunctions of type 1 and type 2.

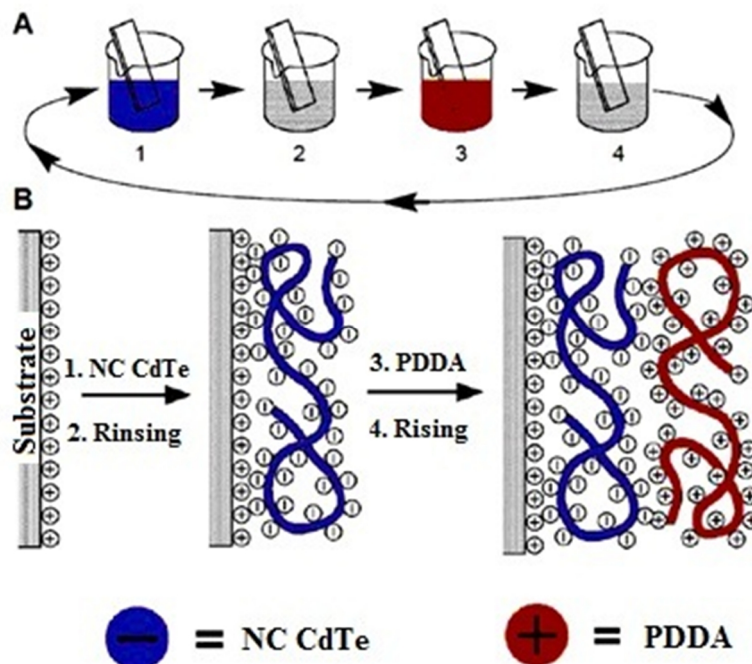


Fig. 7. Schematic representation of the cyclic process of layer deposition of films from colloidal solutions of nanocrystal CdTe [29].

a pre-synthesized nanoparticle of another type (nucleus). Synthesis of the shell is carried out in colloidal solution by the methods of synthesis of one-component nanoparticles. Crystal shells on a nanocrystalline nucleus grow despite the fact that the lattices in CdSe and ZnSe become different by 13%.

The following "core-shell" nanoparticles were also synthesized HgTe/CdS, ZnS/ZnO и ZnO/ZnSe, TiO<sub>2</sub>/SiO<sub>2</sub> and TiO<sub>2</sub>/SnO<sub>2</sub>, TiO<sub>2</sub>/ZnO. Similar heteronanoparticles can, in turn, be coated with another layer of

semiconductor.

### III. Methods of synthesis

The synthesis of nanomaterials of controlled size and shape plays a decisive role in the field of nanotechnology. Since the initial study of their properties began in the 1980s, semiconductor nanocrystals (so-called quantum dots (QDs)) demonstrated many different properties, characteristics and potential applications. The

microscale and larger semiconductor crystals have electronic and optical properties that depend on their bulk band structures. When the size of these crystals is reduced to a nanoscale, they essentially become a molecule, and this leads to the unique properties that distinguish nanocrystals from their bulky analogues. Most important in QD, certain properties no longer depend on their band structure, but only on the size and shape. For example, in the case of CdS, bandgap widths can range between 4.5 (275 nm) and 2.5 eV (450 nm), since their size varies from macro to molecular level. The energy above the bandgap, needed to add excess charge, also decreases by 0.5 eV. This range in the fundamental properties of the material opens up further possibilities to trace the evolution of the electronic and optical properties of matter from the bulk material to the atomic cluster.

There are currently quite a large number of methods for synthesizing QDs. These can be approaches based on the "crushing" substance (from the top down), and on the "cultivation" of QDs (from the bottom to the top). Below we consider methods for obtaining colloidal quantum dots - nanocrystals of a semiconductor, coated with a coil of stabilizer molecules. For synthesis of colloidal quantum dots, chemical methods based on the growth of nanocrystals are used. In general, the synthesis of high-quality nanocrystals with a narrow size distribution presents some difficulties. QDs of the best quality are obtained by increasing in nonpolar environments - the method proposed by C.B.Murray in 1992. and gave rise to a rapid growth of interest in colloidal quantum dots.

- Synthesis of colloidal QDs in nonpolar environments

The method of colloidal quantum dots in nonpolar media has a number of undoubted advantages:

1. possibility to control the process of growth of QDs
2. good passivation of surface states QDs
3. narrow size distribution (at 5-8% level)
4. possibility of further allocation and clearing of QDs

- Missing matrix

The currently developed version of this method includes synthesis in

coordinating solvent and synthesis with a coordinating precursor:

- Synthesis in a high boiling stabilized solvent (e.g. trioctylphosphine oxide) [30].

Precursors are demethylcadmium and trioctylphosphine chalcogenide

- Synthesis in a high boiling inert solvent (non-coordinating solvent, for example, diphenyl ether) using stabilizing precursors (e.g., cadmium oleate) [31]

In colloidal synthesis, you can distinguish three main stages:

- 1) Nucleation;
- 2) Growth of the embryos;
- 3) oswald maturation stage;

#### 1. Nucleation stage in a supersaturated solution

In accordance with the classical theory of nucleation, nucleation occurs spontaneously: in some unstable regions, the saturated solution of a molecule or dissolved ions in itself can crystallize, forming embryos. Homogeneous nucleation is due to thermodynamically,

since the supersaturated solution is unstable in energy [32].

#### 2. Stage of growth of embryos

The colloidal theory in interpreting the phenomena associated with the growth of crystals proceeds from the relationship between the shape of the crystal and the surface energy of all its faces. According to the diffusion interpretation of the growth of crystals, the process of formation of a crystalline face proceeds at a high velocity and depends only on the diffusion velocity. Since the process is diffusive, the main parameter for its regulation is the temperature [33].

#### 3. Oswald's maturation

When reagents are exhausted due to the growth of particles, Ostwald's maturation process begins, in which large particles continue to grow due to smaller dissolution, reducing the superficial energy of the system. In this case, defocusing occurs. With a decrease in the degree of over-saturation of a critical size, the embryos grow and particles smaller than the critical size dissolve. If the reaction is quickly stopped at this stage, the particles will have a wide distribution in size. At the stage of Ostwald's maturation, it is impossible to obtain monodisperse particles. The size that remains after the complete disappearance of the supersaturation of particles can reach several micrometers, so that nanocrystals of good size distribution can be obtained only with explosive nucleation and reaction stops quickly after its completion and before the start of Ostwald's maturation. For explosive nucleation it is necessary to create a high degree of supersaturation.

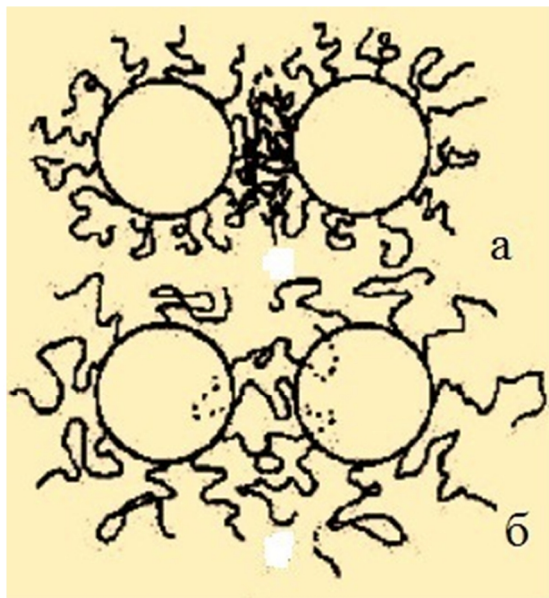
#### Particle stabilization.

An important issue is the stabilization of the sol. It is necessary to achieve not only the given dispersion, but also to stabilize the properties of the solution in time. This can be achieved by reducing the energy of the surface tension on the boundary of the phase separation, which leads to the presence in the system, in addition to the dispersion medium and the dispersed phase of another component - the stabilizer of the disperse system. Adsorbing on the boundary of phase separation, it creates a structural and mechanical barrier and thus prevents aggregation. The successful choice of the stabilizer is crucial in the colloidal synthesis of QDs. Requirements for the stabilizer are relatively high: first, it must prevent the aggregation of particles of the dispersed phase. Second, do not interfere with the diffuse growth of QDs. Thirdly, by focusing on the surface of the colloidal particle, the stabilizer molecule must be firmly bound to it with a covalent bond on one side, whereas the free end of the molecule must have a strong affinity for the solvent, that is, in the dispersion medium, thereby providing a rather high "solubility" of QDs, Fig. 8 and, fourthly, the stabilizer molecules should pass the surface of the QDs, eliminating the broken ties and localizing within it the charge carriers.

#### Dimensional-selective deposition.

"Solubility" of QD different sizes is different. This phenomenon is based on the method of dimensional-selective deposition. Larger particles can be precipitated and get a solution of substantially identical particle size. At present, it is possible to separate quantum dots with diameters, which differ only by several percent [30]. The





**Fig. 8.** Mechanisms that counteract the convergence of colloidal particles due to local increase in concentration (a) and due to mechanical repulsion (b).

resulting dispersion of an average size is 5-10%, depending on the composition of the used semiconductor.

#### IV. Application of quantum dots

QDs have a great potential for practical application. First of all, this is due to the possibility of controlling the variation of the effective band gap width when resized. This will change the optical properties of the system: the wavelength of the luminescence, the absorption region. The second practical - an important feature of quantum dots - this is an opportunity to exist in the form of sols (solutions). This makes it easy to obtain coatings from quantum dot films by optimal methods, or apply quantum dots through inkjet printing on any surface. All of these technologies can be avoided when creating devices based on quantum dots of expensive vacuum technologies, traditional for microelectronic technology. Also, due to soluble technologies, you can enter quantum dots in the required matrix and create composite materials.

The possibility of varying the wavelength of luminescence is a fundamental advantage for the creation of new laser environments. In existing lasers, the wavelength of luminescence is a fundamental characteristic of the medium and the possibilities of its variation are limited (lasers with variable wavelength use the properties of resonators and more complex effects). Another advantage of quantum dots is the high photo stability versus organic dyes. QDs show the behavior of inorganic systems.

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Creating solar cells is one of the most promising areas for colloidal quantum dots. Currently, the highest conversion factor (up to 25%) has traditional silicon batteries. However, they are quite expensive and existing technologies do not allow the creation of large-sized batteries (this is too expensive production). In 1992, M.Gratzel [34] proposed an approach to the production of solar cells, based on the use of materials with a large specific surface (eg, nanocrystalline  $\text{TiO}_2$ ). Activation to the visible range of the spectrum is achieved by the addition of a photosensitizer (some organic dyes). QDs can perfectly act as a photosensitizer, since they allow you to control the position of the absorption band. Another important advantage is the high extinction coefficient (the ability to absorb a significant proportion of photons in a thin layer) and the high photostability inherent in an inorganic core.

The use of QDs arrays as conductive layers in microelectronics is very promising, since it is possible to use simple and cheap "solutions" with drawing technology. However, the possibility of application is limited at this time to an extremely high ( $\sim 10^{12}$  Ohm \* cm) resistance of layers from quantum dots [35]. One of the reasons is the large distance between the individual quantum dots, which is made using standard stabilizers such as trioctylphosphine oxide or oleic acid of 1 to 2 nm, which are too large for efficient tunneling of charge carriers.

Creating fluorescent labels based on QDs is very promising. One can distinguish the following advantages of QDs before organic dyes: the ability to control the wavelength of luminescence, high extinction coefficient, solubility in a wide range of solvents, stability of luminescence to the action of the environment, high photostability. It is also possible to note the possibility of chemical or even biological modification of the surface of quantum dots, which allows for the selective connection with biological objects [36].

Introduction to the production of chemo - and biosensors is the subject of intensive research in the field of quantum dots. In such sensors, QDs is an integral part of the system. The interest in QDs-based lasers was largely motivated by early theoretical works, which provided a number of their advantages over quantum wires, quantum wells, and bulk materials based on lasers: lower limit current density, higher temperature stability, higher gain. However, later detailed theoretical analysis showed that the advantages of lasers on QDs can be realized only with careful optimization as the characteristics of the ensemble of the points themselves (density, size distribution, depth of localization of the ground state, etc.), and the design of the laser (the thickness of the waveguide, profiles of doping, etc.). In this regard, the development of methods for controlling the characteristics of an array of QDs, achieving a deep understanding of the physical properties of

optoelectronic devices on QDs and optimizing their parameters is extremely important and topical task. In recent years, the research interest in InAs / GaAs semiconductor heterostructures with zero-dimensional intense nanosized InAs (quantum dots (QD)) has grown considerably [37,38,39]. Such nanoblocks have a high quantum yield of photoluminescence and is a promising material for the creation of lasers in the near infrared spectrum [40], which can be varied by the deformation parameter; p-n structures on a tight heterojunction QD-matrix; quantum computers [41] and a multi-mode laser based on quantum dots doped with various impurities.

Examples of sensors based on QDs that are fully operational in electrochemical mode are rarely encountered in the literature due to the difficulties associated with the electrochemical experiment of semiconductor nanocrystals. Thus, electrochemical sensors based on quantum dots are based on a nanocrystal fragment as an integral component capable of increasing the electrochemical signal rather than exploiting the electrochemical signal of a quantum dot for a read function. Such sensors are typically intended for target molecules that have biological activity.

Electrochemical neutron neutral detection sensors based on quantum dot CdTe have been described in [42]. The CdTe quantum dots were synthesized in water and subjected to further processing for the purpose of introducing terminal carboxyl groups onto the surface, which ultimately offers a point of deposition for the neutravidin compound.

The prepared quantum dots were activated with 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide and 4-hydroxysuccinimide, and finally mixed with neutravidin solution to reach the compound. The golden electrode was modified from the self-assembled cysteine monosaccharide subsequently biotinylated. QD-neutravidin compounds have been captured at the top of the modified electrode due to the ability of biotin to recognize neutravidin. The formation of the sensor node was controlled using experiments on impedance and atomic force microscopy measurements.

At the end, captured QDs were dissolved in an acid solution to cause the release of  $\text{Cd}_2^+$  ions, the amount of which was determined by anode inversion voltammetry. The dependence of the intensity of the peak on the concentration of the QD-neutravidin compound and on the degree of binding on the surface of the electrode was detected. In this system, QDs play a decisive role in amplifying the current signal, which was not observed in controlled experiments (in the absence of QD).

The selectivity of the sensor was demonstrated by competing analyzes with other proteins (peroxidase of chromium, hemoglobin and myoglobin) that do not lead to changes in the current strength observed for neutravidin, Fig. 9 The specificity of the sensor is based on the occurrence of specific and strong interactions between neutravidin and biotin (this coupled receptor-substrate has constant stability, high as  $10^{15}\text{m}^{-1}$ ). In addition, the sensor signal was proved linearly, ranging from 0.5 to 100 ng / l with a detection limit of up to 0.5 nM.

Light-controlled electrochemical sensor based on QD [43]. It consists of a semiconductor in combination

with an insulating layer. Due to the presence of the latter, a device of this type shows a very poor conductivity. However, the conductivity of the alternating current can be increased by lighting the device with modulated light, thereby creating electron-hole pairs. The irradiated portion of the surface of the device determines the active point where the photocurrent can flow. Using a point source of radiation provides high spatial resolution (several microns), despite the fact that the photocurrent flows also in the immediate periphery of the illuminated area, where electron-hole pairs are well generated.

In particular, QDs of different sizes and compositions coated with dithiol-ligand were immobilized on a gold electrode by incubation or spin-coating. The photocurrent was generated by locating the potential on the electrode and irradiating it with a focused light spot obtained from a common lamp and a lens. Measurement of the photocurrent was carried out using a potentiostat using a golden film as a working electrode. The effect of QD material and the surface of immobilization on photocurrent drifts, noise, and the effect of photocurrent lighting were tested in order to optimize the preparation of the device. Indeed, a strong dependence of sensory characteristics on the basis of quantum dots attached to the electrode was detected.

Amperometric biosensor [44]. was constructed by the functionalization of a glassy carbon electrode with several walls of carbon nanotubes adorned with golden nanoparticles. The modified CEE is further developed by covalent attachment of the quantum dots of CdTe with thiol coating, on which the enzyme methyl parathion has been immobilized. A golden element in carbon nanotubes was introduced, both for increasing the surface area and for catalysis of the enzymatic reaction, thus providing a synergistic effect on signal strength. In voltamperograms of methyl parathion, anode peak associated with the oxidation of p-nitrophenol, the product of hydrolysis, catalyzing the methyl parathion, is manifested. The anode peak increases with increasing the concentration of methyl parathion. The sensor shows a low detection limit (1.0 ng / ml) and high selectivity for the detection of organophosphorus compounds containing P-S bond.

An important feature of the sensor is the possibility of re-use, which makes it suitable in principle, for continuous monitoring.

The design of chemo - and biosensors is the subject of intensive research in the field of QD. In such QD sensors, they are connected to other components and, hence, they are an integral part of the system, or they are simply dissolved in the same medium of an analyte or other chemical substances necessary for the development of a biosensor signal. The first application of electrothermal generation of the chemical luminescence originating from quantum dots was reported in 2004 by Zou and Ju. CdSe nanocrystals were used which were deposited at the top of the paraffin-impregnated graphite electrode. The prepared sensor was used to detect hydrogen peroxide under reduced concentrations up to 0.1 pM. The operational mechanism of the sensor depends on the ability of the analyte to act as an electrochemiluminescent coagulant.

The principle of operation of semiconductor gas

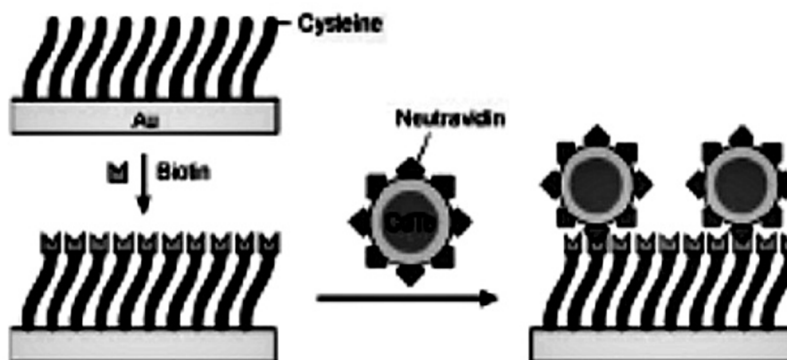


Fig. 9. Collection of electrochemical sensor based on quantum dots CdTe.

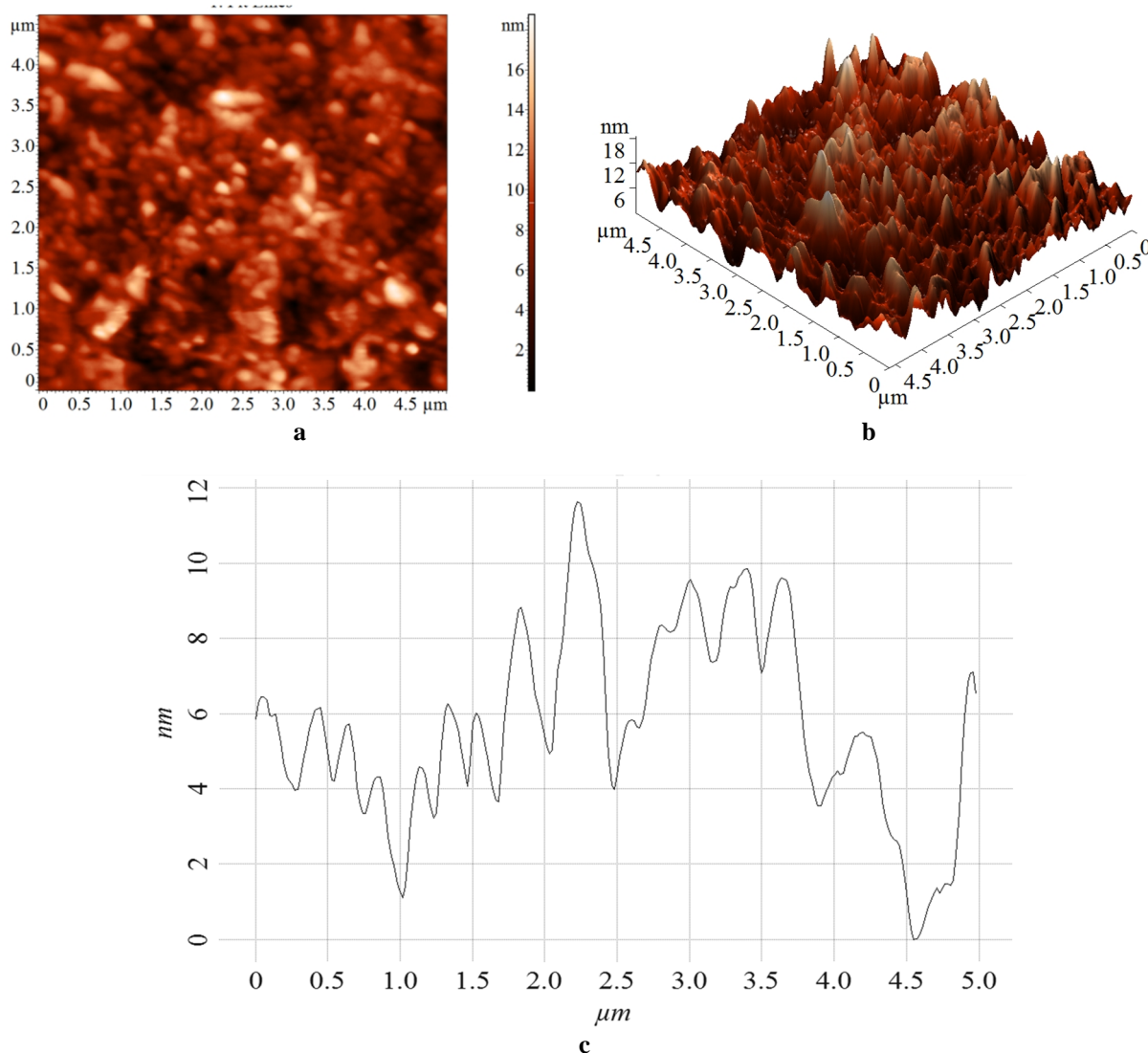


Fig. 10. ACM image of the received films ZnO a) 2D, b) 3D, c) profile of the surface.

sensors is based on the change in the electrical conductivity of a semiconductor gas-sensitive layer at the chemical adsorption on its surface of gases. This allows us to effectively use them to fire alarm devices and signaling devices in hazardous to human concentrations of toxic gases, where they may be considered as alternative devices alongside existing being optical, thermal and electrochemical signaling devices. The main

materials of sensitive elements of such sensors are semiconductor metal oxides [45], [46] and metal-containing organic polymers [47]. The listed materials have an acceptable sensitivity to changes in the composition of the ambient gas environment.

Among the wide range of metal oxides, as a material for gas sensors, zinc oxide (ZnO) is of particular interest [48]. The interest is due to the properties of zinc oxide,

such as chemical inertness, temporary stability, resistance to atmospheric influence, high sensitivity to toxic and explosive gases [48,49]. Different types of zinc oxide gas sensors are known: sensors on thick and thin films, nanoparticles and nanotubes [49]. It is shown that the sensitivity of the material is proportional to the ratio of surface area to its volume.

Nanocrystalline ZnO films were applied to the Sitall substrate by high-frequency reactive magnetron sputtering at the BOC Edwards Auto 500 [50]. Zn 99.999% purity target was used. Sputtering was carried out in an atmosphere of a mixture of oxygen and argon. The oxygen content in the gas mixture was 20, 50 and 80%; Magnetron discharge capacity - 100, 125 and 150 W; working pressure in the process of spraying - 0,2, 0,5 and 0,8 Pa; the preheating temperature varied. The thickness of the films obtained was maintained at  $60 \pm 4$  nm. The study of gas-sensitive characteristics of the films obtained was carried out on the bench for measuring the gas sensitivity. It consists of a chamber with the ability to pump gas and its ventilation, a temperature table that provides heating to a given temperature, and a control and measuring system.

Microscopic studies have shown that the resulting ZnO films are homogeneous, with the grain size value ranging from 50 to 250 nm.

Optical sensors include a nanophoton sensor based on quantum-sized detector structures for the determination of surfactants in water bodies of the environment. The principles of nanophotonics are realized in this device by applying ordered monomolecular layers of semiconductor QDs of the type CdTe, applied by the Langmuir-Blodgett method (LB) to the working electrode of the sensor. The basic principles of nanophotonics are the spatial constraints on nanoscale interactions of photons and electrons located in a quantum object. The solution of this problem is carried out by means of the corresponding Schrödinger equations for finding the wave functions and the energy value of the quantum system.

The nano-photon sensor as detector elements contains ordered monoshares of nanomaterials, which are applied with the help of the LB nanomaterial method, are QD-type CdTe.

The main advantages of QDs in their application as detectors in comparison with organic phosphors are their photochemical stability, narrow spectrum of radiation, optical and neoptic (at recombination of charge carriers) excitation, high quantum yield and intensity of luminescence, dependence of the spectrum of luminescence on the sizes of QDs. With a decrease in the size of the QDs core, the maximum wavelengths of absorption and radiation become smaller [51]. Thus, changing the radius of the QDs core, you can adjust not only the wavelength of the radiation, but also to identify other analysts.

For QDs, like for most organic molecules, oxidation and reduction processes are characteristic. Electrochemistry dissolved in a suitable QDs solvent is based on the transfer of charge (electron) between the electrode and the QD to form charged particles analogous to the ion radicals of organic molecules.

The recombination of the anion and cation radicals

of the given QD can lead to the formation of an electron-excited state of QD, the radiation deactivation of which represents the optical analytical signal of the sensor.

For a correct choice of the size of QD, you can use the quantum mechanical method of calculation using the semi-empirical extended Hartree-Fock method with the help of mathematical and quantum-chemical programs.

The developed nano-photon sensor device is a flow cell with two electrodes - the working and auxiliary. The working electrode represents an optically transparent glass substrate, which is modified to provide electrical conductivity with a transparent conductive coating of  $\text{In}_2\text{O}_3$ . At last, an ordered monomolecular layer is applied by means of the LV method to the semiconductor QD. Modification of the working electrode QD not only significantly reduces the amount of consumable reagents, but also enables the reusable use of the sensor device.

Magnetic sensors today are used in many technical systems, including modern anti-tank missiles. Information carriers built on the basis of magnetoresistive structures are resistant to temperature fluctuations, ionizing radiation and radiation, and therefore promising for use in microprocessor systems for guiding modern warheads. Magnetic field anomaly sensors are also used in marine border control systems because in the water space the perturbation of a magnetic field can not be random in nature, but is due to the appearance of large ferromagnetic objects that move in a nonmagnetic environment.

Magnetic sensors numerically register these perturbations of the background magnetic field of the Earth, and modern methods of digital processing of analog signals allow to determine with a fairly high accuracy the mass, direction and speed of the aforementioned objects [52].

Magnetoresistive structures - objects that have the ability to change their volt-ampere characteristics, depending on the change of the external magnetic field. Sensors based on magneto-resistive structures have high sensitivity to magnetic field changes.

Modern biosensors are analytical devices in which high-specific chemical reactions are used to determine the components of the sample, are catalyzed by enzymes, immunochemical reactions or reactions occurring in organelles, cells or tissues. The main function of the biosensor is to convert highly specific biochemical processes into an analytical signal captured by the detector and serves as a source of information on the qualitative and quantitative content of one or more bioanalysts in the test sample. There are several classifications of biosensors in various features.

There are two main classes of biosensors - enzyme and affinity biosensors. Affinity biosensors include DNA-sensors and immunosensors. The main difference lies in the nature of the analytical signal. In the enzyme biosensors, the analytical signal reflects the rate of conversion of the substrate under the action of the enzyme and reaches a maximum in the first minutes of the reaction. After that, the intensity of the signal is sharply reduced. The basis of the affine biosensors is the reversible biochemical reactions of the type "antigen-antibody" and "DNA-protein". In this case, the maximum analytical signal is observed when the biochemical

response is reached.

Enzyme sensors - one of the most common types of biosensors, with a unique structure and properties of enzymes - selective catalysts of biological processes.

There are 2 groups of enzyme biosensors: catalytic (substrate) and inhibitor [53]. Catalytic or substrate biosensors are based on the direct interaction of an enzyme immobilized on a substrate or plate, and a substrate to form a reaction product, dividing energy and a low molecular weight substance (for example, hydrogen peroxide). In this case, one or more analytical signals and methods of their detection can be identified. The principle of action of inhibitory biosensors is to inhibit the activity of the enzyme by the substrate. An analytical signal in this case is the degree of decrease in the activity of the enzyme (degree of inhibition). Inhibition of the activity of the enzyme can be as specific due to interaction with the substrate, and non-specific because of the presence of strong acids, bases, organic solvents in the sample, as well as the effects of high temperature, radiation, etc.

The main problem with the development and use of enzyme sensors is the immobilization of the enzyme on the surface of the transducer. Many enzymes are extremely labile and physical sorption is often not enough for strong containment. Chemical methods of immobilization can lead to partial or complete loss of enzyme activity [54]. The use of nanomaterials based on carbon [55] or metals [56] due to the developed surface and low resistance allows more firmly fix the enzyme on the substrate and improve the sensitivity of the sensor. Difficulties are also possible in the analysis of multicomponent systems, since such admixtures in the matrix as heavy metals, some organic compounds can also cause a decrease or complete disappearance of the catalytic activity of the enzyme.

Bicatalytic enzyme sensors based on the detection of an integral index of enzyme activity and catalytic activity of a nonbiological component in relation to a specific component, can significantly improve the sensitivity of detection [57].

One of the modern trends in the development of enzyme sensors is the development of biosensors based on inorganic catalysts and enzyme-like complex compounds. The authors [58] developed a catalytic non-enzyme sensor based on organic complex compounds of nickel to determine creatinine. The signals of the oxidation of certain components were amperometrically recorded.

DNK sensors. A distinctive feature of this group of biosensors is the use as a biological component of a DNK fragment. The principle of action of DNK sensors is based on the complementary interaction between single-stranded DNK sections of the component of the analyzed sample and the receptor of a known structure.

The most promising among biosensors for the identification of bacterial pathogens are sensors and methods based on the use of immunoreactive reactions.

For use in the bioanalysis, the most commonly used nanoparticles consist of an "inorganic" core and a polymeric membrane. Inorganic material that exhibits

magnetic properties often acts as a nucleus, and a polymeric membrane, modified by various functional compounds, allows for a signal from a nanocomposite. In order to obtain such nanocomposites, physical encapsulation is generally used.

This method consists of one-step or sequential adsorption of a polymer on a charged nanoparticle. Adsorption is mainly regulated by electrostatic interaction processes and is realized by means of charged groups or specific interactions (eg, ion-ion interaction, complex formation, dipole-dipole interaction, etc.). The main purpose of this method is to physically involve inorganic nanoparticles inside a charged polymer, which ultimately forms a shell of nanocomposite particles. Sometimes they conduct a multi-stage layered coating process. In this way, "quantum dots" are often encapsulated, for example, by covering the nanoparticles with a polysaccharide, by electrostatic interaction [59].

Polymerization is one of the most widely used methods for coating nanoparticles on the surface. In the process of microemulsion polymerization, to obtain identical submicron droplets of the monomer, which are converted into stable colloidal particles, intensive phase mixing takes place. The monomer in most cases is a lipophilic substance dispersed in an aqueous solution of surfactant. Large surface area of small droplets of microemulsion (submicron sizes) increases their ability to capture growing radicals that are generated by the initiator used. Consequently, each drop is converted into a micro-reactor in which polymerization takes place. At the final stage, each drop is converted into a solid particle. Unlike conventional emulsion polymerization, in a microemulsion there is no diffusion of monomer (s) through a continuous disperse phase, which makes it possible to obtain mono-size particles. A large number of papers are devoted to the coating of inorganic nanoparticles by polymer using radical microemulsion polymerization. The literature describes a similar coating of nanoparticles: titanium oxide ( $TiO_2$ ), calcium carbonate ( $CaCO_3$ ), coal tar particles, ZnO, quantum dots (CdSe / CdS) and iron oxide ( $Fe_3O_4$ ) [60].

Shotber's method is one of the simplest methods that allows obtaining a durable coating on the surface of the nanoparticle with a wide range of possibilities for functionalization and further modification. Simplicity of synthesis also plays an important role in attracting the attention of researchers to this class of materials. The advantages of the method [61], which allowed him to be widely used in the synthesis of nanocomposites, in particular, include:

- the possibility of creating different structures with an ultrafine phase;
- ensuring high purity of both the source material and the resulting product;
- the homogeneity of the distribution of components (including small modifying additives) and, as a consequence, the possibility of reducing the micro-heterogeneity of the formed compounds to the molecular and ionic levels;
- the possibility of obtaining new crystalline and amorphous phases, materials with cations in their



unusual degrees of oxidation, the synthesis of which by traditional methods is difficult or impossible.

Heterostructures with spatial constraints of charge carriers in all three directions (quantum dots) realize the limiting case of dimensional quantization in semiconductors, when the modification of electronic properties of a material is most pronounced. The electron spectrum of an ideal quantum dot (QD) is a set of discrete levels separated by areas of forbidden states and corresponds to the electron spectrum of a single atom, although the real quantum dot can thus consist of hundreds of thousands of atoms.

## Conclusions

A review is made of the practical application of semiconductor nanoparticles as materials for different types of sensors on their basis, different methods of obtaining quantum dots and their properties, depending on the synthesis conditions, have been shown that quantum-dimensional effects play a key role in optoelectronic properties of QD. The energy spectrum of

QD is fundamentally different from the spectrum of a bulk semiconductor. Materials possessing controlled luminescence, in particular on the basis of semiconductor nanocrystals, are promising as chemical sensors, for fluorescence microscopy, as components of information processing and storage systems.

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## **Напівпровідникові квантові точки як матеріали для лазерів на їх основі**

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Квантові точки (КТ), на сьогодні належать до центральних об'єктів досліджень багатьох наукових груп. Вивчення властивостей структур малих розмірів важливе як для подальшого розвитку електроніки, так і для удосконалення вже існуючих напівпровідникових приладів. На сьогодні є багато методів отримання КТ в лабораторних умовах: метод ультразвукового подрібнення, молекулярно-променевої епітаксії, імпульсної лазерної абляції, а також за допомогою таких методів хімічного синтезу, як металорганічний синтез, синтез у зворотніх міцелах, сольвотермальний синтез, золь-гель синтез, синтез з використанням тіол-стабілізаторів, синтез у неводному середовищі. Найбільш перспективними на даний момент є методи, засновані на використанні явища самоорганізації. Це методи молекулярно-променевої епітаксії та колоїдної хімії, причому останнім методом можна синтезувати кристали, які мають розмір декілька нанометрів і в яких ширина забороненої зони та енергія максимуму піку люмінесценції, визначаються розміром частинок.

**Ключові слова:** квантові точки, гідротермальний синтез, сольвотермальний синтез, напівпровідникові квантові точки, квантово-розмірний ефект.