

Optics of colloidal quantum-confined CdSe nanoscrolls

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Abstract. Nanostructures in the form of 1.2-nm-thick colloidal CdSe nanoplatelets rolled into scrolls are investigated. The morphology of these scrolls is analysed and their basic geometric parameters are determined (diameter 29 nm, longitudinal size 100–150 nm) by TEM microscopy. Absorption and photoluminescence spectra of these objects are recorded, and the luminescence decay kinetics is studied. It is shown that the optical properties of CdSe nanoscrolls differ significantly from the properties of CdSe quantum dots and that these nanoscrolls are attractive for nanophotonic devices due to large oscillator strengths of the transition, small widths of excitonic peaks and short luminescence decay times. Nanoscrolls can be used to design hybrid organic–inorganic pure-color LEDs with a high luminescence quantum yield and low operating voltages.

Keywords: nanophotonics, colloidal semiconductor nanostructures, nanoscrolls and nanoplatelets, absorption and photoluminescence spectra, luminescence decay kinetics.

1. Introduction

To solve a number of fundamental and applied problems of nanophotonics, optoelectronics and subwave optics, one must know the optical properties of new composite nanostructures and nanomaterials and the effects of their interaction with light fields. Currently, various hybrid nanostructures and materials are used to design solar cells [1–3], nanolasers [4–6], photonic switches [7, 8], optical tweezers [9] and sensors [10–11]. Composite materials are applied in optical nanowaveguides with metal coating [12–15], hybrid plasmonic waveguides [16, 17], and tapered metal-coated near-

field optical probes with dielectric [18–22] and semiconductor [23–26] cores, which make it possible to form nanoscale-localised light fields.

The study of the optical properties of hybrid nanostructures, containing both organic and inorganic components (see review [27]), is an independent line of research in nanophotonics and optoelectronics. A large number of experimental and theoretical studies have been devoted to the spectral characteristics and effects of interaction of light with two- and three-layer metalorganic nanostructures of different shapes and sizes, consisting of a metal core and an external shell formed by ordered molecular J aggregates of cyanine dyes [28–35]. Organic–inorganic materials and structures of another type, which are currently intensively studied, are based on organic transport layers and a layer of colloidal semiconductor nanocrystals of different shapes and sizes. The interest in these hybrid materials and structures is related to a great extent to the necessity of developing efficient organic–inorganic light-emitting devices for energy-efficient and stable light sources, designed for illumination, indication, and imaging.

These devices are based on composite layers, consisting of a semiconductor organic matrix with inorganic semiconductor (CdSe, CdS, CdSe/ZnS, CdSe/CdS, ZnSe/CdSe/ZnS, etc.) nanocrystals of different shape and size, incorporated into it. Varying the shape and size of these nanocrystals, one can change their luminescence properties. When an electric current is transmitted through a diode structure, carriers pass through organic layers to interact with nanocrystals; as a result, the latter become radiative recombination centres. The advantages of semiconductor colloidal nanostructures are their relatively narrow luminescence bands and high quantum yield; in addition, light-emitting devices based on these nanostructures are characterised by high stability and durability.

To date, light-emitting diodes (LEDs) based on organic materials and spherical nanocrystals have been developed. These nanocrystals have either a simple structure, i.e., a single core (for example, CdZnSe), or a composite structure based on a core coated by one or several shells of other semiconductor materials (CdSe/ZnS, CdSe/CdS, ZnSe/CdSe/Zn, etc.) [36–42]. A study of the electroluminescence in hybrid organic–inorganic LEDs was also performed for tetrapod CdSe/CdS nanocrystals [43].

The development of colloidal synthesis techniques in the recent years has led to the formation of a new class of semiconductor nanocrystals (CdSe, CdS and CdTe nanoplatelets) and planar heterostructures (CdSe/CdS and CdSe/CdZnS), grown on their basis [44–47]. The thickness of these platelets can be controlled with accuracy of one atomic layer. These planar nanostructures are characterised by a quantum-con-

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finement effect, which is similar to that observed in epitaxial semiconductor quantum wells. Advantages of colloidal nanoplatelets are high radiative recombination rates and narrow luminescence bands (due to the giant oscillator strength of radiative transition [48] and highly uniform thickness of nanoplatelets). The band full widths at half maximum (FWHMs) $\Delta\lambda$ at $T = 300$ K for one-component nanoplatelets (CdSe, CdS, and CdTe) and composite nanoplatelets consisting of a core and shells (e.g., CdSe/CdZnS) are, respectively, 5–10 and 20 nm [44, 45].

A possibility of applying nanoplatelets as active emitting elements in organic LEDs has been recently demonstrated by us in [49–51], where a hybrid LED based on one-component CdSe nanoplatelets, emitting at wavelength $\lambda = 515$ nm, was devised. A LED based on two-component CdSe/CdZnS nanoplatelets, emitting at wavelength $\lambda = 650$ nm, was developed in [52].

In this study, an original technique of fabricating colloidal nanostructures in the form of nanoscrolls (rolled semiconductor colloidal CdSe nanoplatelets with a thickness of few atomic layers) is proposed and implemented. In our experiments, platelets were rolled into scrolls when the lateral platelet size exceeded 100 nm. The geometric and physical properties of these quantum-confined structures are of great interest. We analysed their morphology and determined the size of synthesised CdSe nanoscrolls (Fig. 1). Their spectra were recorded and the photoluminescence decay kinetics was investigated at room temperature.

The use of these CdSe nanoscrolls as a flat inorganic layer (emitter) is expected to allow one to develop pure-colour light-emitting devices, operating in the short-wavelength visible spectral region at low operating voltages. Another advantage of the obtained CdSe nanoscrolls is that they can poten-

tially be used to store organic molecules, clusters and quantum dots (QDs) (as carbon and boron nitride nanotubes of smaller size were proposed to store hydrogen, methane and carbon dioxide molecules [53, 54], in particular, to solve problems of hydrogen power engineering). Since the synthesis of colloidal semiconductor nanoscrolls is fairly simple and inexpensive, they can become a serious alternative to other possible screw and tubular structures.

2. Structure and size of nanoscrolls

Colloidal CdSe nanoscrolls were obtained using the technique described in [49, 50], which was somewhat upgraded. The difference is that the selenium precursor was a 1-M solution of selenium in trioctylphosphine (TOP–Se) rather than a 0.1-M solution of selenium in octadecene (Se–ODE). The replacement of the selenium precursor in the synthesis of quasi-two-dimensional CdSe nanostructures makes it possible to significantly increase their longitudinal size (up to 150 nm, in contrast to the previously synthesised nanoplatelets with a longitudinal size of 50 nm; see [49, 50]). It is energetically favourable for thin (of few atomic layers) planar structures with such a large longitudinal size to roll into a tube, as a result of which CdSe nanoscrolls are formed.

A study of the synthesised nanoscrolls with a Carl Zeiss LEO 912 AB Omega transmission electron microscope (TEM) (see Fig. 1b) made it possible to determine their average size: $D = 29$ nm and $d = 23$ nm (outer and inner scroll diameters, respectively); $L = 100$ nm (scroll length); $h = 3$ nm (distance between scroll neighbouring layers); $A = 100$ – 150 nm and $B = 100$ – 150 nm (longitudinal sizes of the platelets forming a scroll); and $l = 1.2$ nm (platelet thickness). The TEM analysis showed that the average number of turns in a scroll is 2.5.

3. Absorption and luminescence spectra

Light absorption spectra of solutions of CdSe nanoplatelets and nanoscrolls in hexane were recorded using a PerkinElmer Lambda 45 spectrophotometer. Measurements were performed in a quartz cell filled with the solution; the optical path length was 5 mm. When recording photoluminescence spectra of nanoplatelets and nanoscrolls, a sample (glass substrate with nanostructures deposited on it from a solution in hexane) was excited by a PicoQuant 800-B pulsed semiconductor laser with a wavelength of 405 nm, a pulse repetition rate of 40 MHz and a pulse duration of 75 ps. An Ocean Optics Maya Pro 2000 spectrometer was used as a detector. Luminescence decay curves were recorded using a PicoQuant PMA-C 192-N-M photoelectron multiplier with a monochromator and a PicoQuant TimeHarp-100 single photon counting system.

Figure 2 shows the absorption spectra of hexane solutions of synthesised CdSe nanoscrolls, obtained from nanoplatelets with a large longitudinal size, and flat CdSe nanoplatelets with a smaller characteristic lateral size. Both nanostructures exhibit characteristic excitonic peaks with energies of 2.85 ($\lambda = 435$ nm) and 2.69 ($\lambda = 462$ nm) eV, which are due to the transitions from the subbands of light (lh) and heavy (hh) holes in the valence band to the conduction band. Note that the amplitude of the lh peak is somewhat reduced (by $\sim 5.5\%$), and the area under this peak is changed by approximately 10%. This fact indicates that rolling of nanoplatelets into nanoscrolls slightly reduces the oscillator strength of the

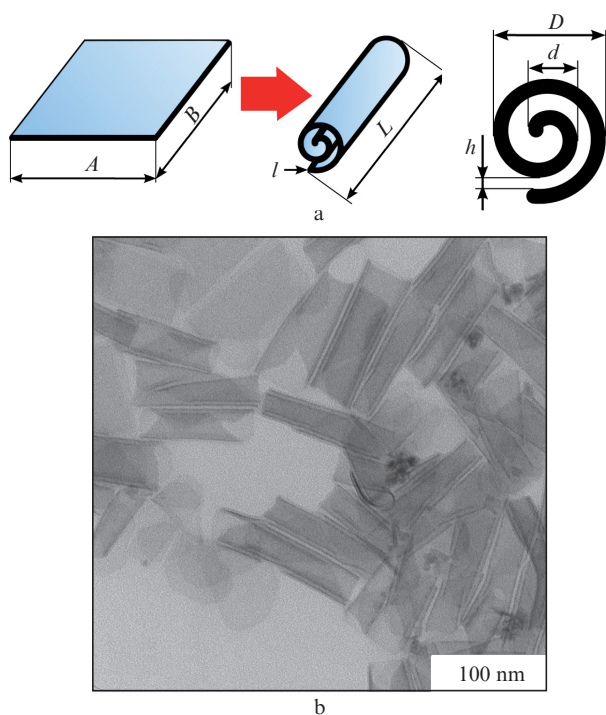


Figure 1. (a) Schematic presentation of rolling of nanoplatelets into nanoscrolls and their geometric parameters ($A, B = 100$ – 150 nm, $L = 100$ nm, $l = 1.2$ nm, $D = 29$ nm, $d = 23$ nm and $h = 3$ nm) and (b) TEM images of synthesised CdSe nanoscrolls.

corresponding transition. Nevertheless, comparison of curves (1) and (2) in Fig. 2 shows that rolling of nanoplatelets with a large longitudinal size (100–150 nm) into nanoscrolls does not radically change their absorption spectra in comparison with rectangular nanoplatelets having a characteristic longitudinal size of 20–70 nm. This is explained by the fact that the optical properties of these objects are primarily controlled by the quantum confinement in the transverse direction (i.e., along the thickness of a platelet or one scroll layer), whereas the transverse size is practically the same for the nanoscrolls and nanoplatelets under study. Figure 2 shows also the absorption spectrum of spherical CdSe QDs with a radius $r = 2.1$ nm [curve (3)], obtained in [55]. A comparison shows that the exciton absorption bands of nanoscrolls are much narrower than those of spherical nanocrystals, which are now widely used to design nanophotonic and optoelectronic devices.

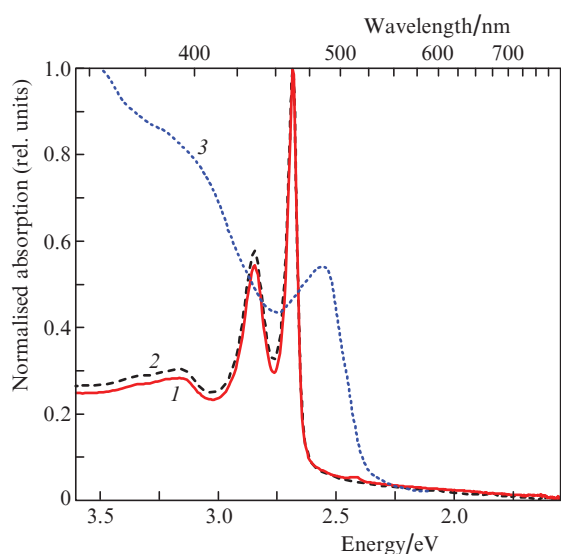


Figure 2. Light absorption spectra of solutions of (1) CdSe nanoscrolls and (2) CdSe nanoplatelets in hexane and (3) absorption spectrum of CdSe QDs ($r = 2.1$ nm) according to the data of [55].

Figure 3 shows photoluminescence spectra of CdSe nanoscrolls and corresponding nanoplatelets at $T = 300$ K. The interband luminescence peaks correspond to energies of 2.601 eV (477 nm) for nanoscrolls and 2.634 eV (471 nm) for nanoplatelets. An estimate of the luminescence bandwidths shows that $\Delta E \approx 140$ meV ($\Delta\lambda \approx 25$ nm) for nanoscrolls and ~ 110 meV (20 nm) for nanoplatelets. Thus, the interband luminescence spectrum of nanoscrolls is red-shifted with respect to nanoplatelets by $\delta E \approx 33$ meV (~ 6 nm). This shift can be explained by the influence of the internal electric field, caused by the stress arising in bent nanoplatelets when they roll into nanoscrolls. In addition, the change in the geometry of a quasi-two-dimensional structure from planar to rolled may somewhat modify the positions of energy levels in the potential well and, therefore, cause a red shift of the luminescence spectrum. In turn, the widening of the interband luminescence band of nanoscrolls in comparison with nanoplatelets is explained by the fact that it is overlapped to a greater extent with the luminescence band of defects than in the case of nanoplatelets.

For comparison, Figure 3 shows a photoluminescence spectrum of spherical CdSe QDs with a radius $r = 2.1$ nm

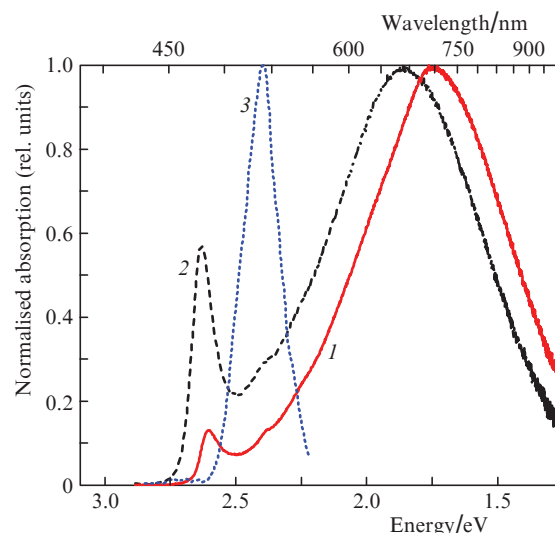


Figure 3. Photoluminescence spectra of (1) CdSe nanoscrolls and (2) CdSe nanoplatelets and (3) photoluminescence spectrum of CdSe QDs ($r = 2.1$ nm) according to the data of [55].

[curve (3)], taken from [55]. The interband luminescence peak corresponds to the energy $E = 2.4$ eV (517 nm), and the interband luminescence bandwidth is $\Delta E = 170$ meV ($\Delta\lambda = 40$ nm). Thus, the interband luminescence spectra of nanoscrolls and nanoplatelets are much narrower than for spherical nanocrystals. Therefore, quasi-two-dimensional colloidal nanocrystals of helical or planar geometry are more favourable for solving fundamental and applied problems of nanophotonics than spherical or tetrapod QDs, as narrow luminescence bands are needed for such purposes.

Figure 3 shows also that the interband luminescence intensity of nanoscrolls is significantly reduced as compared with nanoplatelets. The reason is that rolling of nanoplatelets is accompanied by a charge redistribution at the opposite facets of the scroll layer because of the change in the distance between neighbouring atoms, which may lead to luminescence quenching. In addition, the deformation of nanoplatelets caused by their rolling may lead to the formation of various defects on their surface (for example, vacancies, which can play the role of traps for photoexcited carriers). Along with the interband luminescence peaks in the vicinity of photon energies of 2.7–2.8 eV, there are wide photoluminescence bands on the right from them for both nanoscrolls and nanoplatelets (Fig. 3); these bands are due to the presence of defects. They are peaking at energies $E = 1.74$ eV (694 nm) for nanoscrolls and 1.86 eV (640 nm) for nanoplatelets. The FWHM values for these bands are, respectively, $\Delta E = 660$ meV ($\Delta\lambda = 254$ nm) and 690 meV (242 nm). Thus, the luminescence bands due to defect states are also red-shifted.

4. Photoluminescence decay kinetics

We obtained decay curves for the photoluminescence intensity I in the corresponding interband luminescence peaks ($\lambda = 477$ and 471 nm) at $T = 300$ K for the synthesised CdSe nanoscrolls with a longitudinal size of 100–150 nm and CdSe nanoplatelets with a longitudinal size of 50–70 nm (Fig. 4). The results for nanoscrolls and rectangular nanoplatelets almost coincide [they are presented by curve (1) in Fig. 4]. This means that rolling platelets into scrolls with an increase

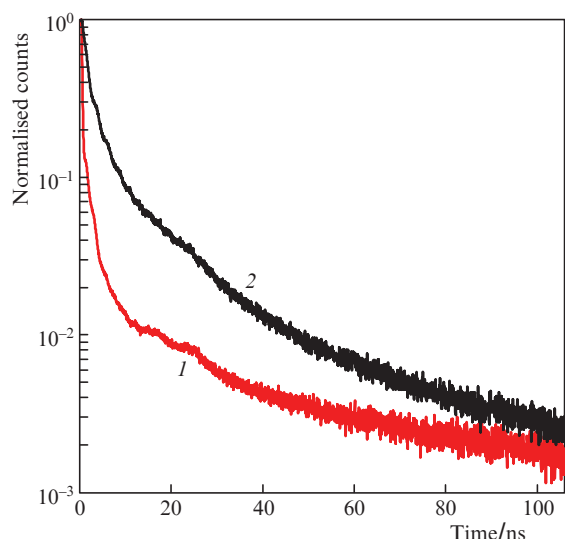


Figure 4. Photoluminescence decay curves in the interband luminescence peaks for (1) CdSe nanoscrolls and (2) CdSe QDs at $T = 300$ K. The decay curve for CdSe nanoplatelets is identical to curve (1).

in their longitudinal size does not change significantly the relaxation dynamics of excited states.

It is of great interest to compare the character of the luminescence decay of the CdSe nanoscrolls under consideration with the corresponding behaviour of the luminescence of spherical CdSe nanocrystals. To this end, Fig. 4 presents luminescence decay curves for CdSe QDs in the maximum of the exciton peak ($\lambda = 586$ nm). An analysis of the decay curves shows that the experimental dependences $I(t)$ within the entire time interval under study, $t \in [0; 100$ ns], are far from exponential in both cases. Nevertheless, to describe the initial stage of luminescence decay for CdSe nanoscrolls and CdSe QDs, we estimated the characteristic values of their time constants ($\tau_{\text{scr}} = 1.6$ ns and $\tau_{\text{QD}} = 3.5$ ns), the use of which allows one to adequately describe the behaviour of curves (1) and (2) at $t \lesssim 10$ ns, by a simple relation: $I(t)/I_0 \propto \exp(-t/\tau)$. Constants τ_{scr} and τ_{QD} differ by more than a factor of 2, which indicates a significant difference in the characteristic photoluminescence decay times for nanoscrolls and QDs in the initial relaxation stage. Moreover, the analysis of the results suggests that the quasi-two-dimensional CdSe nanocrystals synthesised by us have the smallest time constants (which determine the photoluminescence decay kinetics) among all known types of colloidal semiconductor nanocrystals, including spherical [42] and tetrapod [56] QDs.

5. Conclusions

Colloidal semiconductor nanoscale structures (CdSe nanoscrolls) were fabricated and studied. Their morphology was investigated, sizes were determined and photoluminescence spectra and kinetics were analysed. It was found that, at a sufficiently large longitudinal size of CdSe nanoplatelets ($A, B \gtrsim 100$ nm), their synthesis is accompanied by the formation of nanoscrolls rather than thin flat rectangular nanostructures. It was demonstrated that these nanoscrolls have optical properties attractive for various applications and possess certain advantages over colloidal semiconductor QDs for solving complex fundamental and applied problems of nanophotonics. These advantages include, in particular, large oscillator

strengths of the radiative transition, small widths of luminescence spectra and short luminescence decay time. This circumstance makes CdSe nanoscrolls promising elements of optoelectronic devices. They can be applied, for example, as emitters in hybrid organic–inorganic LEDs, operating in the short-wavelength part of the visible spectrum and characterised by pure colour, high external quantum yield and low operating voltages.

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