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Photoluminescence of CdTe colloidal quantum wells in external electric field

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ABSTRACT

We investigated emissive properties of CdTe colloidal quantum wells (CQWs) in applied electric fields in the range of 0–90 kV/cm. Photoluminescence spectra and time-resolved intensities of the excitonic peak were studied. The emission was centered at 2.47 eV with a width of 40 meV. No visible broadening or redshifts were detected in the whole range of applied fields. We obtained the dependencies of integrated intensity and maximum intensity of interband luminescence on the magnitude of applied electric field with the decrease of these characteristics by 15–20% at 90 kV/cm due to field-controlled charge carrier separation. Deceleration of the radiative decay rates with increasing electric field provides the evidence of field-controlled temporary exciton storage as two-fold increase of the number of photons registered in the 1–40 ns time range is observed. This assertion is also confirmed by the growth of the luminescence integrated intensity in the electric fields in the range 30–60 kV/cm. These findings are an important step toward understanding of the influence of electric fields on the operation of hybrid organic-inorganic LEDs with CQW-based emissive layers.

1. Introduction

Since their first successful synthesis [1] spherical colloidal semiconductor nanocrystals (or quantum dots, QDs, 0D nanocrystals) have attracted enduring interest due to their superior optical properties and chemical stability [2–4]. Further advances in colloidal chemistry led to the appearance of the particles with various shapes, i.e. nanorods [5] and nanowires [6] (1D nanocrystals), branched nanocrystals (tetrapods, octapods and etc.) [7,8] and, finally, nanoplatelets (NPLs) or quantum wells [9] (2D nanocrystals) which were later shown to form nanoscrolls [10,11]. Colloidal quantum wells were demonstrated to be the fastest emitters among semiconductor nanocrystals at nanosecond time scales [12] with the most narrow emission lines of about 10 nm [9,12]. They found application for fabrication of different devices such as light-emitting diodes [13–15], lasers [16,17] and photodetectors [18] alongside with spherical quantum dots [19] and tetrapod-shaped nanocrystals [20].

Operation of the devices based on semiconductor nanocrystals sets a number of fundamental problems, such as radiative and non-radiative energy transfer between the components of the devices [21], influence of interfacial and environmental phenomena, charging of the nanocrystals and the impact of electric field on their emissive properties [22,23]. The understanding of all these effects is essential for designing stable and efficient devices as photoluminescence quantum yields of colloidal nanocrystals reach 90% in solutions [24] but still quantum efficiencies of QD-based LEDs do not surpass 20% [25]. The influence of electric field was addressed in several works. Namely, in [26] CdSe/CdS core-shell quantum dots were studied in capacitive structures. Field-dependent luminescence lifetime measurements as well as tight-binding calculations revealed strong quenching of the luminescence quantum yield with significant charge carrier separation and reduction of the overlap integrals between electron and hole wavefunctions. This led to a substantial decrease of radiative decay rates. Absorption and luminescence of CdSe quantum dots and anisotropic CdSe quantum rods were investigated by Gurinovich et al. [27]. Strong luminescence quenching along with significant broadening of excitonic absorption and emission lines were observed. In addition, distinguishable redshifts associated with quantum-confined Stark effect (QCSE) were detected. Selective influence of electric field on luminescent properties (S- and

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P-polarized luminescence) of anisotropic nanocrystals was demonstrated. It was established that 0D nanocrystals are more sensitive to the impact of external electric field than 1D ones. Emissive properties of CdTe quantum dots were investigated by Ohshima et al. as well [28]. Photoluminescence spectra of the nanoparticles revealed significant quenching of photoluminescence in the presence of electric fields. Additionally, photoluminescence decay measurements demonstrated that this quenching was due to field-induced decrease in lifetime and initial population of the exciton-emitting state. Field-dependent absorption of 0D, 1D and 2D CdSe nanocrystals was considered by Achtstein et al. [29]. It was shown that the absorption of 2D nanocrystals is very sensitive to the influence of DC electric field with strong reduction of the absorption efficiency within the lowest-energy band due to QCSE and Franz-Keldysh effect. Interestingly, no redshifts were observed in the absorption spectra of 2D nanocrystals. The nanorods were confirmed to have the weakest DC response among the nanocrystals with different geometries as well.

Here we perform an investigation of luminescent properties of CdTe colloidal quantum wells under exposure to DC electric field inside capacitive structures. The corresponding information on the behavior of luminescence spectra and luminescence decays would be an important step toward improvement of the characteristics of CQW-based devices and understanding of the basic optoelectronic properties of 2D objects.

2. Experimental

The synthesis of CdTe colloidal quantum wells was conducted according to the modified protocols given in [30]. Morphology of the synthesized CQWs was studied with the use of a JEOL JEM-2100 transmission electron microscope (TEM). To study optical properties of the quantum wells we embedded them into a capacitive structure similar to the one described in [26–29] with polystyrene (PS) serving as an insulator matrix. We prepared two parts with [glass+ITO substrate/PS/CdTe/glue film] and [glass+ITO substrate/glue] structures which were joined together to form the experimental sample (Fig. 1). PS layers and CdTe CQWs were deposited by spin-coating. The sample was connected to a Tesla TV2 constant voltage source. The average gap between ITO contacts was measured with the use of a micrometer and was equal to 35 μm. Photoluminescence spectra were recorded with the use of an Ocean Optics Maya Pro 2000 spectrometer with a PicoQuant 800-B pulsed diode laser (emission wavelength: λ = 405 nm; repetition rate: ν = 40 MHz; pulse duration: $\Delta\tau$ = 75 ps) acting as the excitation source. Luminescence decay measurements were performed with a PicoQuant PMA-C 192-N-M photoelectron multiplier coupled to a monochromator and a PicoQuant TimeHarp-100 single photon counting system with the same excitation source at 100 kHz repetition rate.

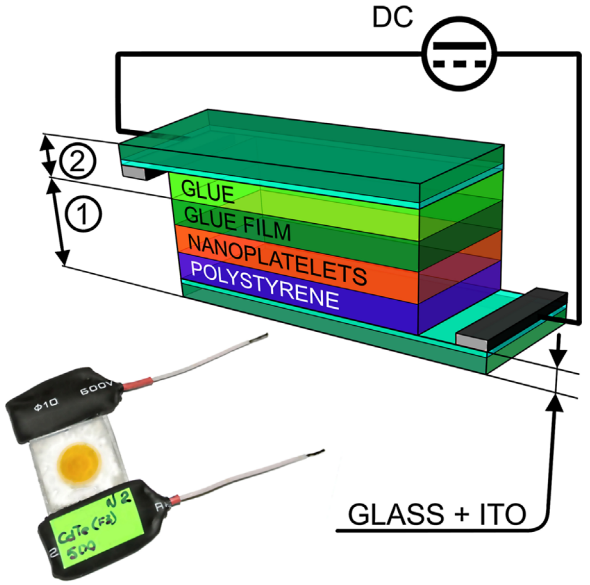

Fig. 1. Schematic view of the sample: two sandwiched structures (glass/ITO/polystyrene/glue film (1) and glass/ITO/glue (2)) are joined together to form the experimental sample, which is connected to a DC power supply. A photograph of the structure is given in the inset.

3. Results and discussion

3.1. Morphology

TEM studies (Fig. 2) show that the synthesized CdTe CQWs have a rather regular rectangular shape with the characteristic lateral size of about 100 nm. The thickness of the nanoparticles could not be obtained with the use of the images as the nanocrystals lie flat on the TEM grid. Nevertheless, spectroscopic measurements allowed us to ascribe the thickness of 1.9 nm to the synthesized 2D nanocrystals as an unambiguous correlation between the thickness and emission (absorption) maxima was established in [30] for different populations of CdTe CQWs.

3.2. Photoluminescence spectra

Photoluminescence (PL) spectra of CdTe CQWs are presented in Fig. 3. We observe a clearly distinguishable peak with the maximum at 2.47 eV. This peak is associated with interband luminescence and corresponds to the 1.9-nm-thick CdTe colloidal quantum wells [30]. Additionally, we observe broadening in the wings of the spectra, which could be attributed to phonon-assisted processes [31,32]. Visible quenching of the photoluminescence intensity occurs with increasing the electric field from 0 to 90 kV/cm (Fig. 3). With diminishing the electric field we find that PL intensity is restored with a slight increase of its maximum at 0 kV/cm compared with the initial intensity (see the lower inset in Fig. 3). The electric field did not induce any visible broadening or redshifts of the spectra.

Differential photoluminescence spectra $\Delta I = I(0) - I(F)$ in Fig. 4 demonstrate that about 20% of the PL intensity is lost due to field-induced quenching in the case of the highest applied field (90 kV/

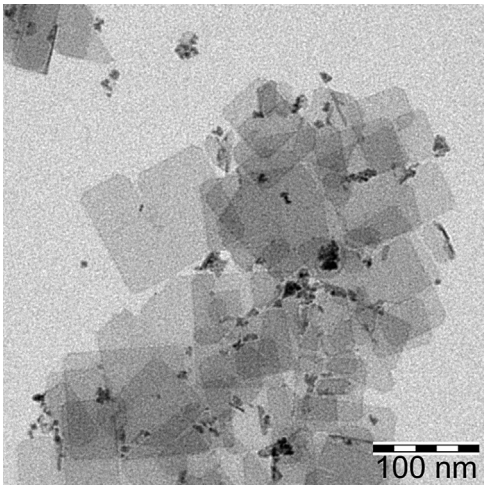

Fig. 2. TEM image of the synthesized CdTe colloidal quantum wells.