PHYSICS AND CHEMISTRY OF SOLID STATE
V. 23, No. 4 (2022) pp. 720-727

Section: Physics

DOI: 10.15330/pcss.23.4.720-727

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ФІЗИКА І ХІМІЯ ТВЕРДОГО ТІЛА
Т. 23, № 4 (2022) С. 720-727

Фізико-математичні науки

PACS: 33.20.Fb; 36.40.Gk; 73.20.Mf; 78.30.-j ISSN 1729-4428

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Tuning the photoluminescence of CdTe quantum dots by controllable coupling to plasmonic Au nanoparticles

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A controllable variation of the photoluminescence (PL) intensity of semiconductor quantum dots (QDs) via their coupling to plasmonic nanoparticles (NPs) is the potential basis for optoelectronic and sensing applications. In this work, the effect of Au NPs on the PL of colloidal CdTe QDs is investigated in solution and solid films. A PL enhancement for the QDs synthesized in water was observed in case of spectral overlap of the plasmon absorption band and QD PL band. In the case of Au NPs synthesized in dimethyl sulfoxyl the trend is to a reduction of the PL intensity. For the reference samples prepared by mixing QDs not with Au NP solutions but with corresponding pure solvent, certain PL enhancement was observed and presumably attributed to a reduction of self-absorption or non-radiative interparticle interaction in less concentrated QD solution. However, the contribution of this dilution-related enhancement is expected to be independent of the spectral properties of NPs and QDs. Therefore, the observed in this work different behavior of QD PL in certain combinations of QDs and NPs is attributed to the interaction between electronic excitation in the QD and plasmon.

Keywords: quantum dot; semiconductor nanocrystal; CdTe; photoluminescence; LSPR; Au nanoparticles.

Received 11 October 2022; Accepted 08 November 2022.

Introduction

Colloidal quantum dots (QDs) are quantum-confined semiconductor nanocrystals that have unique electronic and optical properties, with a range of promising potential applications in novel light-emitters, next-generation solar cells, sensing, and biomedical diagnostics [1–3]. One of their major advantages over bulk semiconductor counterparts and organic semiconductors and chromophores is the tunable narrow-band photoluminescence (PL) and optical absorption spectra which can cover the whole optical range from UV to NIR [4]. A controllable increase or decrease of the QD PL intensity via their coupling to plasmonic nanoparticles (NPs) is the basis for many potential optoelectronic and sensing applications [5,6,15–17,7–14]. The underlying mechanism is an interplay of the absorption and PL enhancement by a strong electric field of the localized surface plasmon resonance (LSPR) of noble metal NPs and PL decrease due to non-radiative charge and energy transfer between the QD and NP. The applications of these effects can lead to improved performance of light emitters [11,15,17,18], enhancement of the QD absorption in solar cells [5], and temperature or pressure sensors [10,19,20].

The PL spectra of QDs can contain not only the narrow and tunable excitonic (interband) PL band, which is valuable for applications but also a broad band related to surface states, which compete with excitonic recombination and are regarded as a drawback for most QD application [21]. The advantage of CdTe QDs is the absence or negligible intensity of this defect-related band, even for QDs obtained at low-temperature and other mild conditions, in particular at room-temperature colloidal synthesis in aqueous media [22–25]. This fact is an additional motivation for the investigation of CdTe QDs for optical and optoelectronic applications. However, the investigations of optical properties of CdTe QDs have been rarely reported compared, for instance, with CdSe or ZnO QDs.

In this work, we studied the effect of colloidal Au NPs on the PL spectra of colloidal QDs synthesized at mild conditions in aqueous solutions, using thioglycolic acid as a stabilizer [22–24]. We observe that the PL intensity can be enhanced or reduced depending on the relative spectral position of the QD PL and LSPR band of Au NPs, as well as on the kind of dispersion medium (solvent) and the presence of additional stabilizers, such as polymers. In particular, Au NPs synthesized in water and dimethyl sulfoxyl (DMSO) were used. The results obtained shed light on the mechanism of excitonic PL coupling with the environment and suggest applications of this coupling in potential functional structures.

I. Experimental

CdTe NCs were synthesized by means of colloidal chemistry, following the protocol described by the part of the authors previously [26]. Briefly, the low-temperature colloidal synthesis has been performed in the reactor of complete mixing in the presence of TGA as a stabilizer. Deionized water was used as the dispersion medium. CdI2 was dissolved in water, and TGA was added under stirring, followed by adjusting the pH to 10 by dropwise addition of NaOH solution. H2Te gas was passed through the solution using argon as a carrier gas. The solution was then refluxed at 100 °C at a different time to control the size of the CdTe QDs. The particle sizes of the CdTe QDs increase with the reflux period. Three different particles sized CdTe QDs (#0) were prepared by refluxing the above solution for 5h (#1), 10h (#2), and 15h (#3), respectively.

Gold nanoparticles were synthesized by citrate reduction of hydrogen tetrachloroaurate (III) trihydrate solution mixed with trisodium citrate dihydrate following the modified Turkevich method [27]. The molar ratio of NaCt to HAuCl4 was the primary factor controlled to achieve the desired particle size. Deionized water or DMSO was used as the dispersion medium [28].

Optical absorption and PL spectra were obtained using StellarNet Silver Nova 25 BWI6 spectrometer and Shimadzu RF-1501 spectrofluorometer, correspondingly.

II. Results and discussion

Figure 1a shows the optical absorption and PL spectra of a series of CdTe QDs used in this work, and the optical absorption spectra of Au NPs are presented in Fig. 1b. Different spectral position of the QD absorption and PL was achieved by varying the synthesis conditions as described in the experimental section. For the Au NPs the variation of the synthesis conditions was also aimed at obtaining a different spectral position of the SPR band, but for the given synthesis route and NP size range, the SPR position does not exhibit a noticeable size dependence.

In Fig. 2 shown are the PL spectra of the aqueous mixtures of CdTe QDs and Au NPs. In particular, spectra are grouped for each of the QD samples combined with three Au NP samples. The ratio of QD to NP solution was 1:5, so the dilution of the QD concentration in the mixtures compared with the initial QDs solution was taken into account by multiplying the intensity of the mixtures by a factor of 5.

It can be seen in Fig. 2 that in most cases the QD PL increases upon adding the Au NP solution. The strongest enhancement takes place for CdTe#2, around one order of magnitude (Fig. 1c). Noticeable enhancement is also observed for the CdTe#0 upon coupling with plasmonic NPs Au1 (Fig.1a). The common property of the latter two CdTe samples is the closer spectral position of their emission spectra to that of the SPR peak of all the Au NPs. Although among these two QD samples the spectral distance to SPR maximum is closer for CdTe#0 than for CdTe#2, the stronger enhancement for all Au NP samples is observed for CdTe#2. This fact may be explained by an interplay of the enhancement of the radiative recombination and non-radiative recombination or transfer of the photoexcited charge carriers in the QDs. In particular, these processes can be strongly affected by the thickness of the CdS shell that forms on the selenide or telluride QDs synthesized using thiols [29,30]. According to the (difference in the) synthesis conditions of the QD samples CdTe#0 and CdTe#2, the latter may contain a thicker CdS shell than the former. This assumption is in agreement with the position of its PL and absorption peaks being shifted towards longer wavelengths compared to the sample CdTe#0. The thickness of the shell is known to have a tremendous effect on the QD PL, its change by even a fraction of a nanometer can change the PL intensity by several times [31,32]. The role of the shell in (increasing) the PL intensity is generally twofold. Firstly, it passivates its traps on the surface of the CdTe core, which can be centers of non-radiative recombination of defect-related PL. Secondly, it creates an energy barrier that reduces the probability of trapping the charge carriers from the QD to the environment (solvent, matrix, etc).

Therefore, in the present case of CdTe QDs interacting with plasmonic Au NPs, the presumably slightly thicker CdS shell in the CdTe#2 QDs may reduce the efficiency of non-radiative processes (in particular charge transfer to Au NPs) compared to CdTe#0 sample, resulting in a stronger PL intensity. The effect induced by the plasmonic NPs on the QD PL is very reproducible and stable in time. In particular, for the sample CdTe#2 the intensity of the PL band and its spectral position is well preserved even after a week of storage of solution at ambient conditions (dotted lines in Fig. 2c).

It has been observed in the literature that the solvent can noticeably influence the PL intensity of colloidal QDs [33–36]. The underlying mechanisms of such influence may include a change in the potential barrier at the outer surface of the QD due to different dielectric properties of the solvent, rearrangement of the intrinsic ligand molecules due to their interaction with solvent molecules, or direct involvement of the solvent molecules into the electronic interaction with the QD. Here we investigated the effect of dimethyl sulfoxyl (DMSO), as well as Au NPs synthesized in this DMSO in analogy to the synthesis of Au NPs in water [28].

For Au-DMSO NPs a different effect on the PL of the

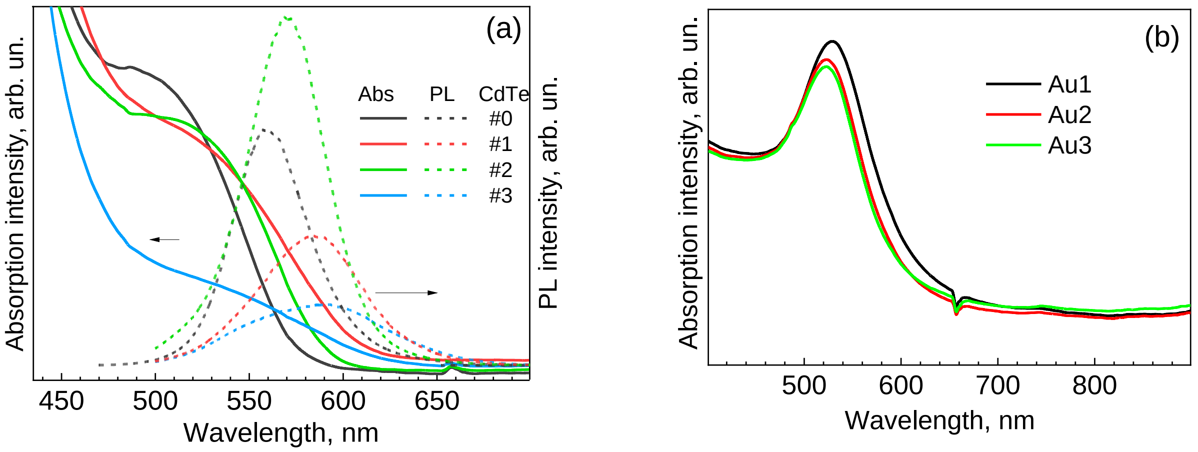

Fig. 1. Optical absorption and PL spectra of a series of CdTe QDs (a) and SPR absorption spectra of the aqueous Au NPs (b) were used in this study.

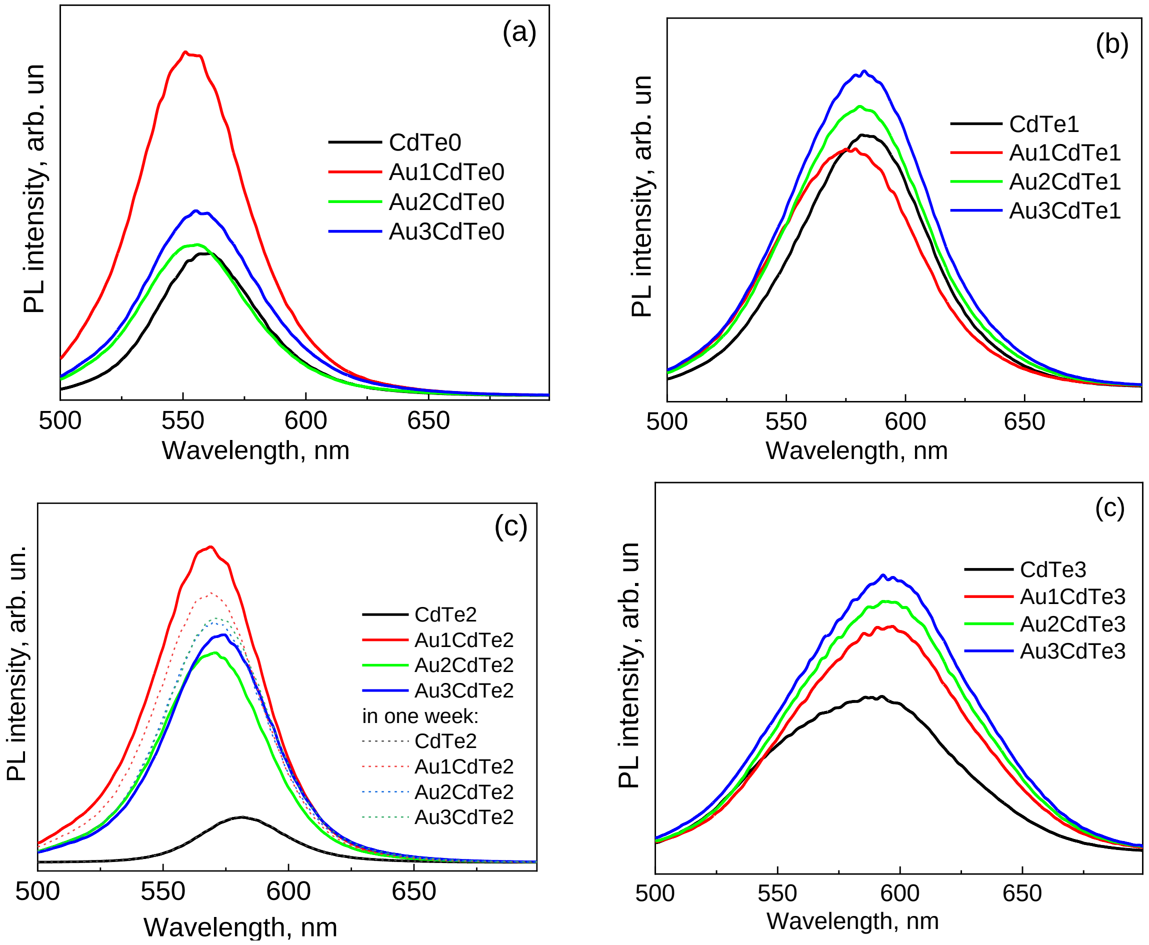

Fig. 2. PL spectra of mixed solutions of CdTe QDs and Au NPs, measured immediately after preparing the mixture. The same solutions measured in one week are shown for CdTe#2 (dotted line).

same series of CdTe QDs was observed (Fig. 3). The PL intensity decreased (roughly by a factor of two) for the samples CdTe#0 and CdTe#1 (Fig. 3 a,c), almost did not change for CdTe#3 (Fig. 3b) and increased significantly for the sample CdTe#3 (Fig. 3d). Comparing the spectra of the QDs mixture with Au NPs in DMSO with the spectra of the mixtures prepared by adding the same amount of pure DMSO we can draw several conclusions. The first is that DMSO causes only weak or no effect on the PL of QD samples CdTe#0, CdTe#1, and CdTe#2 (representative spectra only for CdTe#2 are shown in Fig. 3a), therefore the reduced PL intensity for the corresponding mixtures of these QDs with Au NPs in DMSO can be attributed to the effect of the Au NPs, not DMSO. For the CdTe#3 sample adding DMSO boosts the QD PL by a factor of twelve, while for the corresponding mixture with Au NPs in DMSO the intensity increase is much weaker (Fig. 3b).

The weaker intensity in the mixture with NPs than with pure DMSO is correlates well with the results for other three QD samples and indicates to a partial quenching of the QD PL by Au NPs in all four cases. The only

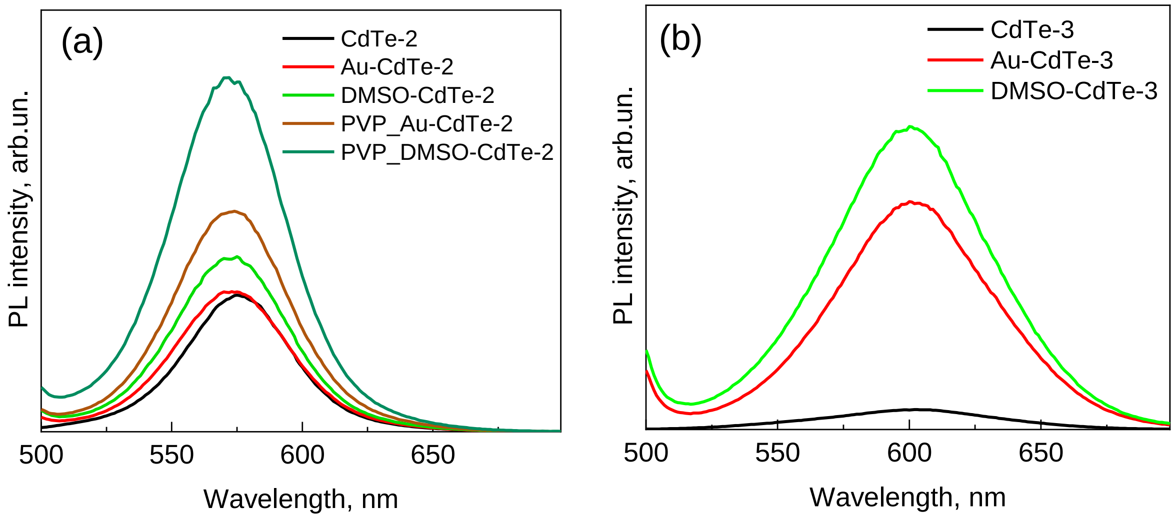

Fig. 3. PL spectra of the series of CdTe QDs with Au NPs synthesized in DMSO. The spectra of the QD solution with the added same amount of pure DMSO are also shown for comparison. For sample CdTe#2 the effect of adding PVP was also studied.

pronounced difference for the sample CdTe#3 is that the PL of this QDs is strongly enhanced by the DMSO itself. Establishing the exact reason or mechanism why this effect is so strong, particularly for this QD sample, would need an additional detailed investigation which is beyond the scope of the present work. Nevertheless, some preliminary assumptions about this mechanism can be made. As DMSO is fully miscible with water, we can expect that DMSO molecules can easily reach the caping (ligand) shell on the QD surface or even the surface of the QDs themselves. The enhancement of the PL intensity can therefore be due to the direct involvement of the DMSO molecules in the (additional) passivation of the QD surface (traps). It has been shown that DMSO used as a solvent allows ZnO QDs with high excitonic PL intensity to be synthesized under mild conditions using no additional stabilizers [37]. It needs to be established yet why this particular QD sample shows a different behavior than the other three samples. Presumably, its ligand shell was more “loose” (as may be assumed from its weakest PL in the series), allowing more efficient access of DMSO molecules to the QD surface.

To further understand the mechanism of the interaction in the semiconductor-metal nanosystems under study, and to explore it in the conditions closer to potential applications, we studied the PL properties of the films prepared from the discussed above mixture of CdTe QDs and Au-DMSO NPs. Below we discuss the results obtained for the QD sample CdTe#2 as a representative case that, an intermediate situation among the results obtained above (Fig. 4) on the series of solution samples. As one of the key factors studied for the film samples is the addition of the polymer, polyvinylpyrrolidone (PVP), we first investigated its possible effect on the QD in the solution.

In Figure 3c the PL spectra of the film samples are shown for CdTe QDs in the following samples: bare QDs, QDs with Au-DMSO NPs, QDs with pure DMSO, as well as the latter two mixtures with adding polymer polyvinylpyrrolidone (PVP). From inspecting the latter

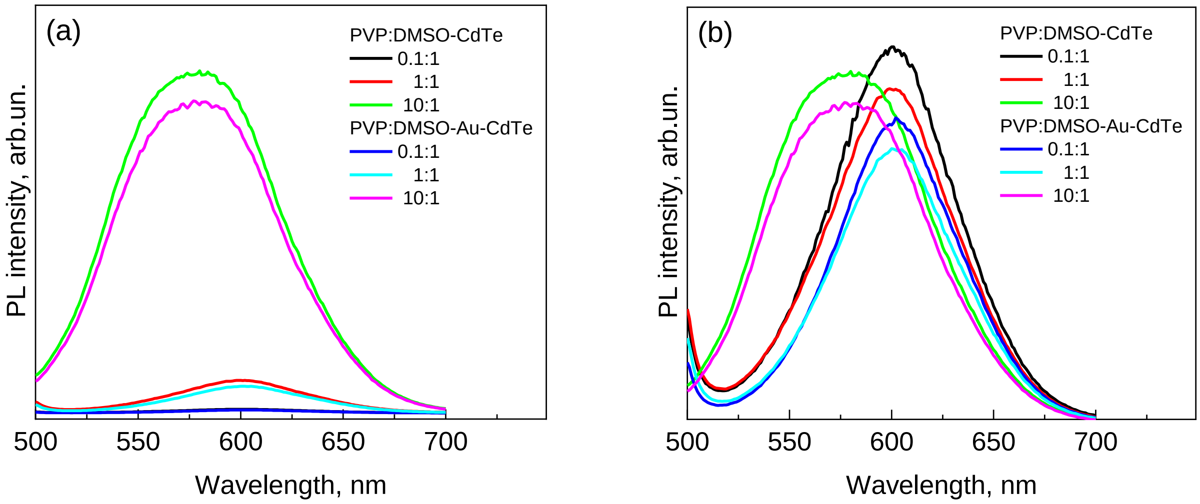

Fig. 4. PL spectra of the CdTe QDs blends with PVP at three polymer/QD ratios (0.1, 1, and 10) and added Au NPs in DMSO or pure DMSO (as a reference sample). The spectra with intensity correction for the different QD concentrations due to the addition of a different amount of polymer are shown in (a). In (b) the spectra shown without intensity correction, in this presentation, the difference in the spectral line shape is more obvious.

figure, we can conclude that the quenching effect of Au NPs is preserved also in presence of PVP – the corresponding QD PL intensity is lower than for the PVP-DMSO-CdTe. The effect of the polymer in both cases is a noticeable increase in the PL intensity. It should be noted that the increase of the QD PL intensity upon adding DMSO can be partially caused by decreasing the concentration of QDs and thus reducing the effects of re-absorption or non-radiative processes between the QDs (such as charge or energy transfer) which may be causing partial PL quenching in the concentrated (as-synthesized) QD solutions [38,39]. We made this assumption based on a similar observation also in the case of diluting the QD solution with water, although the magnitude of the effect was much smaller than for DMSO, thus this is not a purely mechanical effect but there is a specific to each solvent chemical effect on the PL intensity. Moreover, the effect of dilution on the QD PL is expected not to be dependent on the spectral properties of the QD PL and Au NP plasmon, therefore, the different behavior of QD PL in combinations of different QDs and NPs can be indeed attributed to the interaction between the plasmon and electronic excitation in the QD, as discussed above.

As to the choice of the polymer to use in this study, PVP was taken due to its good solubility in both water and DMSO and broad area of applications, in particular use in pharmaceutical and biomedical fields [40], as well as in the synthesis of composites with nanoparticles [41] for various optoelectronic applications [3]. Polymers can affect the properties of the embedded semiconductor QDs or metal NPs via chemical or electronic interaction [3,42–44]. In the present case of PVP blends with CdTe QDs and Au NPs, we also observed a strong effect of the polymer on the QD PL properties. When the ratio of polymer-to-QDs increases significantly, e.g. 10:1, not only QD PL continue to grow proportionally (Fig. 4a), but a drastic change in the QD PL lineshape takes place (Fig. 4b).

The increase of the QD PL intensity upon adding a polymer is a well-documented effect that is explained by the passivation of the surface traps of the QD [3,32,45]. The strong spectral changes of the PL at the high polymer concentration may indicate the involvement of the polymer in the emission process, as was observed for other QD/polymer composites [3,42–44], or some induced changes in the electronic structure of the QDs themselves. This effect needs a separate detailed study.

In the solid films prepared from the studied above sets of samples, all the trends and observations were qualitatively preserved. In particular, the PL intensity is stronger when pure DMSO is added than in the case of added Au NPs in DMSO, both with and without polymer matrix (Fig. 5).

For the CdTe#3 QDs, for which DMSO caused a dramatic boost of PL intensity in the solution (Fig. 3d), this is also the case in the solid polymer film, while the blend with Au-DMSO NPs is less luminescent than the reference sample with only DMSO added. The effect of PL increase caused by DMSO being preserved in the solution may indicate that a part of the DMSO molecules remains adsorbed on the QD surface in the films. This may be the case even without a strong chemical bonding because the temperature used to dry the films, ~50 °C, efficiently evaporate water from the film, while may not be as efficient in completely desorbing DMSO molecules. Because of the high boiling temperature, 189°C, DMSO evaporates very slowly at normal pressure. This property is advantageous for performing reactions at heating but requires a long time or temperature for film preparation.

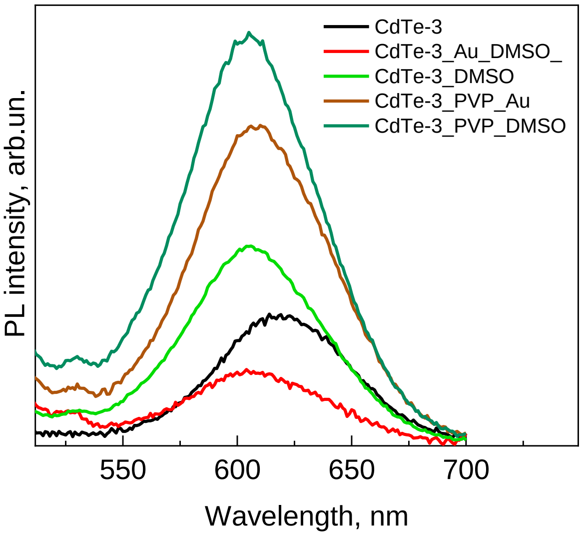

Fig. 5. PL spectra of the solid (dried) film samples prepared from the corresponding solutions (blends) analyzed in previous figures.

Conclusions

We have investigated the effect of the plasmonic Au nanoparticles (NPs) on the photoluminescence (PL) of colloidal CdTe quantum dots (QDs) in solution and solid films. An enhancement of the QD PL can be observed in the case of spectral overlap of the localized surface plasmon resonance (LSPR) absorption band of the Au NPs and PL band of the QDs. An additional condition of the enhancement is concluded to be a sulphide shell of sufficient thickness, which can form an energy barrier that reduces non-radiative charge capture from the QD to the environment. This sulphide shell is known from previous works to form on the QD surface as a result of using thiol molecules as a stabilizer. Qualitatively similar trends in the PL intensity were observed for Au NPs synthesized in water and in dimethyl sulfoxyl (DMSO). Interestingly, the effect of PL change registered in the solutions is preserved upon drying it into a film. By comparing the results on reference samples prepared by mixing the same ratio of QDs not with Au NP solution but with pure solvent, we also established noticeable PL enhancement. In the case of Au NPs synthesized in dimethyl sulfoxyl (DMSO), the general trend is not enhancement but a reduction of the PL intensity. The increase of PL intensity due to a decrease of QD concentration in the solution may be explained by reduced re-absorption or non-radiative interparticle interaction in a less concentrated QD solution. However, the contribution of this dilution-related enhancement is expected to be independent of the spectral properties of PL and the plasmon band. Therefore, the different behavior of QD PL in certain combinations of QDs and NPs can be attributed to an interaction between the plasmon and electronic excitation in the QD.

Acknowledgement

The work was supported by the NAS of Ukraine priority research programme 59/КТ-20-05 "Development and research of hybrid semiconductor nanostructures with metasurfaces for optoelectronic applications" of project "New plasmonic and photonic phenomena and technologies for nano- and optoelectronic devices" (0122U000583).

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Варіювання фотолюмінесценції квантових точок CdTe шляхом контрольованої взаємодії з плазмонними наночастинками Au

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Контрольована зміна інтенсивності фотолюмінесценції (ФЛ) напівпровідникових квантових точок (КТ) зумовлена їх взаємодією з плазмонними наночастинками (НЧ) є основою для потенційних оптоелектронних і сенсорних застосувань. У даній роботі досліджено вплив НЧ Au на ФЛ колоїдних КТ CdTe в розчинах і плівках. Підсилення ФЛ КТ, синтезованих у воді, спостерігається у випадку спектрального перекриття смуги поглинання плазмону та смуги ФЛ КТ. У випадку НЧ Au синтезованих у диметилсульфоксилі, спостерігається тенденція до зниження інтенсивності ФЛ. Для еталонних зразків, отриманих шляхом змішування КТ не з розчинами НЧ Au, а з відповідним чистим розчинником, спостерігалося певне посилення ФЛ, яке, імовірно, пояснюється зменшенням самопоглинання або невипромінювальної взаємодії між КТ в менш концентрованому їх розчині. Однак внесок концентраційного вкладу в підсилення не повинен залежати від спектральних властивостей НЧ і КТ. Таким чином, спостережувана в цій роботі різна поведінка ФЛ КТ у певних комбінаціях КТ і НЧ пояснюється взаємодією між електронним збудженням у КТ і плазмоном.

Ключові слова: квантова точка; напівпровідниковий нанокристал; CdTe; фотолюмінесценція; LSPR; наночастинки Au.