

## OPTICAL PROPERTIES OF CdTe NANOPARTICLES IN WATER

A. I. KASHUBA <sup>1\*</sup>, R. Y. PETRUS <sup>2</sup>, O. S. KUSHNIR <sup>3</sup>, H. A. ILCHUK <sup>1</sup>, I. V. SEMKIV <sup>1</sup>  
AND N. Y. KASHUBA <sup>1</sup>

<sup>1</sup> Department of General Physics, Lviv Polytechnic National University, 12 Bandera Street, 79013 Lviv, Ukraine

<sup>2</sup> Carl von Ossietzky University of Oldenburg, Institute of Physics, Carl-von-Ossietzky-Straße 11, 26129 Oldenburg, Germany

<sup>3</sup> Department of Optoelectronics and Information Technologies, Ivan Franko National University of Lviv, 107 Tarnavsky Street, 79017 Lviv, Ukraine

\* Corresponding author: \*andrii.i.kashuba@lpnu.ua

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**Abstract.** We report on the optical properties of CdTe nanoparticles (NPs) located in water and the same NPs in water placed onto a fused quartz substrate, i.e., the structure (CdTe NPs in water) / quartz. The optical parameters are calculated for different concentrations of the CdTe NPs in water (in the interval 0–100% with the step 10%). The optical transmission and reflection, the refractive index, and the extinction coefficient are found using a transfer-matrix method. All the optical properties are analyzed in the spectral range 500–1500 nm. The optical dielectric functions  $\epsilon_1$  and  $\epsilon_2$  are calculated from the spectral dependences of the refractive index and the extinction coefficient. The high-frequency dielectric constant, the ratio of the carrier density to the effective carrier mass, and the relaxation time are obtained from the dispersion of the optical dielectric function. These parameters are analyzed depending on the CdTe NP concentration in water.

**Keywords:** CdTe nanoparticles, relaxation time, refractive index, optical transmission, optical reflection, optical dielectric function

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### 1. Introduction

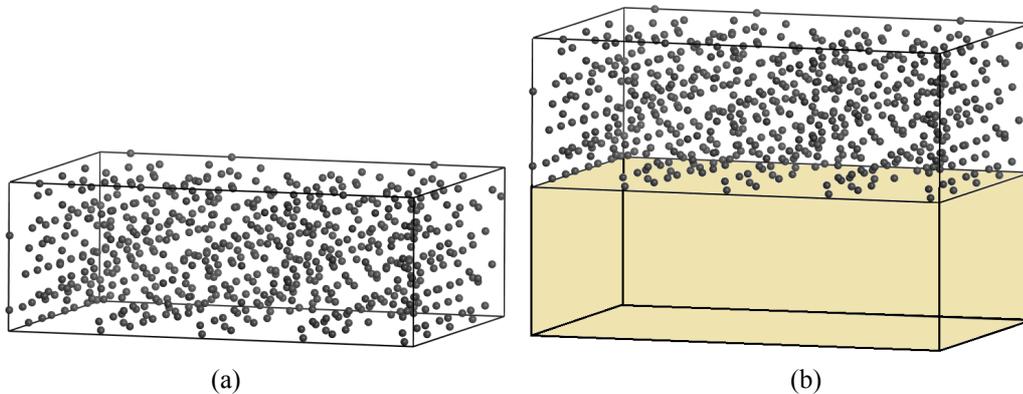
Nanocrystalline semiconducting materials of the II–VI group manifest unique electronic and optical properties due to quantum-confinement effects. The optical characteristics of these materials can be tuned by altering the size of constituting nanoparticles (NPs). Cadmium telluride (CdTe) NPs represent a key II–VI semiconducting material famous for its narrow bulk bandgap and relatively large exciton Bohr radius ( $\sim 7.3$  nm). These NPs hold significant potential for many applications in novel light emitters, next-generation solar cells, sensing technologies, and biomedical diagnostics [1–5].

One of the problems with the synthesis of colloidal CdTe NPs is a restriction imposed by their compatibility with water and/or air [6]. This type of compatibility is especially important in the field of nano-biotechnology because water is a natural solvent. The authors [1, 5–9] have studied experimentally the optical properties of CdTe NPs in water. As reported in Ref. [6], the transfer of the NPs (in particular, the CdTe NPs) from organic to aqueous solutions is a time-consuming procedure with a number of drawbacks such as a limited stability, a low emission quantum yield and a large increase in the actual NP size. Computer modelling of the optical processes occurring in the CdTe NPs would allow for predicting their expected properties. This can also help to optimize the experimental researches and enable forecasting the properties of the CdTe NPs in water. A well known

transfer-matrix method [10] can be used for the corresponding theoretical calculations implemented with Oppromus software [11].

To the best of our knowledge, there are no reports in the literature concerned with the optical dielectric function and the refractive indices of the CdTe NPs maintained in water and a more complex though more practical structure, in which the CdTe NPs in water are placed onto a fused-quartz substrate (the notation (CdTe NPs in water) / quartz is used hereafter). However, the optical properties of those structures can prove to be of primary importance due to their potential practical applications in biotechnology (see also Refs. [1, 5]).

In this work, we focus on the room-temperature spectral behaviour of the optical properties of the CdTe NPs located in water (see Fig. 1a) and of the structure (CdTe NPs in water) / quartz (see Fig. 1b). Basing on the calculated optical functions, we have determined the concentration dependences for the high-frequency dielectric constant, the ratio of the carrier density to the effective mass of carriers and the relaxation time. In particular, we have found the optimal concentration of the CdTe NPs and water, which manifests the shortest relaxation time.



**Fig. 1.** Schematic 3D view of the CdTe NPs in water [panel (a)] and the structure (CdTe NPs in water) / quartz [panel (b)]. Gray balls correspond to CdTe NPs, and a cream layer in panel (b) to quartz substrate. Light propagates top-down through the sample.

## 2. Methods of calculations

For simplicity, we analyze the optical properties of an aqueous colloidal layer of CdTe NPs with the thickness of  $1\ \mu\text{m}$  and a plane-parallel shape (see Fig. 1). We consider a normal light incidence on this layer. The dependences of the transmission ( $T$ ) and reflection ( $R$ ) coefficients of this structure on the wavelength  $\lambda$  of light are calculated using a known transfer-matrix method [10].

The effective refractive indices of colloidal solutions of the CdTe NPs are calculated using a Bruggeman mean-field approach [12, 13]. The latter assumes that different components of the colloidal solution (the NPs and the solvent in our case) are mixed uniformly to form a homogeneous mixture. The size of the NPs remains much smaller than the wavelength  $\lambda$  (usually up to  $0.1\lambda$ ), which allows for neglecting any scattering effects.

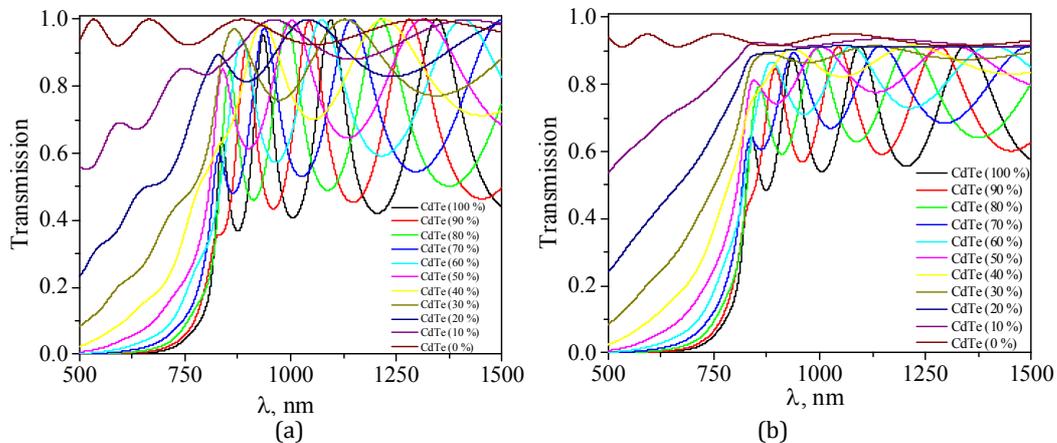
The calculations are based on the assumption that the shape of the CdTe NPs does not differ significantly from a spherical one. The particles are isolated, do not stick together, and they form no aggregates. The complex dielectric functions for CdTe and  $\text{H}_2\text{O}$  required for the calculations have been taken from Refs. [14–16].

Below, we will study the spectral behaviour of the optical functions depending on the content of CdTe NPs in H<sub>2</sub>O as a solvent. An important separate case is also studied when the NPs are placed onto quartz substrates. We note that the above spectral characteristics depend first of all on the CdTe NP concentration  $C_{\text{CdTe}}$  (with  $C_{\text{CdTe}} + C_{\text{H}_2\text{O}} = 100\%$ ), while the dependence on the size of the NPs can be ignored in a reasonable approximation. In our case, the particle size is equal to  $30 \pm 10$  nm.

We will also analyze the limit  $C_{\text{CdTe}} = 100\%$  since the appropriate results for a monolayer with the thickness of  $1 \mu\text{m}$  should coincide with the experimental data known for the bulk CdTe crystals. Note that the case  $C_{\text{CdTe}} = 100\%$  implies that the light propagates through the centres of all the CdTe spheres, which are tightly aligned in the vertical direction if Fig. 1. Then, in effect, the light does not enter the water in the structure (CdTe in water). The above check-up allows us to evaluate the accuracy and reliability of our calculations since no independent information on the CdTe NPs in water with different content ratios is currently available in the literature.

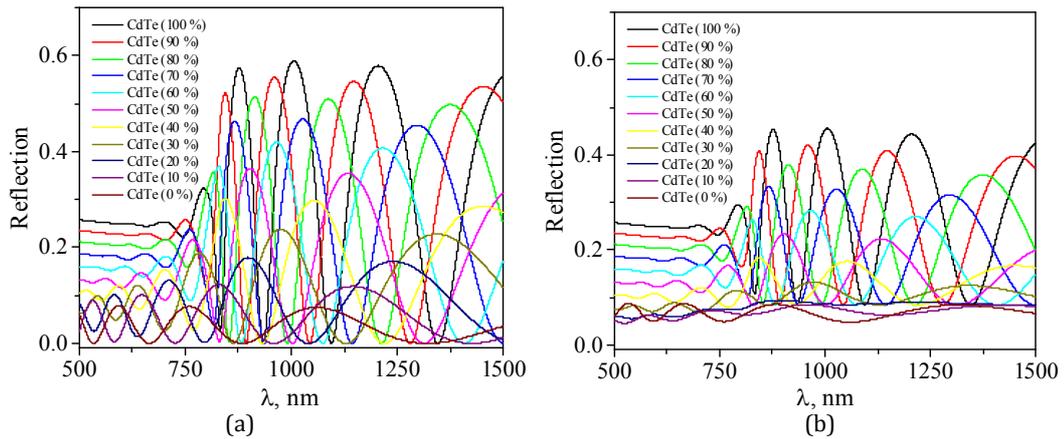
### 3. Results and discussions

Fig. 2 and Fig. 3 show the spectral behaviour of the transmittance ( $T$ ) and reflectance ( $R$ ) for the CdTe NPs contained in water [panels (a)] and for the structure (CdTe NPs in water) / quartz [panels (b)], respectively. We recall again that all our calculations are made at the room temperature. Apart from some general regularities, the spectra of the optical transmittance and reflectance for the CdTe NPs of all concentrations manifest clear signs of optical interference. These phenomena have been observed repeatedly for the similar physical systems (see, e.g., Refs. [17, 18]). This is the interference in the thin film of CdTe NPs in water [17, 18]. It has been analyzed theoretically for both a simplified case of two-beam interference [18] and a general case of multiple light reflections from the thin-film surfaces [19].



**Fig. 2.** Optical transmission spectra calculated for the CdTe NPs in water [panel (a)] and the structure (CdTe NPs in water) / quartz [panel (b)].

The true transmission and reflection coefficients could be obtained from the transmittance and the reflection only after appropriate averaging over the interference oscillation phase [19]. One can see from Fig. 2 and Fig. 3 that the integral values of the optical transmittance and reflectance increase with decreasing CdTe NP concentration in both cases. Also, a sharp increase in those parameters located near 830 nm is obtained for the samples with large enough contents of the CdTe NPs ( $C_{\text{CdTe}} > 30\%$ ). Such behaviour of the optical

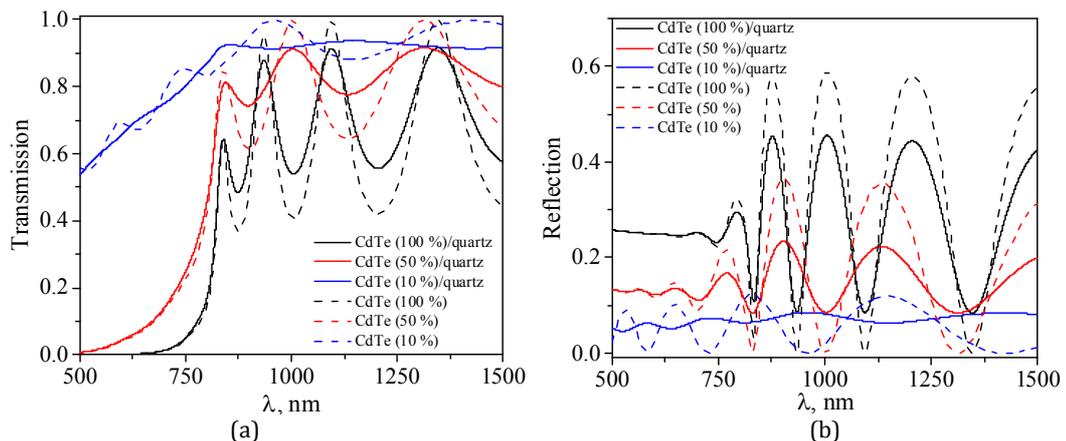


**Fig. 3.** Optical reflection spectra calculated for the CdTe NPs in water [panel (a)] and the structure (CdTe NPs in water) / quartz [panel (b)].

transmittance and reflectance is typical for semiconductor materials and is related to the band-to-band transitions. The energy corresponding to this transition is equal to the bandgap  $E_g$ . We obtain  $E_g = 1.49$  eV for CdTe with the thickness of  $1 \mu\text{m}$ . This value is close to the earlier experimental data ( $E_g = 1.42$  eV) for the thin CdTe films with the thickness of  $1.393 \mu\text{m}$  [17].

For the CdTe NP concentrations higher than 30%, one can see periodic peaks and dips of high enough contrast in the transmission region of CdTe ( $\lambda > 830$  nm). In the spectral region below 830 nm, the transmittance and reflectance are close to zero for any notable CdTe concentrations. This means that the sample absorbs light ( $T + R + A + S = 1$ , where  $A$  denotes the absorption and  $S \rightarrow 0$  – see Section 2).

The optical transmittance and reflectance for the CdTe NP concentrations  $< 30\%$  reveal periodic maxima and minima in the overall spectral range. This can be attributed to the spectral properties of a medium (i.e., water) in which the CdTe NPs are placed (see the data of Fig. 2 and Fig. 3 for the particular case of CdTe concentration 0%).

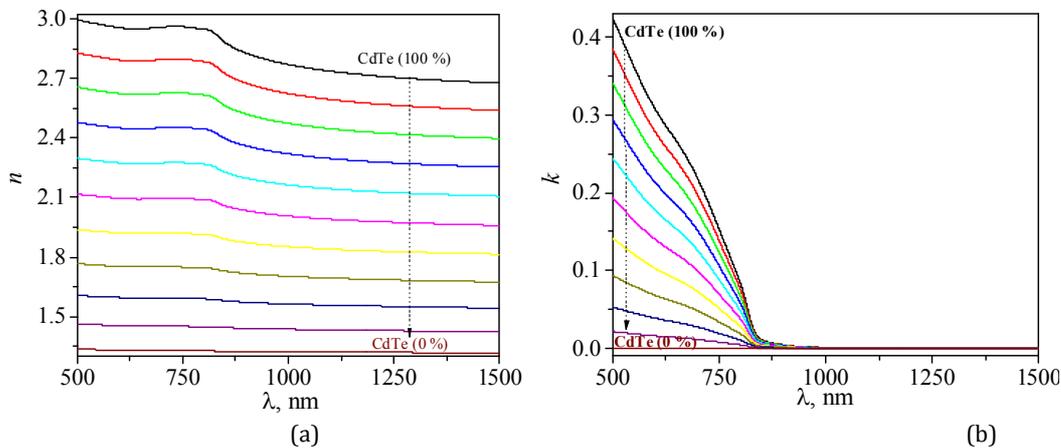


**Fig. 4.** Comparison of the optical transmission [ $T$ , panel (a)] and reflection [ $R$ , panel (b)] for the CdTe NPs in water with the structure (CdTe NPs in water) / quartz.

The transmittance and reflectance for the CdTe NPs located in water and the structure (CdTe NPs in water) / quartz are compared in Fig. 4. One can see a similar spectral behaviour of

the optical functions for the two materials. This indicates that the refractive index does not change when one uses quartz as a substrate for the CdTe NPs in water. In what the absolute values of the optical transmittance and reflectance are concerned, the interference peaks become weaker for the CdTe NP contents above 50%, and they are completely destructed for the zero CdTe concentration. The main reason is the absorption of light by the substrate. Since the spectral behaviour of the transmission and reflection are almost the same for the CdTe NPs in water and the structure (CdTe NPs in water) / quartz and their refractive indices reveal a similar behaviour, further on we present only the data obtained for the case of the CdTe NPs in water.

Fig. 5 displays the spectral behaviours of the refractive index  $n$  and the extinction coefficient  $k$  calculated for the CdTe NPs in water. The refractive index shows a broad peak near 830 nm, which is associated with the band-to-band transition (the energy  $E_g$ ). Also, we obtain a tendency to decrease for the refractive index observed with decreasing content of the CdTe NPs in water. The dispersion of the refractive index is normal ( $dn/d\lambda < 0$ ). Our refractive index values correlate well with the known literature data [20]. At the wavelengths less than 830 nm, the extinction coefficient decreases sharply with increasing wavelength and becomes close to zero above 830 nm.



**Fig. 5.** Spectral dependences of the refractive index [ $n$ , panel (a)] and the extinction coefficient [ $k$ , panel (b)] as calculated for the CdTe NPs in water.

The complex dielectric function ( $\epsilon = \epsilon_1 + \epsilon_2$ ) can be found from the extinction coefficient and the refractive index:

$$\epsilon_1 = n^2 - k^2, \quad (1)$$

$$\epsilon_2 = 2nk. \quad (2)$$

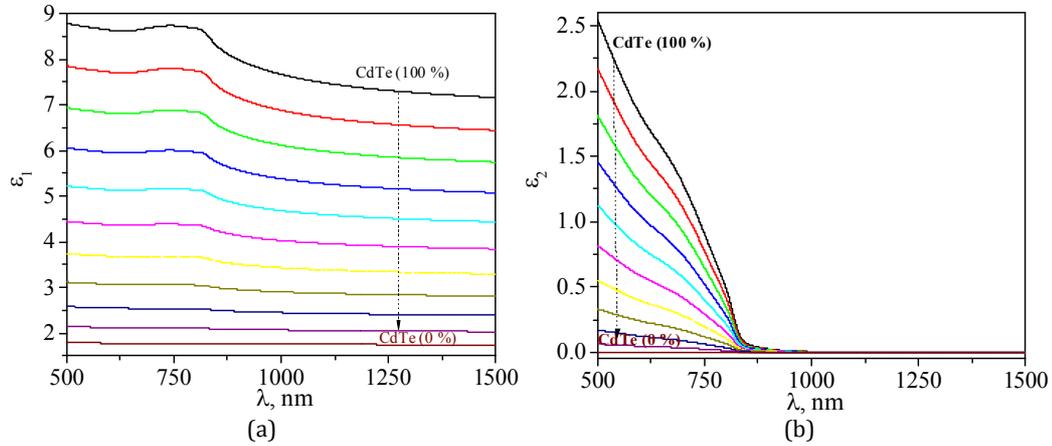
Fig. 6 shows the spectral dependences of the real ( $\epsilon_1$ ) and imaginary ( $\epsilon_2$ ) parts of the dielectric function for the CdTe NPs in water. The behaviours of  $\epsilon_1$  and  $n$  and of  $\epsilon_2$  and  $k$  are similar.

For the  $n$  values much larger than  $k$  (in our case, at  $\lambda > 830$  nm), the  $\epsilon_1$  parameter is approximately equal to  $n^2$ , and the dependence of  $\epsilon_1$  on the wavelength  $\lambda$  can be examined using the relation [21]

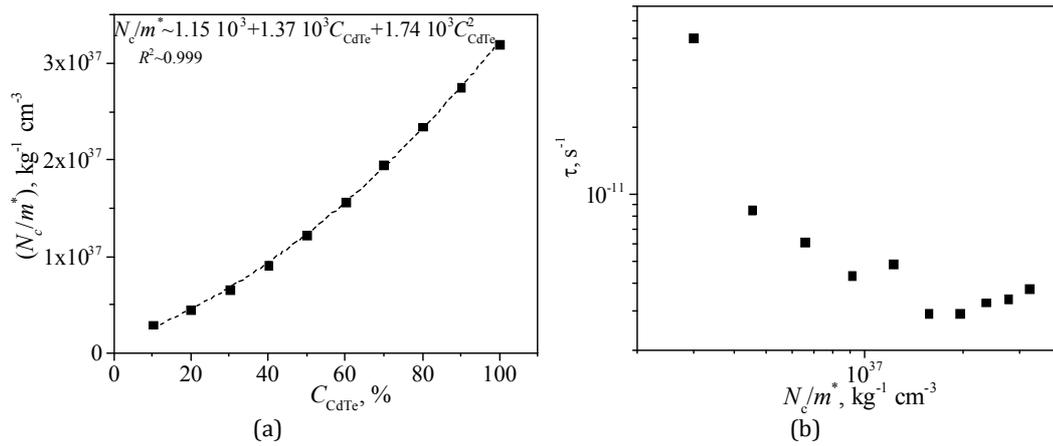
$$\epsilon_1 = n^2 = \epsilon_\infty - \left( \frac{e^2}{\pi \cdot c^2} \right) \left( \frac{N_c}{m^*} \right) \lambda^2, \quad (3)$$

where  $c$  is the speed of light in vacuum,  $m^*$  the effective mass of carriers,  $N_c$  the carrier

density,  $e$  the electronic charge, and  $\varepsilon_\infty$  the high-frequency dielectric constant. In order to obtain  $\varepsilon_\infty$ , we plot  $\varepsilon_1$  as a function of  $\lambda^2$  and extrapolate the linear part of this curve towards the point  $\lambda^2 = 0$ . One can see that the ratio  $(N_c/m^*)$  increases with increasing content of the CdTe NPs in water (see Fig. 7a). The values of the  $\varepsilon_\infty$  and  $(N_c/m^*)$  parameters obtained by us are listed in Table 1. A comparison of our value  $\varepsilon_\infty = 7.67$  for the CdTe NPs (100%) with the literature data  $\varepsilon_\infty \approx 7.15(5)$  [22] known for the bulk CdTe testifies a fair agreement.



**Fig. 6.** Spectral dependences of the real [ $\varepsilon_1$ , panel (a)] and imaginary [ $\varepsilon_2$ , panel (b)] parts of the dielectric function as calculated for the CdTe NPs in water.



**Fig. 7.** Dependence of the ratio  $(N_c/m^*)$  on the concentration of CdTe NPs in water [panel (a)] and dependence of  $\tau$  on  $(N_c/m^*)$  [panel (b)].

The relaxation time  $\tau$  can also be found from Eq. (4). To obtain this parameter, we plot the  $\varepsilon_2$  vs.  $\lambda^3$  dependence and take the value  $(N_c/m^*)$  from the data found above on the basis of Eq. (3):

$$\varepsilon_2 = \left( \frac{e^2}{4\varepsilon_0\pi^3 \cdot c^3} \right) \left( \frac{N_c}{m^*} \right) \left( \frac{1}{\tau} \right) \lambda^3. \quad (4)$$

The relaxation time  $\tau$  for the CdTe NPs can vary depending on different factors, including the particle size, the temperature, and the surrounding medium. Our calculations have been performed for the CdTe NPs with the size  $30 \pm 10$  nm placed in water at the room temperature. The relaxation time can also be influenced by a phonon scattering, electron-phonon interactions and surface effects [2, 3, 5, 23]. The relaxation times typical for the CdTe

NPs can range from picoseconds to nanoseconds, depending on specific conditions. Moreover, the authors of Ref. [5] have obtained a femtosecond dynamics for the CdTe quantum dots placed in water. Finally, we notice that, in the context of photonics and optoelectronics, the relaxation time is critical for understanding the carrier dynamics.

The relaxation times obtained by us are presented in Table 1. When the content of the CdTe NPs in water decreases,  $\tau$  varies from 3.82 to 8.61 ps, except for the concentration 10%. Note that the  $\tau$  value obtained experimentally for the CdTe NPs with the diameters 3.1 and 3.6 nm, which are placed into neutral water, equals 1.6 ps [5]. The latter value agrees well with our calculated data. Moreover, the value  $\tau = 3.82$  ps obtained for the 100% CdTe NPs in water (i.e., for the case of the CdTe layer with the thickness of 1  $\mu\text{m}$ ) is close in the order of magnitude to the value obtained from the first-principles calculations for the bulk CdTe crystals with the cubic structure ( $\tau = 183$  fs [24]). Short picoseconds-like times found for a general case of NP systems have earlier been assigned to Auger recombination [25], hole relaxation [26], or recombination processes occurring due to shallow traps [27]. Taking the information from the literature into account, we assume that these values of the relaxation time can be attributed to an intrinsic mechanism of electron and hole relaxation.

Finally, the dependence of the  $\tau$  parameter on the ratio ( $N_c/m^*$ ) is shown in Fig. 7b. One can see from this dependence that the minimal relaxation time corresponds to the content of the CdTe NPs in water equal to 60%.

**Table 1.** High-frequency dielectric constant  $\varepsilon_\infty$ , ratio ( $N_c/m^*$ ) of the carrier density to the effective carrier mass, and relaxation time  $\tau$  as functions of the content of the CdTe NPs in water.

Concentration of CdTe NPs in water, %	$\varepsilon_\infty$	$N_c/m^*$ , $\text{kg}^{-1} \text{cm}^{-3}$	$\tau$ , ps
100	7.67	$3.2 \times 10^{37}$	3.82
90	6.98	$2.76 \times 10^{37}$	3.43
80	6.20	$2.35 \times 10^{37}$	3.29
70	5.46	$1.96 \times 10^{37}$	2.95
60	4.74	$1.57 \times 10^{37}$	2.93
50	4.08	$1.22 \times 10^{37}$	4.88
40	3.47	$9.13 \times 10^{36}$	4.36
30	2.94	$6.57 \times 10^{36}$	6.13
20	2.47	$4.51 \times 10^{36}$	8.61
10	2.09	$2.98 \times 10^{36}$	50.87

#### 4. Conclusion

We have calculated theoretically the optical properties of the CdTe NPs placed into water and the same material placed onto the quartz substrate, using the transfer-matrix method. In particular, we have obtained the spectral dependences of the transmittance, the reflectance, the refractive index, and the extinction coefficient. It is shown that the integral values of the optical transmittance and reflectance increase with decreasing content of the CdTe NPs in water for both materials under test. For all CdTe NP concentrations, the optical transmittance and reflectance reveal a similar spectral behaviour. A decrease in the CdTe NP concentration leads to decreasing intensity of the interference peaks. It has also been demonstrated that the quartz substrate does not affect the absolute values of the refractive index and the extinction coefficient of the CdTe NPs in water.

The refractive index and the extinction coefficient decrease with decreasing content of the CdTe NPs in water. The optical dielectric function has been calculated with the Kramers–Kronig relations. On this basis, the high-frequency dielectric constant, the ratio of the carrier density to the effective carrier mass, and the relaxation time have been obtained from the spectral dependences of the  $\epsilon_1$  and  $\epsilon_2$  parameters. The numerical values of the optical parameters obtained from our calculations are consistent with the experimental data known from the literature. The picosecond-interval relaxation times have been obtained for the CdTe NPs in water. The smallest value of  $\tau$  corresponds to the content of the CdTe NPs in water equal to 60%. We assume that our results might be important for better understanding of the optical properties of the CdTe NPs in aqueous media and for developing biotechnology and the related research fields.

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**Disclosures.** The authors declare no conflicts of interest.

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**Анотація.** Ми повідомляємо про оптичні властивості наночастинок (НЧ) CdTe у воді та цих же НЧ у воді, поміщених на підкладку з плавленого кварцу, тобто структури (НЧ CdTe у воді) / кварц. Розраховано оптичні параметри для різного вмісту НЧ CdTe у воді в інтервалі 0–100% із кроком 10%. За допомогою методу матриці трансферу знайдено оптичне пропускання та відбивання, показник заломлення та коефіцієнт екстинкції. Усі оптичні властивості проаналізовано в спектральному діапазоні 500–1500 нм. Оптичні діелектричні функції  $\epsilon_1$  і  $\epsilon_2$  обчислено за спектральними залежностями показника заломлення та коефіцієнта екстинкції. На підставі дисперсії оптичних діелектричних функцій одержано високочастотну діелектричну проникність, відношення щільності носіїв до їхньої ефективної маси і час релаксації. Ці параметри проаналізовано залежно від концентрації НЧ CdTe у воді.

**Ключові слова:** наночастинок CdTe, час релаксації, показник заломлення, оптичне пропускання, оптичне відбивання, оптична діелектрична функція