

Fabrication, structural characterization, optical properties, and temperature-dependent fluorescence of CdTe tetrapods

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Abstract. *CdTe tetrapod nanocrystals (NCs) were synthesized using a colloidal hot-injection method, and their structural, vibrational, and optical properties were systematically investigated. X-ray diffraction and transmission electron microscopy analyses confirmed the formation of tetrapod-shaped NCs with a zinc-blende (ZB) crystal structure. Raman scattering measurements revealed clear phonon features associated with longitudinal optical and surface optical modes, indicating the presence of phonon confinement effects in the NCs. Optical absorption and photoluminescence spectra exhibited two distinct features attributed to excitonic transitions in the tetrapod core and arms. Temperature-dependent photoluminescence measurements demonstrated significant variations in emission energy, intensity, and linewidth with increasing temperature, which are mainly governed by band-gap shrinkage and exciton–phonon interactions. The experimental results were analyzed using well-established theoretical models, providing insight into the thermal stability and carrier recombination processes in CdTe tetrapod NCs. These findings contribute to a better understanding of the optical behavior of branched semiconductor nanostructures and their potential applications in optoelectronic devices.*

Keywords: tetrapods; CdTe; optical property; temperature dependence.

Classification numbers: 78.67.Hc; 78.55.-m; 61.05.cp.

1. Introduction

Semiconductor nanocrystals (NCs) have attracted considerable attention in recent decades due to their unique size-dependent optical and electronic properties arising from quantum confinement effects [1, 2]. When the size of a semiconductor crystal approaches the exciton Bohr radius, its electronic energy levels become discretized, leading to tunable optical absorption and emission [3]. As a result, semiconductor NCs exhibit remarkable photophysical properties such

as high photoluminescence efficiency, narrow emission linewidth, and size-controlled emission wavelengths [4]. These distinctive characteristics make them promising materials for a wide range of applications, including light-emitting devices, lasers, biological imaging, photovoltaic cells, and photodetectors [1–4]. In addition, their large surface-to-volume ratio and tunable electronic structure provide further opportunities for tailoring their physical and chemical properties [1,3–5]. Consequently, semiconductor NCs have become an important research topic in nanoscience and nanotechnology.

Among various semiconductor nanomaterials, CdTe NCs have been widely investigated due to their direct band gap, strong absorption coefficient, and efficient photoluminescence in the visible spectral region [6]. These properties make CdTe NCs particularly attractive for applications in optoelectronic devices, light-emitting diodes, solar cells, and fluorescence-based sensing [7]. Depending on the synthesis conditions, CdTe NCs can be prepared in various morphologies such as quantum dots, nanorods, thin films, and tetrapods [8–10]. Each morphology exhibits distinct electronic and optical characteristics because of differences in quantum confinement and structural anisotropy. In particular, tetrapod-shaped NCs, consisting of a central core with four elongated arms, have attracted increasing interest due to their unique branched geometry. This structure provides spatially separated electronic states and anisotropic carrier transport pathways, which may enhance their optical and charge transport properties. Therefore, the fabrication and investigation of CdTe tetrapods are of considerable importance for understanding the relationship between nanostructure geometry and optical behavior.

The temperature dependence of the optical properties of semiconductor NCs is an important issue for both fundamental studies and practical applications [11]. Variations in temperature can significantly influence photoluminescence intensity, emission energy, and spectral linewidth through mechanisms such as band-gap shrinkage, exciton–phonon interactions, and thermally activated nonradiative recombination [11, 12]. Understanding these temperature-dependent processes is essential for evaluating the thermal stability and performance of nanocrystal-based optoelectronic devices. Several studies have investigated the temperature-dependent optical properties of semiconductor NCs, including InP/ZnS, WS₂, and CdSe QDs [13–15]. However, to the best of the authors's knowledge, there have been no detailed studies on the temperature-dependent fluorescence properties of CdTe TPs in the very low temperature range (15–300 K). In this work, CdTe TPs are synthesized and their structural, vibrational, and optical properties are systematically investigated. In particular, the temperature-dependent fluorescence behavior of the CdTe TPs is analyzed to provide further insight into the exciton–phonon interactions and thermal stability of these nanostructures.

2. Experiment

2.1. Materials

Cadmium oxide (CdO), tellurium powder (Te, 99.99%), oleic acid (OA), trioctylphosphine (TOP), 1-octadecene (ODE), trioctylphosphine oxide (TOPO), and other analytical-grade reagents were used as received without further purification. All chemicals were purchased from commercial suppliers and used directly in the synthesis. Organic solvents such as hexane, ethanol, and acetone were used during the purification process. Deionized water was used in all experimental procedures.

2.2. Synthesis of CdTe tetrapods

CdTe TPs were synthesized using a colloidal hot-injection method. In a typical procedure, cadmium oxide (CdO) was dissolved in a mixture of oleic acid (OA) and 1-octadecene (ODE) under continuous stirring and heating until a clear and transparent cadmium oleate solution was obtained. Meanwhile, the tellurium precursor was prepared by dissolving tellurium powder in trioctylphosphine (TOP) and ODE under an inert atmosphere to form a TOP–Te solution. The Te precursor solution was then rapidly injected into the hot cadmium precursor solution at 250 °C, which initiated the nucleation and growth of CdTe NCs. By carefully controlling the reaction temperature, precursor ratio, and ligand concentration, tetrapod-shaped CdTe NCs were obtained. After the growth process, the reaction mixture was cooled to room temperature. The resulting NCs were purified several times by precipitation with ethanol followed by redispersion in hexane to remove unreacted precursors and excess ligands. Finally, the purified CdTe TPs were dispersed in hexane for subsequent structural and optical characterizations.

2.3. Characterization methods

The crystal structure of the synthesized CdTe TPs was characterized by X-ray diffraction (XRD) using a Bruker D8 Advance diffractometer with Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$). The diffraction patterns were recorded over an appropriate 2θ range at room temperature. The vibrational properties of the samples were investigated by Raman scattering (RS) spectroscopy at room temperature using a Horiba LabRAM HR Evolution Raman spectrometer equipped with a 532 nm laser excitation source. The optical absorption spectra were measured using a Shimadzu UV-2600 UV–Vis spectrophotometer in the visible spectral range. The photoluminescence (PL) spectra were recorded using a FLS1000 fluorescence spectrometer (Edinburgh Instruments, UK) with an appropriate excitation wavelength. The temperature-dependent PL measurements were carried out using the same FLS1000 system coupled with a closed-cycle helium cryostat (Janis Research). The measurements were performed in the temperature range from 15 K to 300 K to investigate the influence of temperature on the emission energy, spectral linewidth, and PL intensity of the CdTe TPs.

3. Results and discussion

3.1. Structural and morphology analyses

Figure 1 shows the X-ray diffraction (XRD) pattern of tetrapod-shaped CdTe NCs. Three main diffraction peaks are observed at approximately $2\theta \approx 25^\circ$, 42° , and 50° , which can be indexed to the (111), (220), and (311) crystallographic planes of cubic zinc-blende (ZB) CdTe (JCPDS card No. 15-0770) [6, 8, 16]. Importantly, no additional diffraction peaks corresponding to impurity phases are detected in the XRD pattern, indicating that the obtained CdTe TPs possess high phase purity.

The absence of secondary phases such as CdO or Te confirms that the synthesis method effectively produced single-phase CdTe NCs. The (111) diffraction peak exhibits the highest intensity, indicating that the CdTe TPs possess a preferential orientation along the (111) plane. Such preferential growth is commonly observed in II–VI semiconductor nanostructures and is closely related to the anisotropic growth kinetics during the synthesis process [10, 11]. The diffraction peaks are relatively broad, indicating that the CdTe TPs are nanometer-sized. For ZB structure,

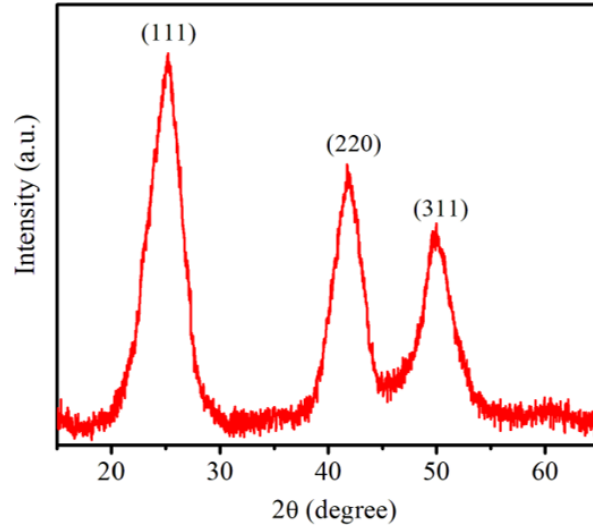


Fig. 1. X-ray diffraction patterns of tetrapod-shaped CdTe NCs.

the relationship between the distance of planes (d_{hkl}) and the lattice constant 'a' of a material is described by the following equation [17]:

$$\frac{1}{d_{hkl}^2} = \frac{h^2 + k^2 + l^2}{a^2}. \quad (1)$$

In the above equation: h , k , and l denote the Miller indices associated with the crystal lattice planes. d_{hkl} represents the interplanar spacing and was calculated using Bragg's equation [17]:

$$n\lambda = 2d_{hkl} \sin \theta, \quad (2)$$

where n is an integer, λ is the X-ray wavelength. Using Eqs. (1) and (2), applied to the diffraction peak (111), the lattice constant a of CdTe is determined to be 6.485 Å.

Figure 2 presents the TEM image of tetrapod-shaped CdTe NCs, together with a schematic model of the tetrapod structure shown in the upper left corner. The TEM image clearly reveals that the synthesized CdTe NCs predominantly exhibit a tetrapod morphology, consisting of a central core from which four elongated arms extend radially. The tetrapod architecture arises from the anisotropic growth of CdTe NCs during the synthesis process. Typically, the central core adopts a zinc-blende structure, while the arms grow preferentially along the $\langle 111 \rangle$ crystallographic directions, leading to

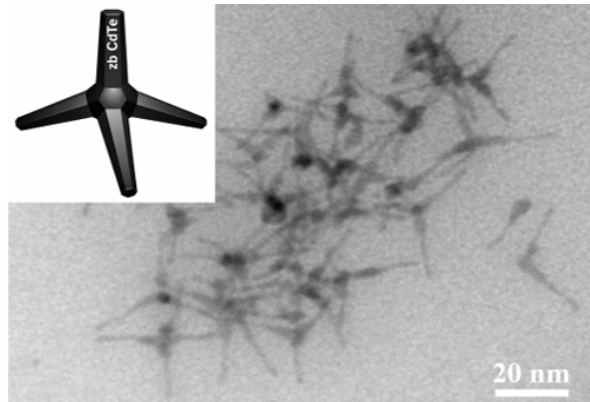


Fig. 2. TEM image of tetrapod-shaped CdTe NCs.

the formation of four branches with approximately tetrahedral symmetry. This growth mechanism is consistent with the structural characteristics observed in many II–VI semiconductor tetrapod nanostructures [18, 19]. From the TEM image, the size of the CdTe TPs can be determined, with a core diameter of approximately 4–5 nm and arms ranging in length from 15–20 nm.

3.2. Vibrational characteristics

Figure 3(a) presents the Raman scattering (RS) spectrum of CdTe TPs measured at room temperature. The multiphonon scattering process, up to the third order, is clearly observed, enabling the investigation of size effects on the vibrational spectra of the NCs. The spectral features of the RS spectrum strongly depend on the size of the NCs. The peak appearing in the low-frequency region is attributed to the surface optical (SO) phonon mode. The first-order longitudinal optical (LO) phonon peak is located at 167 cm^{-1} , which exhibits a blue shift compared with that of bulk CdTe crystals ($159\text{--}163\text{ cm}^{-1}$), accompanied by an asymmetric broadening toward the low-frequency side [20]. The modification of the LO phonon peak characteristics originates from the optical phonon confinement effect in NCs [21]. In addition, the asymmetric broadening on the low-frequency side of the LO peak can also be attributed to the contribution of SO phonons [20, 21]. As the particle size decreases, both the red shift and the asymmetric broadening of the LO phonon peak become more pronounced, indicating the enhancement of the phonon confinement effect in the CdTe TPs.

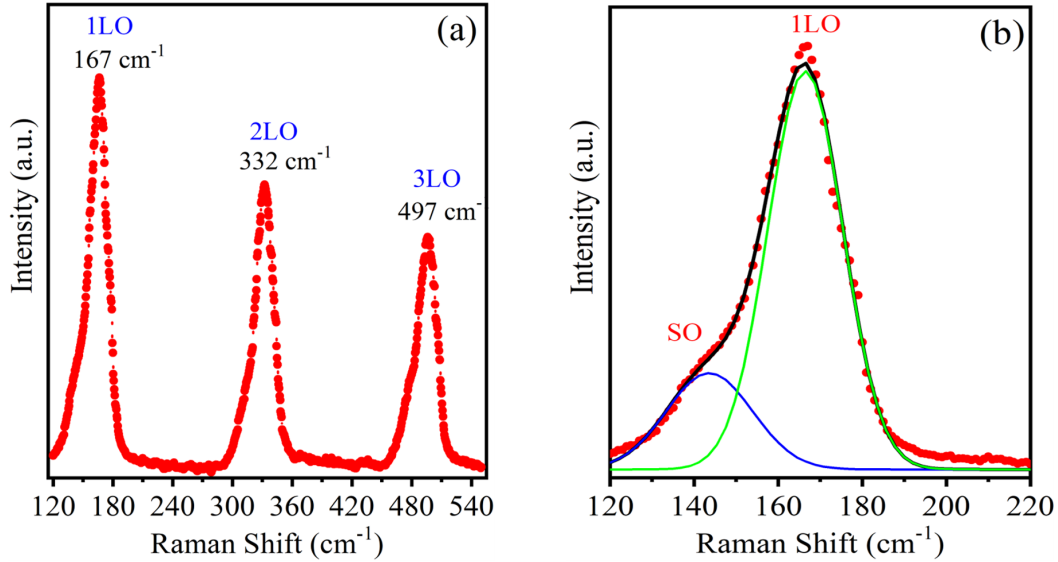


Fig. 3. (Color online) Raman scattering spectra of CdTe TPs.

The RS spectrum was fitted using the phonon confinement (CF) model combined with a Lorentzian function representing the contribution of the surface optical (SO) phonons: $I(\omega) = I_{\text{LO}}(\omega) + I_{\text{SO}}(\omega)$, where $I_{\text{LO}}(\omega)$ is described by the Campbell–Fauchet (C–F) phonon confinement model [22]:

$$I_{\text{SO}}(\omega) = \frac{B\Gamma_{\text{SO}}}{(\omega - \omega_{\text{SO}})^2 + \Gamma_{\text{SO}}^2}, \quad (3)$$

where B is a proportionality constant, while Γ_{SO} and ω_{SO} denote the full width at half maximum (FWHM) and the frequency of the SO phonon, respectively. The fitting result is shown by the solid black curve in Fig. 3(b), while the individual contributions of the LO and SO phonons are represented by the solid blue curves. The obtained fitting parameters are $I_{\text{SO}}/I_{\text{LO}} = 0.32$, $\omega_{\text{SO}} = 144 \text{ cm}^{-1}$, and $\Gamma_{\text{SO}} = 24 \text{ cm}^{-1}$. The obtained fitting parameters indicate a noticeable contribution of the surface optical (SO) phonon to the Raman spectrum. The intensity ratio $I_{\text{SO}}/I_{\text{LO}} = 0.32$ suggests that surface-related vibrational modes play a significant role in the CdTe TPs due to their large surface-to-volume ratio. The extracted SO phonon frequency $\omega_{\text{SO}} = 144 \text{ cm}^{-1}$ is lower than the LO phonon frequency, which is consistent with the typical behavior of surface optical modes in polar semiconductor nanostructures. In addition, the relatively large linewidth $\Gamma_{\text{SO}} = 24 \text{ cm}^{-1}$ reflects the influence of surface disorder and size dispersion in the NCs. These results further confirm that surface effects and phonon confinement significantly affect the vibrational properties of the CdTe TPs.

3.3. Optical properties

The absorption and photoluminescence (PL) spectra of the CdTe TPs measured in solution at room temperature are presented in Fig. 4. The absorption spectrum exhibits two distinct features in the visible region. A pronounced absorption shoulder centered at 586 nm can be observed, which is attributed to the first excitonic transition of the CdTe tetrapod core. This transition originates from the quantum-confined electronic states in the central core region. In addition to the excitonic absorption of the core, another absorption feature appears at longer wavelength ($\sim 665 \text{ nm}$). This feature is associated with the electronic transitions in the elongated arms of the TPs. Due to their larger effective size compared to the core, the branches exhibit a weaker quantum confinement effect, resulting in an absorption contribution shifted towards the red side relative to the core's exciton transition.

The coexistence of these two absorption features indicates that the tetrapod nanostructures consist of two distinct structural domains: a smaller central core and longer nanorod-like arms, each contributing differently to the optical transitions.

The PL spectrum exhibits two emission peaks located at 625 nm and 686 nm, respectively. The emission peak at 625 nm is attributed to the band-edge recombination of excitons confined in the tetrapod core. This emission corresponds well with the first excitonic absorption feature at 586 nm and results in a moderate Stokes shift, which is commonly observed in semiconductor NCs due

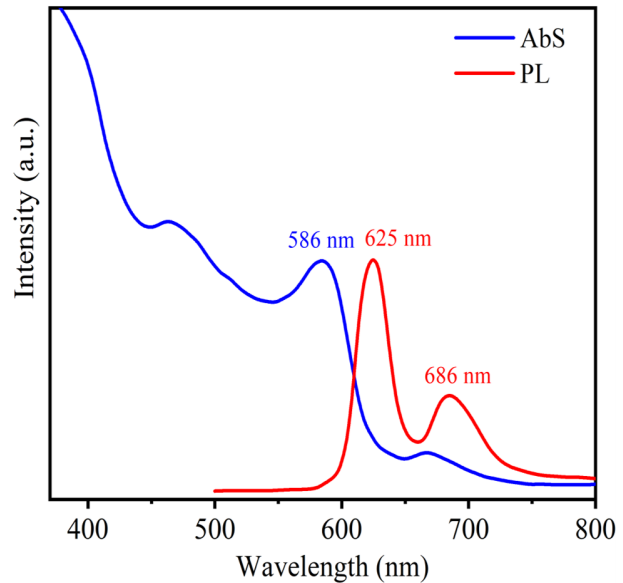


Fig. 4. (Color online) The photoluminescence and absorption spectra of CdTe TPs.

to exciton relaxation and surface effects [11, 13]. The second emission peak centered at 686 nm is assigned to the radiative recombination occurring in the tetrapod arms. Because the arms have a larger effective size and reduced quantum confinement compared with the core, their emission appears at longer wavelengths (lower energies). The presence of this red-shifted PL peak indicates that the arms act as separate recombination sites for charge carriers. Furthermore, the energy difference between the two PL peaks suggests the existence of spatially separated electronic states within the tetrapod structure, where excitons generated in the core may partially migrate toward the arms before recombination.

Previous studies on the optical properties of CdTe or CdSe TPs have reported somewhat different results. A double-peak structure in the optical characteristics of TPs has been observed either in the absorption spectra [19] or in the PL spectra; however, the absorption and PL double peaks have rarely been observed simultaneously. In our experimental results, two distinct peaks are clearly observed in both the absorption and PL spectra of CdTe TPs. Tari *et al.* reported a similar double-peak structure in the PL spectra and attributed it to transitions involving the lowest confined electron and hole states [23]. In their interpretation, the lower-energy emission peak corresponds to the transition between the ground states of the conduction and valence bands ($E_{1e} \rightarrow E_{1h}$), whereas the higher-energy emission peak is associated with the transition between the first excited states in the conduction and valence bands ($E_{2e} \rightarrow E_{2h}$).

3.4. Effect of temperature on the emission properties of CdTe TPs

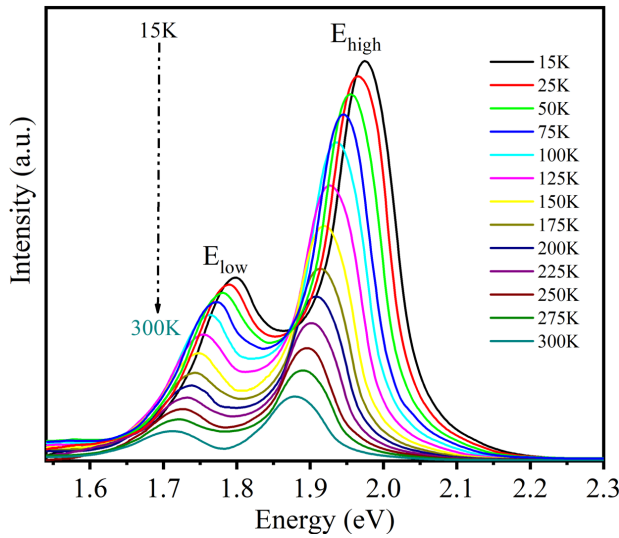


Fig. 5. (Color online) The temperature-dependent PL spectrum of CdTe TPs ranges from 15 to 300 K.

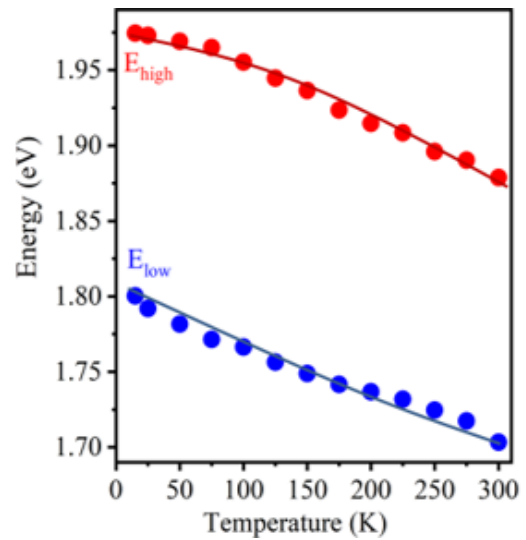


Fig. 6. Temperature dependence of the peak PL energy of the core and arms of a CdTe TP. The solid line is the fit line.

Figure 5 presents the temperature-dependent photoluminescence (PL) spectra of CdTe TPs, measured over the temperature range from 15 to 300 K. Three main effects are observed for both

Table 1. Fitting parameters obtained from the Varshni equation for the experimental data.

CdTe TP	E_{g0} (eV)	α (10^{-4} eV/K)	β (K)
Arms (E_{low})	1.81	3.01	144.3
Core (E_{high})	1.98	3.26	172.7

emission peaks as the temperature increases: (i) a decrease in the PL intensity, (ii) a red shift of the emission peaks toward lower energies, and (iii) a noticeable broadening of the emission linewidth.

Figure 6 presents the temperature dependence of the peak PL energies corresponding to the core (E_{high}) and arms (E_{low}) of CdTe TPs. As the temperature increases from 15–300 K, both emission peaks exhibit a monotonic shift toward lower energies, indicating a clear red shift of the PL emission. For the high-energy emission band, assigned to the tetrapod core, the PL peak energy decreases from approximately 1.97 eV at low temperature to about 1.87 eV at 300 K. Similarly, the low-energy emission band, associated with the tetrapod arms, shifts from around 1.80 eV to 1.70 eV over the same temperature range. The continuous red shift observed for both emission bands is characteristic of the temperature-induced shrinkage of the semiconductor band gap, which is mainly governed by lattice expansion and electron–phonon interactions [11, 12].

Theoretical analyses indicate that these effects can be described by the Varshni relation [11, 12]:

$$E_g(T) = E_{g0} - \alpha \frac{T^2}{T + \beta}, \quad (4)$$

where $E_g(T)$ is the band-gap energy at temperature T (K), E_{g0} is the band-gap energy at 0 K, α is the temperature coefficient, and β is a parameter that is approximately related to the Debye temperature θ_D of the material. The experimental data were fitted using the Varshni equation, and the obtained fitting parameters are summarized in Table 1.

The higher band-gap energy of the core compared with the arms indicates a stronger quantum confinement effect in the smaller central region of the TPs. The larger β value obtained for the core emission may also reflect a higher effective Debye temperature, implying stronger lattice interactions in the core region. The temperature coefficient α obtained for both the core and the arms is consistent with the reported value for CdTe thin film, $\alpha = (2.35 - 4.13)10^{-4}$ eV/K [24]. This agreement indicates that the temperature dependence of the band-gap energy is primarily governed by the intrinsic properties of the crystal lattice in the CdTe TPs. Furthermore, differences in Varshni parameters indicate that the core and branches have different strengths of electron–phonon interactions.

The temperature dependence of the PL intensity (Fig. 7(a)) can be described by the following expression [11, 12]:

$$I(T) = \frac{I_0}{(1 + Ce^{-E_a/k_B T})}, \quad (5)$$

where I_0 is the PL intensity at 0 K, C is a constant related to the radiative lifetime, E_a is the thermal activation energy, and k_B is the Boltzmann constant.

The fitting results yield thermal activation energies of 52 meV for the arms (E_{low}) and 45 meV for the core (E_{high}) of the CdTe TPs as shown in Table 2. The slightly higher E_a value

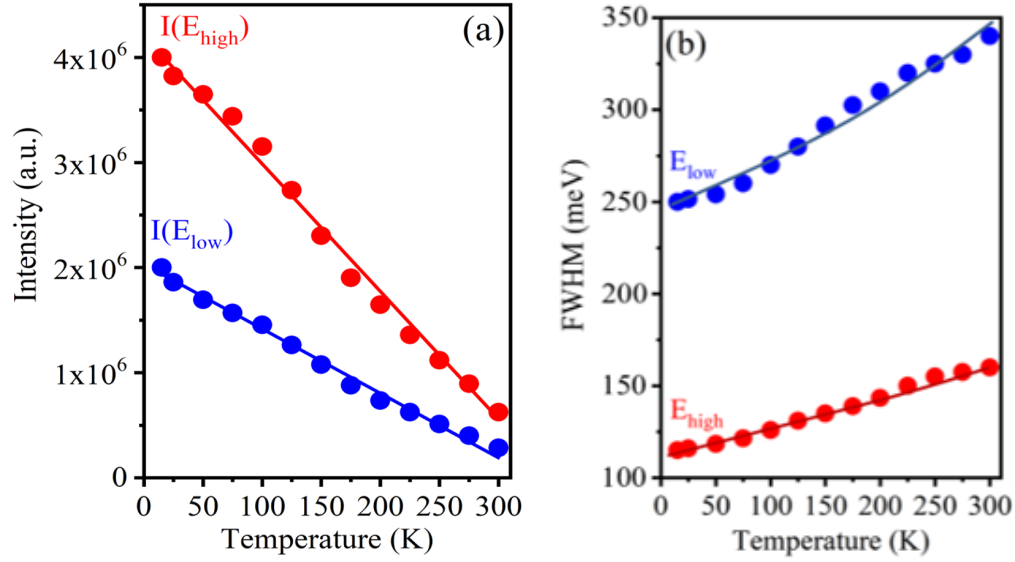


Fig. 7. (a) Temperature dependence of emission intensity of CdTe TPs, (b) Temperature dependence of FWHM of CdTe TPs.

for the arms indicates that carriers in this region require more thermal energy to escape toward nonradiative recombination channels. This suggests that the emission from the arms is thermally more stable compared with that from the core. The difference in activation energies may arise from the distinct confinement environments and carrier localization in the two structural regions of the TPs.

The temperature dependence of the PL full width at half maximum (FWHM) (Fig. 7(b)) is generally determined by exciton–acoustic phonon interactions and exciton–longitudinal optical (LO) phonon interactions, and can be described by the following relation [11, 12]:

$$\Gamma(T) = \Gamma_{inh} + \sigma T + \Gamma_{LO} \left(e^{E_{LO}/k_B T} - 1 \right)^{-1}, \quad (6)$$

where $\Gamma(T)$ is the FWHM at temperature $T(K)$, and Γ_{int} represents the temperature-independent inhomogeneous broadening arising from variations in the size and shape of the NCs. The last two terms correspond to the homogeneous broadening caused by exciton–phonon interactions. Here, σ is the exciton–acoustic phonon coupling coefficient, Γ_{LO} represents the exciton–LO phonon coupling strength, and E_{LO} is the LO phonon energy.

Table 2. Fitting parameters obtained from the Eqs. (5) and (6) for the experimental data.

CdTe TP	E_a (meV)	σ (meV/K)	Γ_{LO} (meV)	E_{LO} (meV)
Arms (E_{low})	52	0.061	67	22
Core (E_{high})	45	0.053	55	20

Figure 7(b) shows the temperature dependence of the PL full width at half maximum (FWHM) for the emissions originating from the arms (E_{low}) and the core (E_{high}) of the CdTe TPs. In both cases, the FWHM increases gradually with increasing temperature, indicating enhanced exciton–phonon interactions at higher temperatures. The experimental data were fitted using Eq. (7), and the obtained parameters are summarized in Table 2. The exciton–acoustic phonon coupling coefficient σ is slightly larger for the arms (0.061 meV/K) than for the core (0.053 meV/K), suggesting stronger exciton–acoustic phonon interactions in the arm region. A similar trend is observed for the exciton–LO phonon coupling strength Γ_{LO} , which is higher for the arms (67 meV) than for the core (55 meV), indicating stronger exciton–LO phonon interactions in the arms. The determined LO phonon energies ($E_{\text{LO}} = 22$ meV for the arms and 20 meV for the core) are close to the reported LO phonon energy of CdTe, confirming the reliability of the fitting model.

4. Conclusion

In this work, CdTe tetrapod NCs were successfully synthesized using a colloidal hot-injection method and their structural, vibrational, and optical properties were systematically studied. XRD results confirmed that the NCs possess a cubic zinc-blende structure with a lattice constant of 6.485 Å, while TEM observations revealed a typical tetrapod morphology with a core diameter of 4–5 nm and arm lengths of 15–20 nm. Raman spectroscopy showed a pronounced LO phonon peak at 167 cm^{-1} together with a surface optical phonon mode at 144 cm^{-1} , indicating strong phonon confinement effects in the NCs. Optical measurements revealed two distinct absorption and PL peaks corresponding to the core and arms, with emission bands located at 625 nm (core) and 686 nm (arms). Temperature-dependent PL measurements from 15 to 300 K demonstrated a continuous red shift of the emission energies, which was well described by the Varshni model with parameters $E_{g0} = 1.98$ eV, $\alpha = 3.26 \times 10^{-4}$ eV/K, $\beta = 172.7$ K for the core and $E_{g0} = 1.81$ eV, $\alpha = 3.01 \times 10^{-4}$ eV/K, $\beta = 144.3$ K for the arms. Thermal activation energies of 45 meV (core) and 52 meV (arms) were obtained from the temperature-dependent PL intensity. Furthermore, analysis of the linewidth broadening yielded exciton–acoustic phonon coupling coefficients of 0.053 meV/K (core) and 0.061 meV/K (arms) and exciton–LO phonon coupling strengths of 55 meV (core) and 67 meV (arms), indicating stronger exciton–phonon interactions in the tetrapod arms. These findings provide valuable insight into the optical behavior and temperature stability of CdTe tetrapod NCs and may be useful for their potential applications in optoelectronic and photonic devices.

Authors contributions

Pham Thi Thuy: Writing – review & editing, Writing – original draft, Methodology, Investigation. Tran Thi Kim Chi: Methodology, Experimentation, Investigation, Data curation.

Conflict of interest

The authors declare that they have no conflicts of interest

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