Supporting Information for:

# Long-lived 1D Excitons in Bright CdTe Quantum Wires

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#### §1. Synthesis and TEM Analysis of CdTe QWs

**§1.A.** Materials. Tellurium granules (Te, -5-+50 mesh, 99.99%, Aldrich), tri-*n*-octylphosphine (TOP, 97%, Strem), and *n*-tetradecylphosphonic acid (TDPA, >99%, PCI Synthesis) were used as received. The TOP contained 0.02 mol% di-*n*-octylphosphinate (DOP), as determined by <sup>31</sup>P NMR following a previously reported procedure.<sup>1</sup> Tri-*n*-octylphosphine oxide (TOPO, 99%, Aldrich) was recrystallized<sup>2-3</sup> before use. DOP was synthesized using a previously reported procedure.<sup>1</sup> Bismuth (Bi) NP stock solutions (0.04 mmol Bi atoms g<sup>-1</sup> solution) <sup>4-5</sup> Cd(di-*n*-octylphosphinate)<sub>2</sub>, tri-*n*-octylphosphine telluride (TOPTe) stock solution (0.025 mmol/g solution), and DOP stock solution (1.01 wt%, 1.45 mol%) were synthesized using previously reported procedures.<sup>6</sup> Toluene (CHROMASOLV for HPLC, Sigma-Aldrich) was distilled under N<sub>2</sub> (g) over Na/benzophenone ketyl, and referred to as dried toluene. Dried toluene (5.30 g) was added in a N<sub>2</sub> (g)-filled glovebox to the as-synthesized CdTe QWs before solidification.

**§1.B.** Synthesis of WZ CdTe QWs. The CdTe QWs (~6.7-nm-diameter, ~1-10- $\mu$ m-length, 98.5% WZ) were synthesized following a previously reported procedure with a slight modification.<sup>6</sup> Briefly, Cd(di-*n*-octylphosphinate)<sub>2</sub> (30.7 mg, 0.0444 mmol), TDPA (2.9 mg, 0.0104 mmol), and purified TOPO (4.0 g, 10.35 mmol) were loaded into a 50-mL Schlenk reaction tube in air. The reaction mixture was degassed under vacuum (0.01–0.1 torr) at ~100 °C and then back-filled with N<sub>2</sub>(g). After repeating this degassing and N<sub>2</sub>(g) back-filling cycle at least four additional times and degassing for additional 1 h, the reaction mixture was inserted into a 300 °C salt bath (NaNO<sub>3</sub>:KNO<sub>3</sub>:Ca(NO<sub>3</sub>)<sub>2</sub>, 21:54:25 mol%) to achieve a clear colorless solution. A 6.3-nm Bi-NP stock solution (20 mg, 0.00080 mmol Bi atoms), TOPTe stock solution (639 mg, 0.0160 mmol TOPTe), and DOP stock solution (60 mg, 0.0015 mmol TOPTe) were combined in a separate vial in a N<sub>2</sub>(g)-filled glove box, which was septum capped. This mixture was brought out of the glove box, loaded into a 3-mL syringe, and then was quickly injected into the Schlenk tube that had been equilibrated in the salt bath for 4 min. The amount of TOPTe (0.0152 mmol) and DOP (0.00212 mmol) injected was calculated by measuring the difference between masses of the syringe before and after the injection. The Cd:Te molar ratio was thus 2.92, and DOP mol% in the reaction mixture was 0.018. The reaction mixture turned dark brown instantly. TOP (182 mg, 0.491 mmol) was injected into the reaction mixture at 3 min of reaction. After 5 min of total reaction the tube was withdrawn from the bath and allowed to cool to room temperature.

**§1.C. TEM analyses.** The purification steps for the CdTe QWs were conducted in the ambient atmosphere. The CdTe-QW-toluene mixture (~1.5 mL) was mixed with methanol (~1.0 mL), followed by centrifugation at 1150 *g* for 5 min and decanting of the supernatant. The QW precipitate was re-dispersed in toluene (~1.0 mL), and was obtained again by adding methanol (ca. 1.0 mL) and by centrifugation and decanting of the supernatant. The precipitate was finally re-dispersed in toluene (~0.5 mL). Carbon-coated copper grids were dipped in the toluene solution, and then immediately taken out to evaporate the solvent. TEM images were collected within 24 hours using a JEOL 2000 FX microscope with an acceleration voltage of 200 kV. High- resolution TEM (HRTEM) was carried out on a JEOL JEM-2100F TEM at 200 kV. The WZ% for ensembles of wires was determined from a previously reported procedure.<sup>6</sup>

Figure S1. TEM analysis and diameter distribution of as-synthesized CdTe QWs



**Figure S1.** Panels **a** and **b**, representative TEM images of 6.7(7) nm as-synthesized CdTe QWs. Panel **c** includes the histogram of CdTe QW diameters measured from TEM images of 42 individual QWs with length  $\ge 1 \mu m$ .

#### §2. Enhancement of CdTe QWs

**Post-synthetic surface modification of CdTe QWs.** The optical properties of CdTe QWs were enhanced with ethanethiol (EtSH, 97%, Aldrich) and trioctylphosphine (TOP, 97%, Aldrich) following a previously published procedure.<sup>7</sup> Briefly, synthesized CdTe QWs, dispersed in dried toluene (~0.265 g), were added to 1.5 mL toluene dropwise via glass pipette to a 1 cm pathlength quartz cuvette in a glovebox. 1.5 mL EtSH was added in air to the cuvette in a fume hood and the sample was submerged in a sand bath to solution level. The sand bath was held at a temperature of 95 - 100 °C for 22 - 24 hours for different samples. After completion, the sample cuvette was centrifuged at a rate of 2300 rpm for 12 minutes and CdTe QWs were visibly collected in a corner of the cuvette. The remaining solution of toluene and EtSH was removed via syringe in a fume hood and 3.75 mL of TOP was added to CdTe QWs remaining in the quartz cuvette. The sample cuvette was then sealed and placed ~50 cm from a 100 W incandescent light bulb for a period of 24 - 100 hours for different QW samples. During this time, CdTe QWs noticeably bundled in solution. All optically enhanced samples remained under incandescence until experiments were performed.



Figure S2. Absorption and PL spectra of enhanced CdTe QWs

**Figure S2**. Panel **a**, UV-visible absorbance (black) and PL (blue) spectra of ensemble CdTe QWs in diluted in TOP,  $\Phi_{PL} = 9.0(3)$ %. Panel **b**, normalized PL spectra of ensemble QWs in TOP with  $\Phi_{PL} < 0.1 - 9.0(3)$ % (black through blue). All PL spectra were collected using an excitation energy of 2.76 eV. The error of the  $\Phi_{PL}$  values of the enhanced CdTe QW samples were obtained from the standard deviation of three consecutive  $\Phi_{PL}$  measurements.

#### **§3. Instrumental Details**

§3.A. Absorption. Ensemble as-synthesized CdTe QWs were prepared for subsequent absorption and PL measurements in a glovebox by dropwise addition of CdTe QW-toluene solution into a quartz cuvette and diluted with TOP. In all cases, samples were prepared with an optical density (O.D) < 0.2 at 2.76 eV ( $\lambda = 450$  nm) without further dilution. Absorption spectra were collected, at room temperature, on a commercial UV-Vis-NIR spectrometer coupled with an integration sphere for collection of forward scattering.

**§3.B.** Photoluminescence. Ensemble steady-state PL spectra and absolute  $\Phi_{PL}$  data were collected using a commercial spectrofluorometer. The light attenuation of internal optics was corrected using PL spectra of NIST certified solid samples (SRM2942, 2943C, 2940B, and 2944B), and absolute  $\Phi_{PL}$  data were obtained using an integration sphere.  $\Phi_{PL}$  values were calculated by collecting PL measurements using a 4-curve method. Sample and blank excitation and emission spectra were recorded over a range of 430-470 nm and 600-800 nm, respectively, with excitation and emission bandpass set at 3 nm. A monochromator (1200g @ 500 nm) was used for emission wavelength selection and a PMT was utilized for detection. A 600-nm longwave-pass filter was placed in front of the monochromator to minimize the detection of scattering from the excitation light when collecting PL spectra. Excitation and emission data were corrected for filters used to obtain the necessary spectral intensities needed for proper linear detection.  $\Phi_{PL}$  values were calculated by taking the ratio of integrated spectral intensity of emitted photons to excitation scatter for each CdTe QW sample. Standard deviations in  $\Phi_{PL}$  values were calculated from three successive  $\Phi_{PL}$  measurements.

**§3.C. PL lifetime measurements.** TRPLDs were recorded using time-correlated single photon counting (TCSPC) with fs pulsed excitation. The pulses were generated by the 5.00 W output of a continuous-wave laser coupled into a wavelength tunable mode-locked Ti:sapphire laser. The output (570 mW, 900 nm, 10 nm FWHM) was then frequency doubled and repetition rates were

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varied using a pulse selector. The doubled output was aligned into the spectrofluorometer sample compartment and emitted light collected using a monochromator. An external focusing lens and neutral density filters were utilized in the excitation beam path to control fluence. In all experiments, a excitation energy of 2.76 eV ( $\lambda = 450$  nm) was used to excite samples. Emitted photons were detected using MCP-PMT cooled by a closed-loop chiller. Typical detection energies and slit width settings were ~1.70 eV and 3-5 nm, respectively. The remaining undoubled light was focused into a silicon photodetector and used as a sync signal for data collection. Photon detection rates were kept below 1% per pulse to avoid signal pile-up effects.





**Figure S3**. The TCSPC instrument response recorded via excitation light scattered off a cuvette filled with TOP using an excitation energy of 2.76 eV (red). The Instrument response has a FWHM ~230 ps for the temporal collection range of 300 ns used when acquiring TRPLDs. The TRPLDs collected at an emission energy of ~1.70 eV for as-synthesized (black) and enhanced,  $\Phi_{PL} = 9.0(3)\%$ , (blue) CdTe QWs in TOP are plotted for comparison.

#### §4. Ensemble PL Lifetime Spectroscopy

**§4.A. Spot-size measurements.** Spot-size measurements were performed using a razor blade mounted on a translation stage at the spatial center of the sample cuvette. The razor blade was translated perpendicularly across the laser beam, and single laser pulse energies were recorded every 0.02mm (Figure S4, black circles). Each profile was then fit to an inverse error function (Figure S4, red curve). Each  $\sigma$  value obtained in the fitting was used to calculate the FWHM of the Gaussian beam using the equation,

$$FWHM = 2\sqrt{2\ln(2)} \sigma \approx 2.355\sigma.$$

The excitation fluence utilized in each experiment was calculated using:

Spot Size Area 
$$(cm^2) = \pi \left(\frac{FWHM(cm)}{2}\right)^2$$

Excitation Fluence 
$$\left(\frac{nJ}{cm^2 \text{ pulse}}\right) = \frac{Pulse \text{ Energy } \left(\frac{pJ}{pulse}\right)}{Spot \text{ Size Area } (cm^2)}$$

## Figure S4. Representative laser spot-size measurement



**Figure S4.** Laser spot-size measurement of the excitation laser used to excite CdTe QWs for PL lifetime studies. For each experiment, single laser pulse energies were recorded as a function of razor blade position (black circles), and the profile was fit to an error function (red curve) in order to estimate the full-width at half-maximum (FWHM) of the excitation spot and the excitation fluence.

**§4.B.** Calculation of the number of charge carriers photogenerated. The number of excitons photogenerated per linear micrometer along each QW per laser pulse,  $N_{\text{exc}}$ , at a given excitation energy, hv in units of Joules, and excitation fluence is calculated using the equation:

$$N_{\rm exc} = \frac{Excitation Fluence}{hv} \chi_{\rm exc}$$

where  $\chi_{exc}$  is the absorption cross section at the excitation energy. The reported absorption crosssection of CdTe QWs vary among the literature. In this report, we estimate values of  $N_{exc}$  using the data reported by Protasenko et al<sup>8</sup>. They reported absorption cross sections at 2.54 and 3.20 eV plotted as a function of  $d^2$ , where d is the diameter of the QWs. We extrapolated each data set to estimate the absorption cross section for QWs with d = 6.7 nm. We obtain absorption cross sections of  $\sim 2 \times 10^{-12}$  cm<sup>2</sup> µm<sup>-1</sup> and  $\sim 4 \times 10^{-12}$  cm<sup>2</sup> µm<sup>-1</sup> for energy of 2.54 and 3.2 eV, respectively. Considering the excitation energy of 2.76 eV used in our studies, we approximate the absorption cross section to be between these two values,  $\sim 3 \times 10^{-12}$  cm<sup>2</sup> µm<sup>-1</sup>. The  $N_{exc}$ values obtained using this absorption cross section are included in **Table S1**.

**§4.C. Multiexponential fitting of TRPLDs.** For each CdTe QW ensemble the TRPLDs are noticeably multiexponential. The role of non-radiative relaxation pathways occurring on differing timescales compete with radiative recombination during intraband<sup>9</sup> and interband<sup>10</sup> relaxation. A useful model for determining the observed relaxation lifetimes in this case is to model the TRPLD data as a sum of exponentials:

$$Counts(t) = A_1 \exp\left(\frac{-t}{\tau_1}\right) + A_2 \exp\left(\frac{-t}{\tau_2}\right) + A_3 \exp\left(\frac{-t}{\tau_3}\right) + \dots, \qquad (1)$$

where  $A_i$  is the amplitude describing the initial contribution of the exponential and  $\tau_i$  is a single lifetime within the sum of exponentials. From the values obtained via multiexponential fitting, two important parameters can be calculated which are presented in this paper. The first parameter is the average lifetime,  $\tau_{avg}$ , which is given by:

$$\tau_{\text{avg}} = \frac{(A_1 \tau_1^2) + (A_2 \tau_2^2) + (A_3 \tau_3^2) + \dots}{(A_1 \tau_1) + (A_2 \tau_2) + (A_3 \tau_3) + \dots}$$
(2)

and Equation 3, the percent contribution, calculated from the ratio of integrated PL intensity contribution of a single lifetime to that of the total integrated intensity of the PL decay,

Contribution (%) = 
$$\frac{A_i \tau_i}{(A_1 \tau_1) + (A_2 \tau_2) + (A_3 \tau_3) + \dots}$$
 100% (3)

These parameters are calculated from the exponential fit parameters detailed below.

Figure S5. TRPLDs of CdTe QWs with varying  $\Phi_{PL}$ 



**Figure S5.** Panel **a**, TRPLDs of as-synthesized,  $\Phi_{PL} < 0.1\%$  and enhanced CdTe QWs with  $\Phi_{PL}$  values ranging from 1.2(1)% to 9.0(3)%. Decays were collected using excitation and emission energies of 2.76 and ~1.70 eV, respectively. The individual TRPLDs of the CdTe QWs included in panel **a** are plotted in **b-g.** Excitation fluence,  $N_{exc}$ , and multiexponential fitting parameters for each measurement are listed in **Table S1**.

| $\Phi_{PL}$ (%) | Fluence<br>(nJ cm <sup>-2</sup><br>pulse <sup>-1</sup> ) | N <sub>exc</sub> *<br>(excitons<br>μm <sup>-1</sup> pulse <sup>-1</sup> ) | A <sub>1</sub> | τ <sub>1</sub> (ns)              | A <sub>2</sub> | τ <sub>2</sub> (ns)             | A <sub>3</sub> | τ <sub>3</sub> (ns)            | A <sub>4</sub> | τ <sub>4</sub> (ns)               | $	au_{avg} (ns)$ |
|-----------------|--|---|----------------|----------------------------------|----------------|---------------------------------|----------------|--------------------------------|----------------|-----------------------------------|------------------|
| As<br>Synth.    | 13.2   | 0.09  | 0.88(6)†       | 0.20(2) <sup>†</sup><br>[27(4)%] | 0.121(8)†      | 1.5(2) <sup>†</sup><br>[28(5)%] | 0.026(1)†      | 11(1) <sup>†</sup><br>[44(5)%] |                |                                   | 5.4(5)†          |
| 1.2(1)          | 1.8  | 0.01  | 0.54(3)        | 0.52(5)<br>[3.1(4)%]             | 0.28(2)        | 3.3(5)<br>[10(2)%]              | 0.127(8)       | 19(2)<br>[26(4)%]              | 0.036(2)       | 155(7)<br><mark>[61(6)%]</mark>   | 100(7)           |
| 1.9(1)          | 14.6   | 0.1   | 0.50(3)        | 0.42(4)<br>[1.7(2)%]             | 0.32(2)        | 2.9(4)<br>[7(1)%]               | 0.14(1)        | 19(2)<br>[21(3)%]              | 0.055(2)       | 157(7)<br><mark>[69(5)%]</mark>   | 113(7)           |
| 4.0(1)          | 1.8  | 0.01  | 0.45(3)        | 0.55(5)<br>[1.0(1)%]             | 0.32(2)        | 3.8(5)<br>[4.7(7)%]             | 0.15(1)        | 26(3)<br>[15(2)%]              | 0.083(4)       | 245(11)<br>[79(7)%]               | 198(15)          |
| 5.9(2)          | 2.4  | 0.02  | 0.46(3)        | 0.63(5)<br>[1.2(1)%]             | 0.27(2)        | 4.4(6)<br>[5.0(8)%]             | 0.15(1)        | 29(3)<br>[18(2)%]              | 0.078(4)       | 230(10)<br>[ <mark>75(7)%]</mark> | 180(13)          |
| 7.2(1)          | 2.9  | 0.02  | 0.44(3)        | 0.54(5)<br>[0.8(1)%]             | 0.30(2)        | 4.2(6)<br>[4.2(7)%]             | 0.16(1)        | 29(3)<br>[15(2)%]              | 0.094(4)       | 254(11)<br><mark>[80(6)%]</mark>  | 207(14)          |
| 9.0(3)          | 2.4  | 0.02  | 0.42(3)        | 0.54(5)<br>[0.8(1)%)             | 0.29(2)        | 3.4(5)<br>[3.3(6)%]             | 0.18(1)        | 25(3)<br>[15(2)%]              | 0.101(5)       | 242(10)<br>[81(7)%]               | 200(14)          |

Table S1. Exponential fitting parameters,  $\tau_{avg}$ , and percent contributions, of CdTe QWs

\* - Since  $N_{\text{exc}}$  is reported in units of excitons  $\mu \text{m}^{-1}$  pulse<sup>-1</sup> and the typical lengths of the CdTe QWs are 1 to 10  $\mu$ m, the number of excitons generated within each QW per laser pulse are 1× to 7×  $N_{\text{exc}}$ .

( )<sup> $\dagger$ </sup> - one standard deviation obtained from the multiexponential fit

() - one standard deviation obtained from data collected from five measurements, see §4.E.

[%] - Percent contribution



Figure S6. TRPLDs of CdTe QWs at varying emission energies

**Figure S6.** Panel **a**, TRPLDs of enhanced CdTe QWs,  $\Phi_{PL} = 7.2(1)\%$ , as a function of emission energies ranging from 1.64 to 1.74 eV. An excitation energy of 2.76 eV was used for all measurements with a fluence 2.9 nJ cm<sup>-2</sup> pulse<sup>-1</sup> yielding  $N_{exc} = 0.02$  excitons  $\mu m^{-1}$  pulse<sup>-1</sup> along the CdTe QWs. The individual TRPLDs included in panel **a** are plotted separately in **b-f**. Multiexponential fit parameters are listed in **Table S2**. Panel **g**, the lifetimes obtained from fitting the TRPLDs were binned into short-, <10 ns (black), intermediate-, 10-35 ns (red), and long-, >35 ns (blue) time constants. The percent contributions of these binned time constants are plotted versus emission energy.

| Emission (eV) | <i>A</i> <sub>1</sub> | τ <sub>1</sub> (ns)   | A <sub>2</sub> | $\tau_2$ (ns)       | <i>A</i> <sub>3</sub> | $	au_3$ (ns)      | $A_4$    | $	au_4$ (ns)        | $	au_{avg}(ns)$ |
|---------------|-----------------------|-----------------------|----------------|---------------------|-----------------------|-------------------|----------|---------------------|-----------------|
| 1.65          | 0.35(2)               | 0.83(7)<br>[0.57(7)%] | 0.28(2)        | 5.4(8)<br>[3.0(5)%] | 0.29(2)               | 32(4)<br>[18(3)%] | 0.144(6) | 274(12)<br>[78(6)%] | 220(15)         |
| 1.67          | 0.38(2)               | 0.52(5)<br>[0.55(7)%] | 0.30(2)        | 3.9(5)<br>[3.3(5)%] | 0.19(1)               | 27(3)<br>[14(2)%] | 0.116(5) | 252(11)<br>[82(7)%] | 210(14)         |
| 1.70          | 0.44(3)               | 0.54(5)<br>[0.8(1)%]  | 0.30(2)        | 4.2(6)<br>[4.2(7)%] | 0.155(9)              | 29(3)<br>[15(2)%] | 0.094(4) | 254(11)<br>[80(6)%] | 208(14)         |
| 1.72          | 0.49(3)               | 0.49(4)<br>[1.0(1)%]  | 0.28(2)        | 4.1(6)<br>[4.6(8)%] | 0.140(8)              | 27(3)<br>[15(2)%] | 0.081(4) | 245(11)<br>[79(7)]  | 199(15)         |
| 1.74          | 0.47(3)               | 0.44(4)<br>[1.0(1)%]  | 0.32(2)        | 3.2(4)<br>[5.1(8)%] | 0.146(9)              | 23(3)<br>[17(3)%] | 0.071(3) | 219(9)<br>[77(6)%]  | 173(12)         |

Table S2. Exponential fitting parameters,  $\tau_{avg}$ , and percent contributions of enhanced CdTe QWs,  $\Phi_{PL} = 7.2(1)\%$ , as a function of emission energy

() - one standard deviation obtained from data collected from five measurements, see §4.E.

[%] - Percent contribution

Figure S7. Average photoluminescence lifetime,  $\tau_{avg}$ , versus emission energy



**Figure S7.** PL lifetime data versus emission energy. The measured average PL lifetimes calculated by Equation 2 are superimposed on the PL spectrum (magenta line) collected for the  $\Phi_{PL} = 7.2(1)\%$  CdTe QW sample. The TRPLDs were collected using an excitation energy of 2.76 eV.

# §4.D. Estimation of long-lifetime component in TRPLDs



# Figure S8. TRPLDs with varying laser repetition rates

**Figure S8.** Panel **a**, TRPLDs of enhanced CdTe QWs were collected using excitation and emission energies of 2.76 and ~1.70 eV, respectively. Laser repetition rates of 20 (black) and 40 kHz (red) were used for the enhanced CdTe QW sample with  $\Phi_{PL} = 9.0(3)$ %, and a repetition rate of 160 kHz (blue) was used for the enhanced CdTe QW sample with  $\Phi_{PL} = 4.0(1)$ %. The individual TRPLDs included in panel **a** are plotted separately in **b-d**.

# §4.E. Estimation of error in multiexponential fitting of TRPLDs



Figure S9. Single and averaged TRPLDs of enhanced CdTe QWs with  $\Phi_{PL} = 7.2(1)\%$ 

**Figure S9.** An individual TRPLD (red circles) and averaged TRPLD (black line) obtained from five individual measurements for an enhanced CdTe QW ensemble with  $\Phi_{PL} = 7.2(1)\%$ . The TRPLDs were recorded using an excitation energy of 2.76 eV with a fluence 2.9 nJ cm<sup>-2</sup> pulse<sup>-1</sup> yielding  $N_{exc} = 0.02$  excitons  $\mu m^{-1}$  pulse<sup>-1</sup> along the CdTe QWs. The emission was collected at an energy of 1.70 eV.

| Scan                 | <b>A</b> <sub>1</sub> | τ <sub>1</sub> (ns)   | A <sub>2</sub>  | $\tau_2$ (ns)       | <b>A</b> <sub>3</sub> | $\tau_3$ (ns)       | <i>A</i> <sub>4</sub> | $	au_4$ (ns)                     | $	au_{avg}(ns)$ |
|----------------------|-----------------------|-----------------------|-----------------|---------------------|-----------------------|---------------------|-----------------------|----------------------------------|-----------------|
| 1                    | 0.508(9)              | 0.72(2)<br>[1.28(6)%] | 0.293(7)        | 4.9(2)<br>[5.0(3)%] | 0.133(4)              | 31(2)<br>[14(1)%]   | 0.090(2)              | 252(8)<br>[79(4)%]               | 205(8)          |
| 2                    | 0.46(1)               | 0.62(2)<br>[0.99(5)%] | 0.307(8)        | 4.2(2)<br>[4.5(3)%] | 0.144(3)              | 29(1)<br>[14.4(7)%] | 0.092(2)              | 252(7)<br><mark>[80(4)%]</mark>  | 206(7)          |
| 3                    | 0.487(8)              | 0.77(2)<br>[1.28(7)%] | 0.257(6)        | 5.6(3)<br>[4.9(4)%] | 0.130(4)              | 35(2)<br>[16(1)%]   | 0.083(3)              | 275(11)<br><mark>[78(5)%]</mark> | 221(12)         |
| 4                    | 0.46(1)               | 0.65(2)<br>[1.08(5)%] | 0.274(8)        | 4.2(2)<br>[4.2(3)%] | 0.145(4)              | 27(1)<br>[14.1(8)%] | 0.090(2)              | 248(6)<br>[81(3)%]               | 204(7)          |
| 5                    | 0.44(1)               | 0.67(3)<br>[1.03(6)%] | 0.291(1)        | 4.1(2)<br>[4.2(2)%] | 0.149(4)              | 28(1)<br>[14.6(8)%] | 0.091(2)              | 252(7)<br>[80(4)%]               | 206(8)          |
| St. Dev.<br>(% Dev.) | 0.03<br>(6.4%)        | 0.06<br>(8.7%)        | 0.019<br>(6.8%) | 0.6<br>(14%)        | 0.008<br>(5.9%)       | 3<br>(11%)          | 0.004<br>(4.5%)       | 11<br>(4.3%)                     | 8<br>(3.8%)     |

Table S3. Error of TRPLD measurements for enhanced CdTe QWs,  $\Phi_{PL} = 7.2(1)\%$ 

() - one standard deviation obtained from the multiexponential fit

[%] - Percent contribution

(% Dev.) - percent deviation calculated from five PL decays

**Reproducibility error analysis.** In order to estimate scan-to-scan errors in the TRPLD measurements for samples with high  $\Phi_{PL}$  values, five consecutive TRPLDs were recorded on the enhanced sample of CdTe QWs with  $\Phi_{PL} = 7.2(1)\%$ . A laser fluence of 2.9 nJ cm<sup>-2</sup> pulse<sup>-1</sup>, corresponding to  $N_{exc} = 0.02$  excitons  $\mu$ m<sup>-1</sup> pulse<sup>-1</sup>, was used. Each of the five TRPLDs was fit to a sum of exponentials. The standard deviation of each of the fitted parameters was calculated using the values obtained from the five different measurements (listed as **St. Dev.** in **Table S3**). This standard deviation was then used to calculate a percent deviation for each parameter (listed as **% Dev.** in **Table S3**.) The percent deviations associated with each  $A_i$  and  $\tau_i$  were then used to estimate the error for the other enhanced CdTe QW samples, with values given in **Tables S1**, **S2**, and **S5**.

# §4.F. Excitation-fluence dependence on TRPLDs



## Figure S10. TRPLD excitation-fluence dependence of as-synthesized CdTe QWs

**Figure S10**. TRPLDs of ensemble as-synthesized CdTe QWs diluted in TOP,  $\Phi_{PL} < 0.1\%$ , under excitation fluences of 2.9 nJ cm<sup>-2</sup> pulse<sup>-1</sup> (black circles) and 382 nJ cm<sup>-2</sup> pulse<sup>-1</sup> (red circles) at an excitation energy of 2.76 eV. The inset is an expansion of the short time region. TRPLDs were fit to a sum of three exponentials which are detailed in **Table S3**. The fit for the 382 nJ cm<sup>-2</sup> pulse<sup>-1</sup> TRPLD is shown (yellow curve).

| Fluence<br>(nJ cm <sup>-2</sup><br>pulse <sup>-1</sup> ) | N <sub>exc</sub> *<br>(excitons<br>µm <sup>−1</sup> pulse <sup>−1</sup> ) | A <sub>1</sub> | τ <sub>1</sub> (ns)               | A <sub>2</sub> | $\tau_2$ (ns)      | A <sub>3</sub> | $\tau_3$ (ns)     | $	au_{avg}(ns)$ |
|--|---|----------------|-----------------------------------|----------------|--------------------|----------------|-------------------|-----------------|
| 2.9  | 0.02  | 0.89(6)        | 0.17(1)<br>[28(3)]                | 0.107(7)       | 1.4(2)<br>[28(5)%] | 0.020(1)       | 12(1)<br>[44(5)%] | 5.8(5)          |
| 7.8  | 0.06  | 0.90(6)        | 0.17(1)<br>[27(3)%]               | 0.108(7)       | 1.6(2)<br>[31(5)%] | 0.020(1)       | 12(1)<br>[42(5)%] | 5.6(5)          |
| 11.7   | 0.08  | 0.87(6)        | 0.17(1)*<br><mark>[26(3)%]</mark> | 0.119(8)       | 1.4(2)<br>[29(5)%] | 0.024(1)       | 11(1)<br>[46(6)%] | 5.5(4)          |
| 13.2   | 0.09  | 0.88(6)        | 0.20(2)<br>[27(4)%]               | 0.121(8)       | 1.5(2)<br>[28(5)%] | 0.026(1)       | 11(1)<br>[44(5)%] | 5.4(4)          |
| 268  | 1.8   | 0.98(6)        | 0.21(2)<br>[40(5)%]               | 0.063(4)†      | 2.5(3)<br>[30(5)%] | 0.011(1)       | 14(1)<br>[29(4)%] | 5.0(5)          |
| 382  | 2.6   | 0.95(6)        | 0.20(2)<br>[39(5)%]               | 0.071(5)†      | 1.9(2)<br>[27(4)%] | 0.014(1)       | 12(1)<br>[34(4)%] | 4.7(4)          |

Table S4. Exponential fitting parameters,  $\tau_{avg}$ , and percent contributions of as-synthesized CdTe QWs with varying excitation fluence at 2.76 eV

\* - Since  $N_{\text{exc}}$  is reported in units of excitons  $\mu \text{m}^{-1}$  pulse<sup>-1</sup> and the typical lengths of the CdTe QWs are 1 to 10  $\mu$ m, the number of excitons generated within each QW per laser pulse are 1× to 7×  $N_{\text{exc}}$ .

() - one standard deviation obtained from the multiexponential fit

[%] - Percent contribution

# Figure S11. TRPLD excitation-fluence dependence of enhanced CdTe QWs, $\Phi_{PL} = 9.0(3)\%$



**Figure S11**. TRPLDs of an enhanced CdTe QW sample with  $\Phi_{PL} = 9.0(3)\%$  excited at 2.76 eV and emission collected at 1.70 eV. The laser fluence was varied from 1.3 (blue circles) to 10.2 nJ cm<sup>-2</sup> pulse<sup>-1</sup> (black line). The inset is an expansion of the short time region. Decays were fit to a sum of four exponentials as plotted for the 1.3 nJ cm<sup>-2</sup> pulse<sup>-1</sup> TRPLD (yellow curve). All fitting parameters are detailed in **Table S5**.

Table S5. Exponential fit parameters,  $\tau_{avg}$ , and percent contributions of CdTe QWs,  $\Phi_{PL} = 9.0(3)\%$ , with varying excitation

| Fluence<br>(nJ cm <sup>-2</sup><br>pulse <sup>-1</sup> ) | N <sub>exc</sub> *<br>(excitons<br>µm <sup>−1</sup> pulse <sup>−1</sup> ) | A <sub>1</sub> | τ <sub>1</sub> (ns)  | A <sub>2</sub> | $\tau_2$ (ns)       | A <sub>3</sub> | $	au_3$ (ns)      | A <sub>4</sub> | τ <sub>4</sub> (ns)              | $	au_{avg}\left(ns ight)$ |
|--|---|----------------|----------------------|----------------|---------------------|----------------|-------------------|----------------|----------------------------------|---------------------------|
| 1.3  | 0.009   | 0.48(3)        | 0.62(5)<br>[0.9(1)]  | 0.27(2)        | 4.7(7)<br>[4.0(7)%] | 0.157(9)       | 29(3)<br>[14(2)%] | 0.097(4)       | 264(11)<br><mark>[81(6)%]</mark> | 217(14)                   |
| 2.4  | 0.02  | 0.42(3)        | 0.54(5)<br>[0.8(1)%] | 0.29(2)        | 3.4(5)<br>[3.3(6)%] | 0.18(1)        | 25(3)<br>[15(2)%] | 0.101(5)       | 241(10)<br>[81(7)%]              | 200(15)                   |
| 10.2   | 0.07  | 0.42(3)        | 0.43(4)<br>[0.6(1)%] | 0.31(3)        | 3.2(4)<br>[3.5(6)%] | 0.16(1)        | 25(3)<br>[14(2)%] | 0.095(4)       | 241(10)<br>[82(6)%]              | 200(13)                   |

fluence excited at 2.76 eV

\* - Since  $N_{\text{exc}}$  is reported in units of excitons  $\mu \text{m}^{-1}$  pulse<sup>-1</sup> and the typical lengths of the CdTe QWs are 1 to 10  $\mu$ m, the number of excitons generated within each QW per laser pulse are 1× to 7×  $N_{\text{exc}}$ .

() - one standard deviation obtained from data collected from five measurements, see §4.E.

[%] - Percent contribution



Figure S12. TRPLD dependence of detected photon counts on excitation fluence

**Figure S12.** The average number of photon counts detected per laser pulse when recording the TRPLDs plotted as a function of excitation fluence. The data for the  $\Phi_{PL} < 0.1\%$  and  $\Phi_{PL} = 9.0(3)\%$  CdTe QW samples are included as filled circles in **a** and **b**, respectively. The red dashed line in each plot represents a linear dependence. The inset in **a** is an expanded view at the lowest fluences. The TRPLDs were recorded with excitation at 2.76 eV and the emission was collected at ~1.70 eV. As indicated in **Table S1**, typical fluences utilized when recording the TRPLDs for determining the PL lifetimes were < 15 nJ cm<sup>-2</sup> pulse<sup>-1</sup>.

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