Electronic Supporting Information

Low threshold lasing from colloidal CdSe/CdSeTe core/alloyed-crown type-II heteronanoplatelets

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Methods:

Synthesis of the four monolayer (4 ML) CdSe NPLs: 4 ML CdSe NPLs were synthesized according to the published procedures.[1] The mixture containing 60 mg of selenium (Se), 60 mL of octadecene (ODE) and 680 mg of cadmium myristate put into 100 mL three-neck flask and was degassed for 20 min at 60°C. Under nitrogen gas, the temperature of the solution was increased to 240°C, and at 205°C 250 mg of cadmium acetate dihydrate was inserted. After reaching 240°C, 2 mL oleic acid (OA) was injected, and the solution was quickly cooled down to room temperature. 4 ML CdSe NPLs were precipitated by adding ethanol and centrifugation. NPLs were further dispersed and kept in hexane.

Synthesis of 4ML CdSe/$CdSe_{1-x}Te_x$ core/crown NPLs: $CdSe_{1-x}Te_x$ crown was grown by slightly modifying the published procedures.[2-4] For CdSe/CdTe core/crown NPLs, x=1, and for CdSe/CdSeTe core/crown NPLs, x=0.2. First, anisotropic growth mixture required for the crown growth (containing 2 mL ODE, 500 mg cadmium acetate dehydrate and 350 µL OA) was synthesized by heating the mixture up to 165°C and repeatedly sonicating until a whitish gel is formed. For CdSeTe crown growth, 30% of the previously synthesized 4ML CdSe NPLs dissolved in hexane, 100 µL of OA, 15 mL of ODE and 600 µL of anisotropic growth mixture was put into a 50 mL three-neck flask. The mixture was kept under vacuum ~30 min at room temperature till the complete removal of hexane. Under nitrogen gas flow, the temperature was set to 240°C and 3 ml 0.03M ODE−TOP−$Te_{1-x}Se_x$ was injected at 15 mL/h. 0.03M ODE−TOP−$Te_{0.2}Se_{0.8}$ was prepared in glovebox by dilution of previously prepared 1M TOP-Te and 1M TOP-Se with ODE. The solution was quickly cooled down to room temperature after completing the injection. CdSe/$CdSe_{1-x}Te_x$ core/crown NPLs were

precipitated by adding ethanol and centrifugation. NPLs were further dispersed and kept in toluene for further usage.

Morphology characterization:

High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images were taken with JEOL JEM-2100F at 200 kV. AFM was performed through an Asylum Research MFP-3D AFM with a silicon cantilever operating in tapping force mode.

Photoluminescence quantum yield (PLQY) measurement:

Absolute PLQY of the NPL solution was determined at the excitation wavelength of 405 nm. The sample was placed in an integrating sphere, and the absorbed and emitted photons were measured via a spectrometer (Ocean Optics USB2000) that couples with an integrating sphere.

Optical characterizations using femtosecond laser:

The pump laser for time-resolved PL (TRPL), transient absorption (TA) and ASE measurements was generated from a 1-kHz regenerative amplifier (Coherent Libra^{TM}). The beam from the regenerative amplifier has a center wavelength at 800 nm, a pulse width of around 100 fs and was seeded by a mode-lock Ti-sapphire oscillator (Coherent Vitesse, 80MHz). 400 nm pump laser was obtained by frequency doubling the 800-nm fundamental regenerative amplifier output using a $BaBO_2$ nonlinear crystal. 550 nm pump laser was obtained by OPA. PL signal was collected by a charge-coupled device array (Princeton Instruments, Pixis^{TM}) coupled to a monochromator (Acton, Spectra Pro^{TM}). The time-resolved PL was resolved by an Optronis Optoscope^{TM} streak camera system. Transient absorption measurements in the time range of fs-ns were performed using a Helios^{TM} setup (Ultrafast Systems LLC). The white light continuum probe beam (in the range from 400 to 800 nm) was generated from a 3-mm sapphire crystal using 800 nm fs pulse from the regenerative amplifier above. The probe beam was collected using a detector for UV-Vis (CMOS sensor). All measurements were performed at room temperature.

Optical characterizations using sub-nanosecond laser:

The pump laser for PL spectra in Figure 5 and 6 was a Nd:YAG laser at 355 nm with a pulse width of 0.5 ns and a repetition rate of 60 Hz. The PL signal was collected with a monochromator (ANDOR Shamrock 303i) and a CCD (ANDOR iDus 401).

The linear absorption spectra were recorded using a Shimadzu UV-1800 UV–VIS–NIR spectrophotometer. Photoluminescence spectra were recorded by using spectro-fluorophotometer at room temperature (Shimadzu RF-5301PC).

Supplementary Figures:

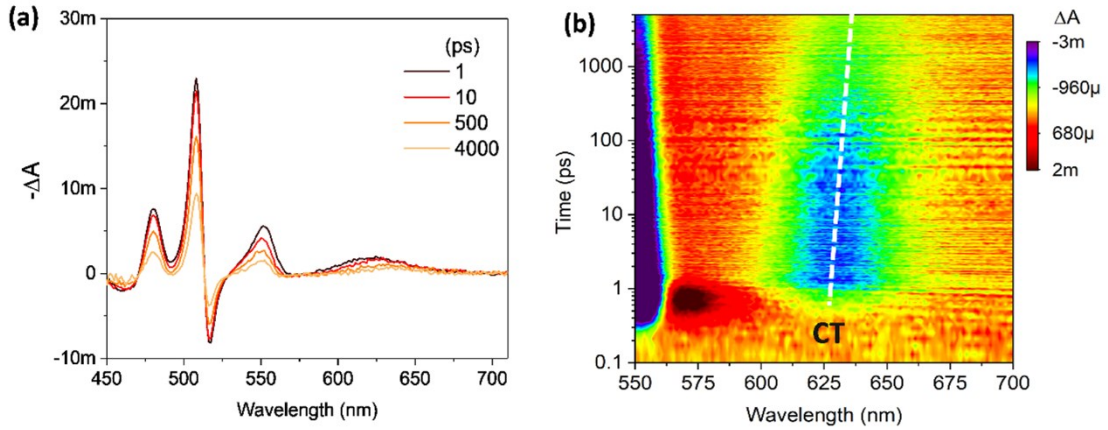

Fig S1 Time evolution of (a) TA spectra and (b) pseudocolor plot of TA spectra of CdSe/CdTe NPLs in solution under 400 nm laser excitation with a pump fluence of 48 μJ cm^{-2}.

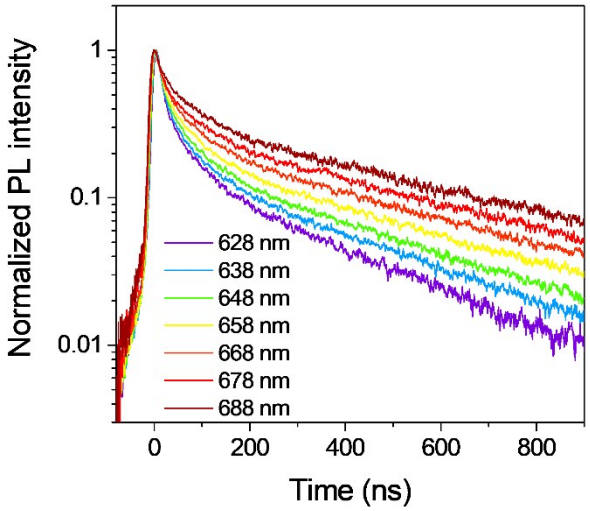

Fig S2 Wavelength-dependent TRPL curves of CdSe/CdTe NPLs under 400 nm excitation.

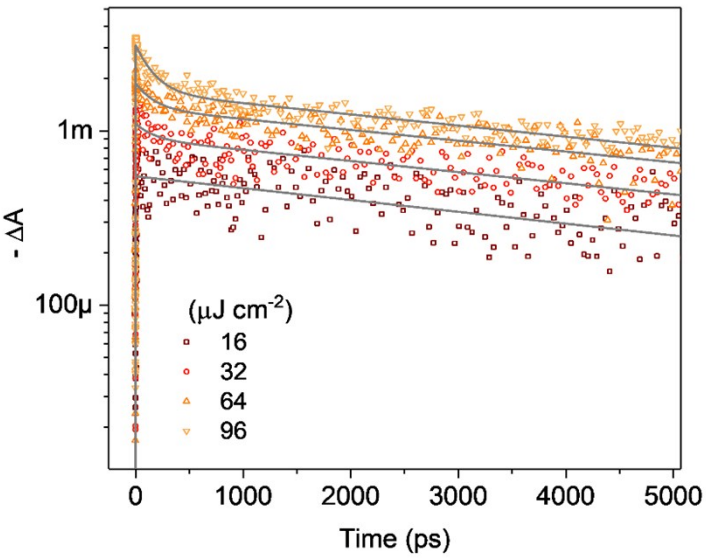

Fig S3 PB decay dynamics probed at CT states of CdSe/CdTe NPLs in solution under 400 nm excitation at different pump fluence levels. The grey lines are numerical fittings using a bi-exponential decay function; the fitted parameters are shown in Table S4.

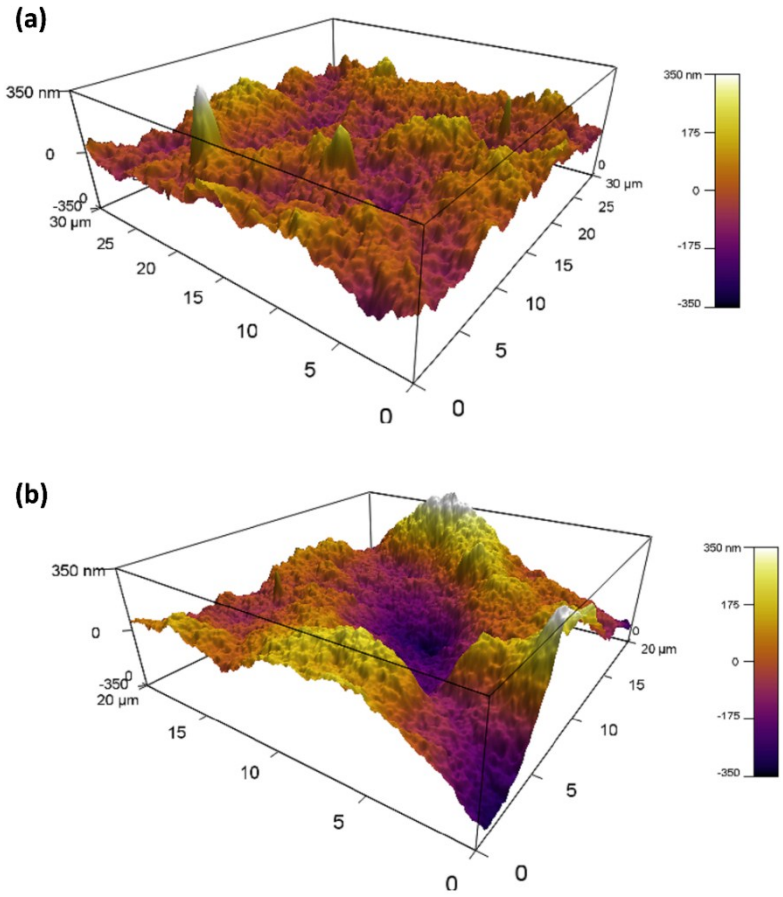

Fig S4 AFM image of drop-casted CdSe/CdSeTe NPLs films on (a) un-scratched and (b) scratched glass slides. The root-mean-square roughness (RMS) is 60.5 and 267.9 nm for the figure (a) and (b) respectively.

Table S1 Fitted rise and decay time with their relative amplitudes (in %) for photobleaching (PB) bands of CdSe/CdTe NPLs in the upper panel of Figure 3b with bi-exponential decay with one growth function.

PB	τ_{rise}	τ_{decay1}	τ_{decay2}
CdSe	0.29 ± 0.02 ps	0.67 ± 0.05 ps (10%)	>> 1 ns (90%)
CdTe	0.29 ± 0.02 ps	0.72 ± 0.05 ps (45%)	>> 1 ns (55%)
CT	0.61 ± 0.05 ps		

Table S2 Fitted rise and decay time with their relative amplitudes (in %) for photobleaching (PB) bands of CdSe/CdSeTe NPLs in the lower panel of Figure 3b with bi-exponential decay with one growth function for CdSe photobleaching (PB) dynamics, and bi-exponential growth function for CdSeTe and CT PB dynamics.

PB	τ_{decay1}	τ_{decay2}	τ_{rise1}	τ_{rise2}
CdSe	0.35 ± 0.02 ps (32%)	>> 1 ns (68%)	0.21± 0.02 ps	
CdSeTe			0.22 ± 0.02 ps (65%)	0.50 ± 0.05 ps (35%)
CT			0.15 ± 0.01 ps (32%)	0.32 ± 0.02 ps (68%)

Table S3 Fitted amplitudes of Auger recombination and spontaneous emission (SE) for TA decay dynamics probed at CT states of CdSe/CdSeTe NPLs at different pump fluence using bi-exponential function of $\Delta A\ (t)=A_{Aug}*\exp(-t/\tau_{Aug})+ A_{SE}*\exp(-t/\tau_{SE})$ with fixed τ_{Aug}=360 ps and τ_{SE}= 9 ns. The shorter time constant of spontaneous emission τ_{SE} than the value measured by streak camera in Fig. 1c is mainly because of the much shorter time delay of TA.

Pump fluence ($\mu J\ cm^{-2}$)	A_{Aug} (Auger recombination)	A_{SE} (spontaneous emission)
8		100 %
16	32 %	68 %
32	37 %	63 %
64	51 %	49 %
96	78 %	22 %

Table S4 Fitted amplitudes of Auger recombination and spontaneous emission (SE) for TA decay dynamics probed at CT states of CdSe/CdTe NPLs at different pump fluence using bi-exponential function of $\Delta A(t)=A_{Aug}*\exp(-t/\tau_{Aug})+A_{SE}*\exp(-t/\tau_{SE})$ with fixed τ_{Aug}=160 ps and τ_{SE}= 7 ns. The shorter time constant of spontaneous emission τ_{SE} than the value measured by streak camera in Fig. 1c is mainly because of the short time delay for TA.

Pump fluence ($\mu J\ cm^{-2}$)	**A_{Aug}** (Auger-recombination)	**A_{SE}** (spontaneous emission)
8		100 %
32	18 %	82 %
64	28 %	72 %
96	46 %	54 %

Table S5 List of reported biexciton Auger lifetimes in various colloidal nanostructures, which were obtained by the similar biexciton lifetime extraction method using femtosecond transient absorption spectroscopy.

Material	**Shape**	**Biexciton Auger lifetime (ps)**	**Size**	**Refs**
CdSe	nanocrystals	6~360	1.2~4.1 nm (radius)	5
CdSe	nanocrystals	~5-150	-	6
CdSe	nanorods	~50-400	50~700 nm^3 (diameter)	
ZnSe/CdSe	core/shell nanocrystals	~8-30	-	
CdTe	nanocrystals	~2-90	2.6 ~ 3.1 nm (diameter)	7
CdSe/CdS	dot-in-rod nanorods	190-300	15~39 nm (length)	8
CdSe	4ML nanoplatelets	150-300	80~270 nm^2 (area)	9
InAs/CdSe/ZnSe	core/shell/shell nanocrystals	28~53	4.9~5.9 nm (diameter)	10
$CsPbBr_3$	nanocrystals	85	8.6 nm (diameter)	11
$MAPbBr_3$	nanocrystals	35-80	65~800 nm^3 (volume)	12
CdSe/CdTe	core/crown nanoplatelets	160	25.9 nm × 18.2 nm	this work

CdSe/CdSeTe	core/alloyed crown nanoplatelets	360	31.7 nm ×15.5 nm	

Reference

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