Supporting Information for

Synthesis of Highly Fluorescent InP/ZnS Small-Core/Thick-Shell Tetrahedral-Shaped Quantum Dots for Blue Light-Emitting Diodes

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Synthetic details of InP/ZnS QDs with tunable emissions

Chemicals

Tris(dimethylamino)phosphine ($(DMA)_3P$, 97%), oleylamine (80−90%), 1-octadecene (> 90%), zinc (II) stearate (Zn 10-12%) were purchased from Aladdin. Indium (III) chloride tetrahydrate (AR) and 1-dodecanethiol (98%) were purchased from Sinopharm Chemical Reagents Co. Ltd., CP. Zinc (II) iodide (98%) was purchased from Alfa Aesar.

Synthesis of InP QDs with tunable colors

a. Green InP QDs

(1) 100 mg (0.34 mmol) indium (III) chloride tetrahydrate, 215 mg (0.68 mmol) zinc (II) iodide and 208 mg (1.53 mmol) zinc (II) chloride are mixed in 5.0 ml (15.00 mmol) oleylamine (OAm) as the precursor solution. The mixed solution is stirred and degassed at 140 °C for an hour to make sure the indium and zinc precursors dissolving. After multi-process of nitrogen injection and degassing to ensure the system under inert atmosphere, the precursor solution is heated up to 200 °C. Then a volume of 1.45 ml phosphorus precursor (0.45 ml $(DMA)_3P$ (2.40 mmol) mixed with 1.00 ml oleylamine) is quickly injected in the precursor solution. The reaction is hold for 20 min, and stopped by extracting the hot solution in cold toluene.

(2) 100 mg (0.34 mmol) indium (III) chloride tetrahydrate and 495 mg (2.20 mmol) zinc (II) bromide are mixed in 5.0 ml (15.00 mmol) oleylamine (OAm) as the precursor solution. The mixed solution is stirred and degassed at 140 °C for an hour to make sure the indium and zinc precursors dissolving. After multi-process of nitrogen injection and degassing to ensure the system under inert atmosphere, the precursor solution is heated up to 200 °C. Then a volume of 1.45 ml phosphorus precursor (0.45 ml $(DMA)_3P$ (2.40 mmol) mixed with 1.00 ml oleylamine) is quickly injected in the precursor solution. The reaction is hold for 20 min, and stopped by extracting the hot solution in cold toluene.

b. Orange InP QDs

100 mg (0.34 mmol) indium (III) chloride tetrahydrate and 300 mg (2.20 mmol) zinc (II) chloride are mixed in 5.0 ml (15.00 mmol) oleylamine (OAm) as the precursor solution (the ratio of $ZnI_2/ZnCl_2$ is 0.44). The mixed solution is stirred and degassed at 140 °C for an hour to make sure the indium and zinc precursors dissolving. After multi-process of nitrogen injection and degassing to ensure the system under inert atmosphere, the precursor solution is heated up to 200 °C. Then a volume of 1.45 ml phosphorus precursor (0.45 ml $(DMA)_3P$ (2.40 mmol) mixed with 1.00 ml oleylamine) is quickly injected in the precursor solution. The reaction is hold for 20 min, and stopped by extracting the hot solution in cold toluene.

c. Red InP QDs

100 mg (0.34 mmol) indium (III) chloride tetrahydrate is mixed in 5.0 ml (15.00 mmol) oleylamine (OAm) as the precursor solution. The mixed solution is stirred and degassed at 140 °C for an hour to make sure the indium and zinc precursors dissolving. After multi-process of nitrogen injection and degassing to ensure the system under inert atmosphere, the precursor solution is heated up to 200 °C. Then a volume of 1.45 ml phosphorus precursor (0.45 ml $(DMA)_3P$ (2.40 mmol) mixed with 1.00 ml oleylamine) is quickly injected in the precursor solution. The reaction is hold for 20 min, and stopped by extracting the hot solution in cold toluene.

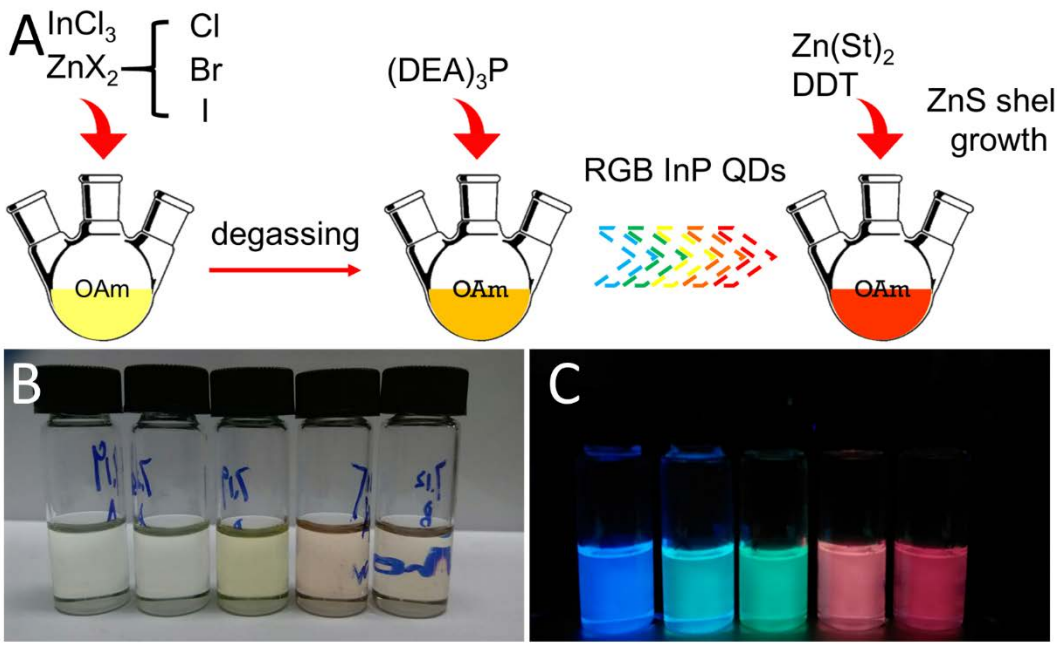

Figure S1. (A) The schematic picture for color tunable InP/ZnS QDs; (B) the as-synthesized InP/ZnS QDs with dispersions in toluene under room light; (C) the as-synthesized InP/ZnS QDs with dispersions in toluene excitation using an ultraviolet lamp.

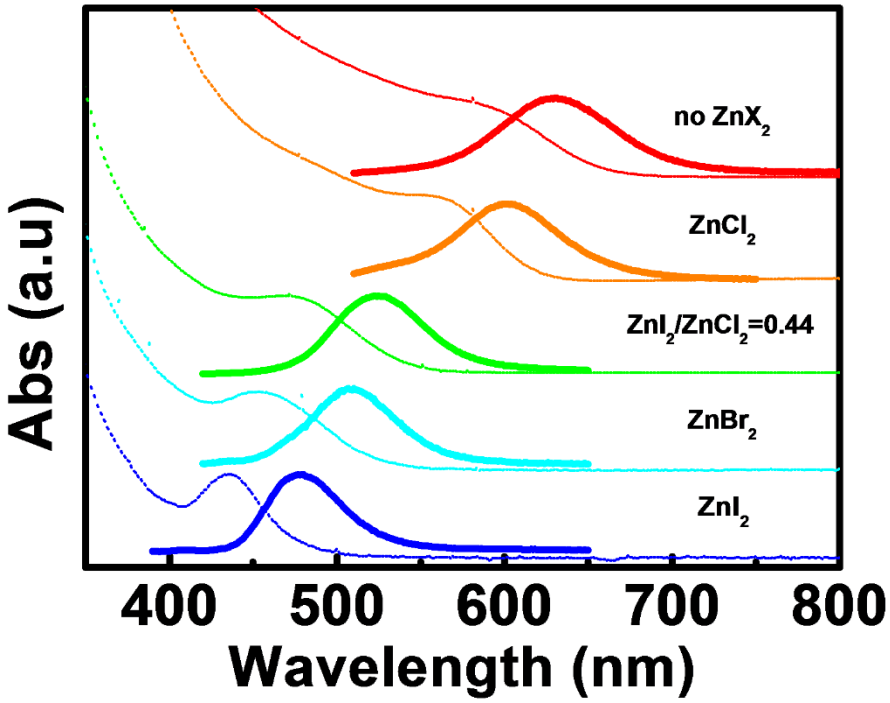

Figure S2. Optical absorption (dots) and PL emission (line) spectra of InP/ZnS synthesized by different methods (Experimental Section). The PL emission peak at 476, 508, 525, 602, and 631 nm, respectively.

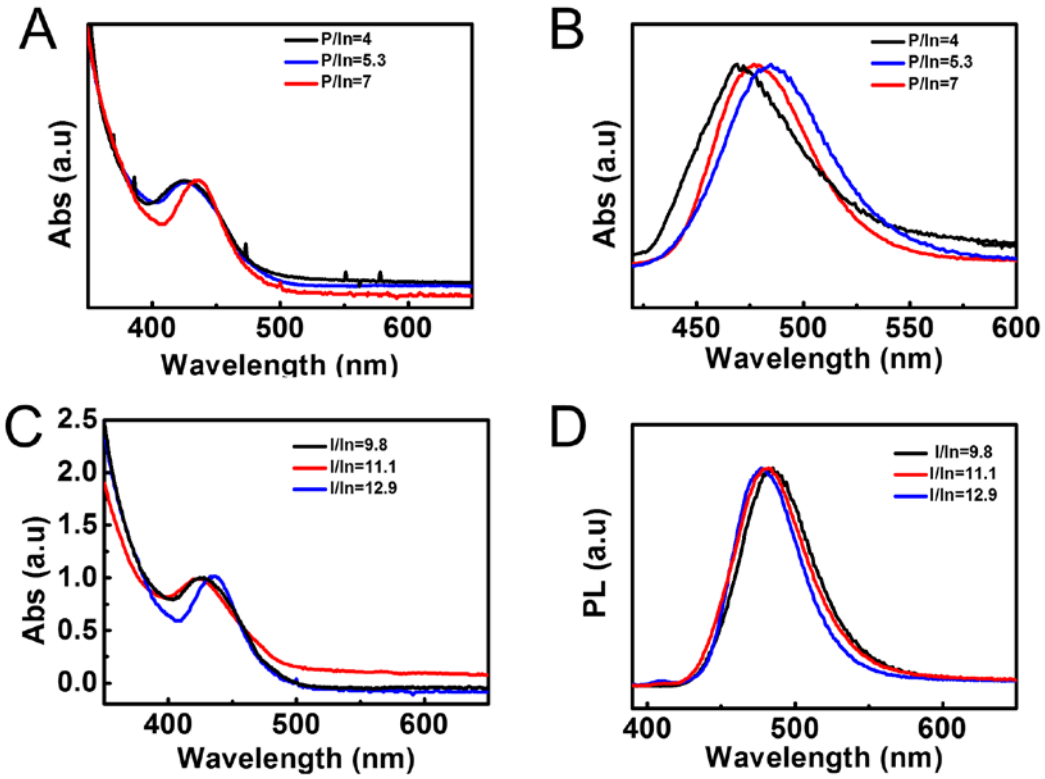

Figure S3. (A) Absorption spectra of InP/ZnS with different P/In; (B) PL spectra of InP/ZnS with different P/In. (A) Absorption spectra of InP/ZnS with different I/In; (B) PL spectra of InP/ZnS with different I/In.

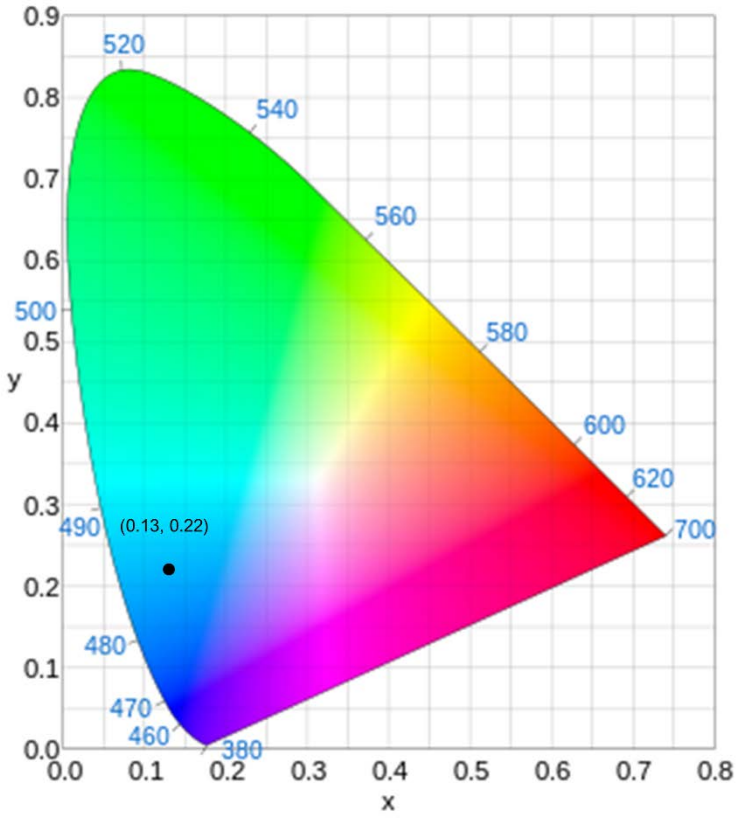

Figure S4. The CIE coordinate of the EL spectra of the LED device using blue-emitting InP/ZnS quantum dots as the active layer.

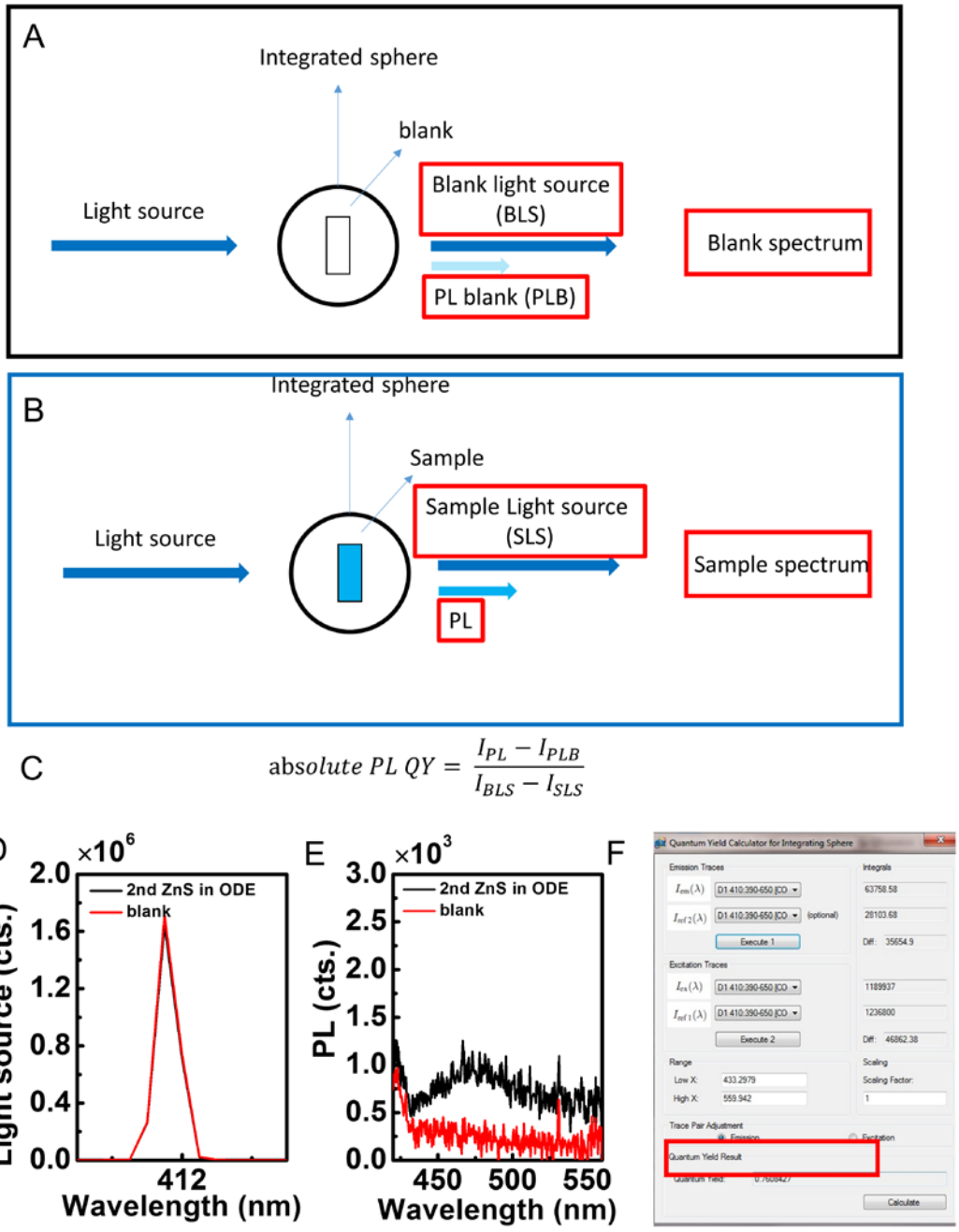

Figure S5. Absolute PL QY measurement. (A) Light source intensity of blank sample measurement. (B) Light source and PL intensity of sample measurement. (C) Absolute PL QY formula. (D) The light source spectra of blank sample and InP/ZnS after 2nd ZnS shell coating in ODE. (E) The PL spectra of blank sample and InP/ZnS after 2nd ZnS shell coating in ODE. (F) The absolute PL QY of InP/ZnS after 2nd ZnS shell coating in ODE.

Table S1. The absolute PL QY of color tunable InP/ZnS QDs.

	λ_{Ex}	PL QY
ZnI_2	420 nm	71.1%
$ZnBr_2$	410 nm	45.3%
$ZnI_2/ZnCl_2$=0.44	410 nm	50.1%
$ZnCl_2$	500 nm	51.2%
No $ZnCl_2$	500 nm	46.8%

Table S2. The time-resolved PL decays of InP/ZnS QDs with different P/In and I/In ratio.

	P/In=4 I/In=9.8	P/In=5.3 I/In=9.8	P/In=5.3 I/In=11.1	P/In=7 I/In=11.1	P/In=7 I/In=12.9
τ_1 (ns)	5.2	6.4	6.3	5.4	20.9
τ_2 (ns)	36.9	36.4	35.5	38.9	82.6
τ_3 (ns)	118.4	125.0	117.4	120.4	-
A1	0.29 (3.0%)	0.33 (4.6%)	0.30 (4.3%)	0.31 (3.6%)	0.64 (26.8%)
A2	0.66 (48.8%)	0.70 (55.2%)	0.69 (57.1%)	0.60 (49.6%)	0.44 (73.2%)
A3	0.20 (48.2%)	0.15 (40.2%)	0.14 (38.6%)	0.18 (46.8%)	-
τ_{avg} (ns)	75.2	70.7	65.9	75.9	66.1

Time-resolved PL decay curves were fitted[1] to a triexponential (see eqs 1 and 2) decay curves of

$$\tau=\tau_0 + A_1*\exp(-(x-x_0)/\tau_1) + A_2*\exp(-(x-x_0)/\tau_2) + A_3*\exp(-(x-x_0)/\tau_3) \quad (1)$$

The average lifetimes were calculated using

$$\tau_{avg} = (A_1\tau_1^2 + A_2\tau_2^2 + A_3\tau_3^2)/(A_1\tau_1 + A_2\tau_2 + A_3\tau_3) \quad (2)$$

Table S3. The time-resolved PL decay curves of InP/ZnS QDs.

	1st ZnS shell coating in OAm	2ndZnS shell coating in OAm	2nd ZnS shell coating in ODE	3rd ZnS shell coating in ODE
τ_1 (ns)	20.9	4.5	22.3	3.1
τ_2 (ns)	82.6	39.0	99.2	33.3
τ_3 (ns)	-	149.2	-	144.6
A1	0.64 (26.8%)	0.43 (3.2%)	0.65 (25.2%)	1.22 (7.1%)
A2	0.44 (73.2%)	0.62 (40.0%)	0.44 (74.8%)	0.52 (32.9%)
A3	-	0.23 (56.8%)	-	0.22 (60.0%)
τ_{avg} (ns)	66.1	100.5	79.8	97.8

Time-resolved PL decay curves were fitted to a triexponential (see eqs 1 and 2) decay curves of

$$\tau=\tau_0 + A_1*\exp(-(x-x_0)/\tau_1) + A_2*\exp(-(x-x_0)/\tau_2) + A_3*\exp(-(x-x_0)/\tau_3) \quad (1)$$

The average lifetimes were calculated using

$$\tau_{avg} = (A_1\tau_1^2 + A_2\tau_2^2 + A_3\tau_3^2)/(A_1\tau_1 + A_2\tau_2 + A_3\tau_3) \quad (2)$$

Table S4. The comparison of previously reported results with this work.

	$P_{precursors}$	λ_{em} of blue QDs	PL QY	Blue QD-LED
1. J. Mater. Chem., 2008,18, 2653-2656	$P(SiMe_3)_3$	480 nm	-	×
2. Adv. Mater., 2012, 24(30): 4180-4185	$P(SiMe_3)_3$	~480 nm	~48%	×
3. Nanotechnology, 2012, 23(48): 485609	$P(SiMe_3)_3$	475/485/497 nm	5%/10%/25 %	×
4. J. Am. Chem. Soc., 2012, 134 (8), pp 3804–3809	$P(SiMe_3)_3$	490 nm	15%	×
5. Chem. Mater. 2015, 27, 4893−4898	$(DEA)_3P$	-	-	×
6. J. Mater. Chem. C 2015, 3, 3582-3591.	$(DMA)_3P$	-	-	×
7. Angew. Chem. Int. Ed. 2016, 55, 3714-3718	$(DEA)_3P$	-	-	×
8. This work	$(DMA)_3P$	477	76%	√