Supporting Information

Water-stable All-inorganic Perovskite Nanocrystals with Nonlinear Optical Properties for Targeted Multiphoton Bioimaging

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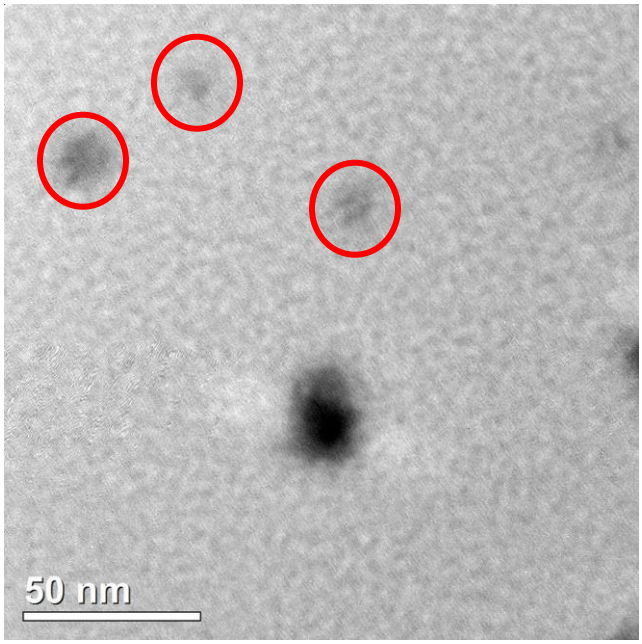

Figure S1: TEM image of $CsPbBr_3/SiO_2$ NCs added with 10 µl of TMOS during the reaction process. Free silica spheres were formed (circled in red) as a result of excess TMOS.

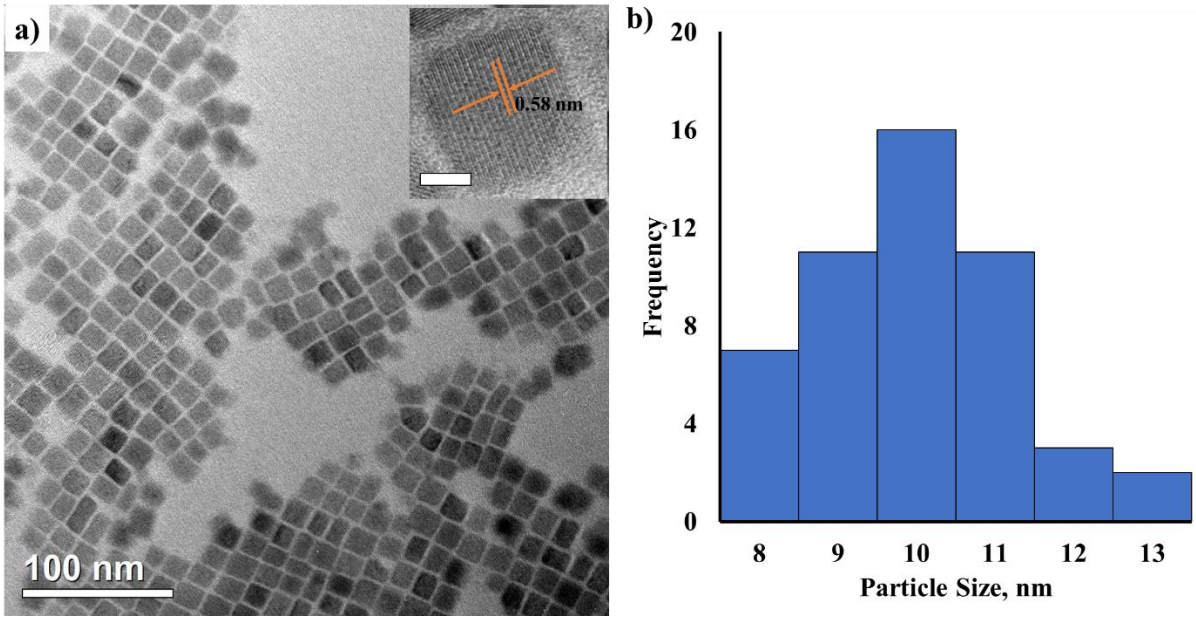

Figure S2: a) TEM images of $CsPbBr_3/SiO_2$ NCs after 1-minute of stirring, inset is the HRTEM of the NCs showing the lattice fringe, the scale is 5 nm. b) Size distribution of $CsPbBr_3/SiO_2$ NCs.

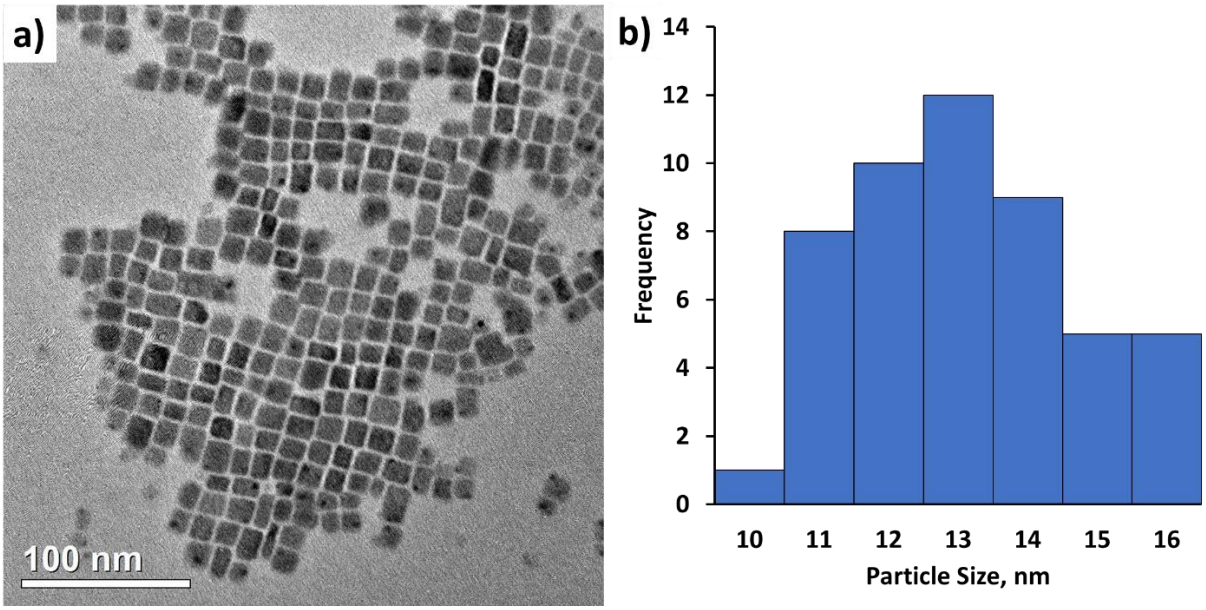

Figure S3: a) TEM image of $CsPbBr_3/SiO_2$ NCs after 120 minutes of stirring. b) Size distribution of $CsPbBr_3/SiO_2$ nanocrystals.

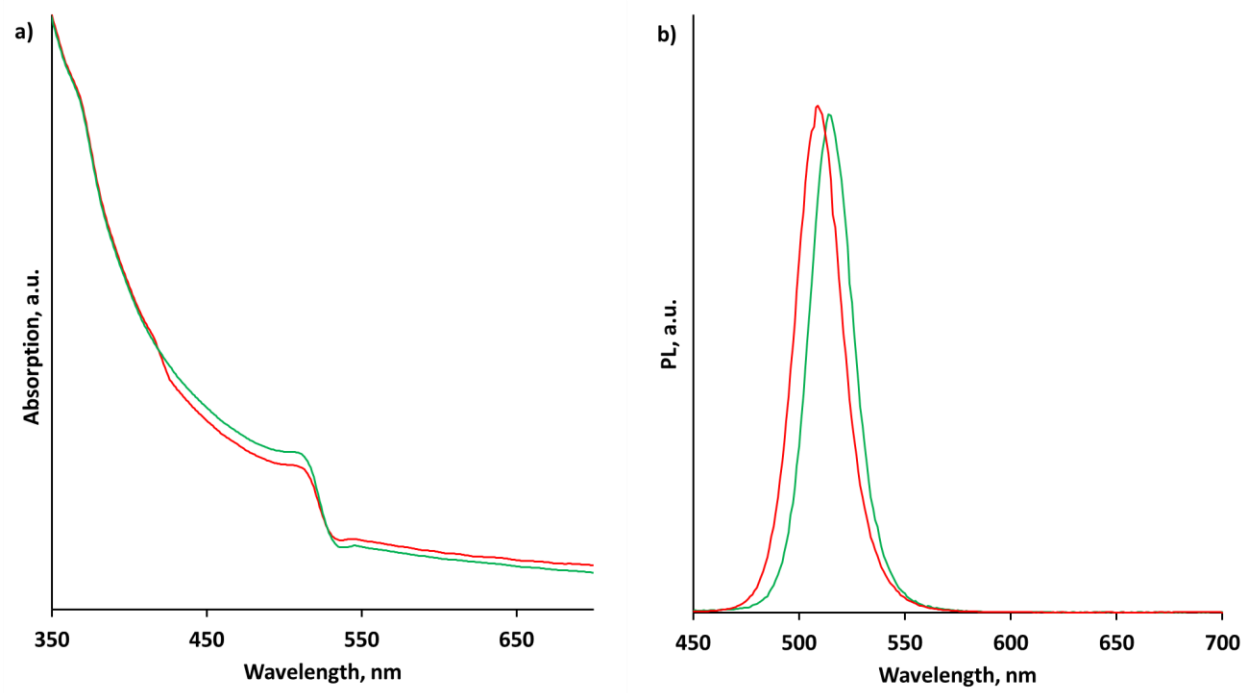

Figure S4: The absorption spectra (left) and PL spectra (right) of $CsPbBr_3/SiO_2$ (red) and $CsPbBr_3/SiO_2$/mPEG-DSPE (green).

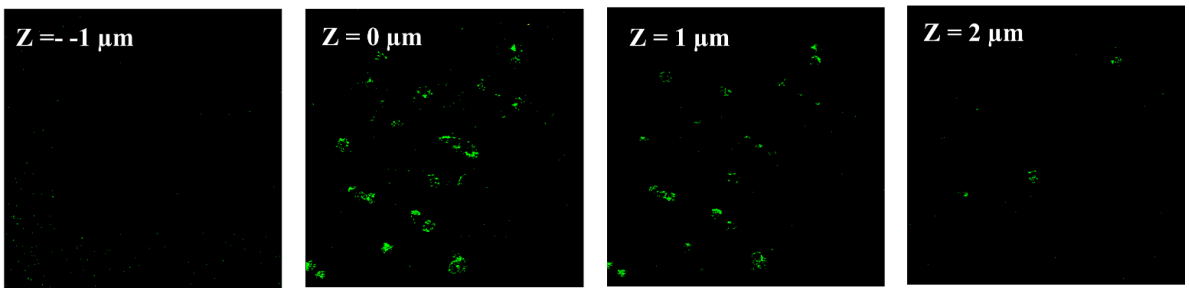

Figure S5: Z-axis scanning confocal images of Mia PaCa-2 cells cultured with $CsPBBr_3/SiO_2$/mPEG-DSPE NCs.

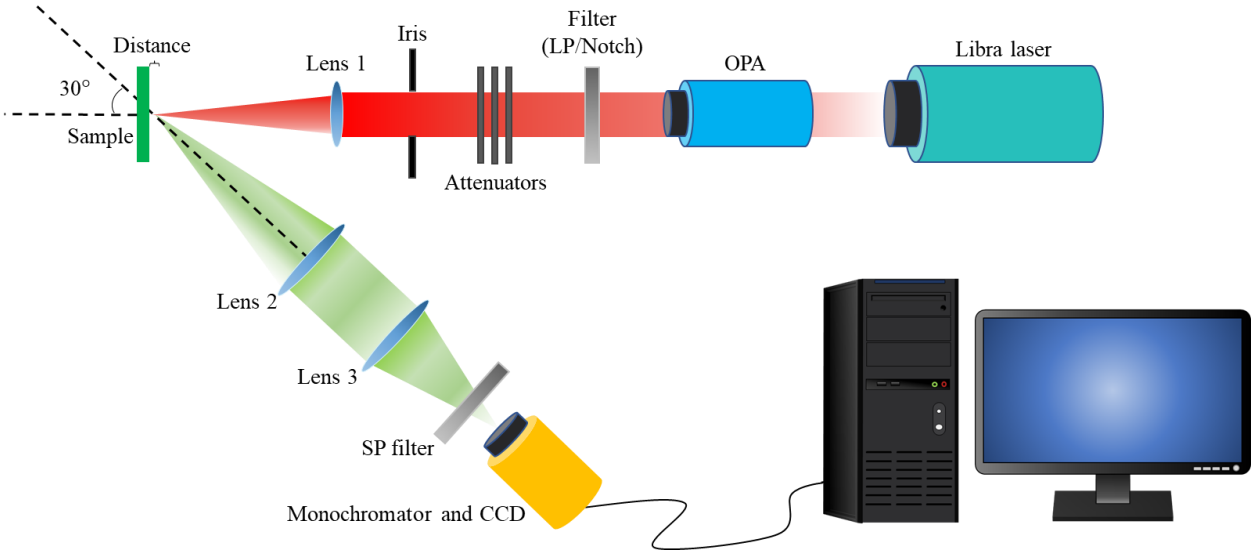

Figure S6: Schematic of the experimental setup.

Table S1: Comparison of the 2PA cross-sections of $CsPbBr_3/SiO_2$/mPEG-DSPE with the reported values from traditional semiconductor quantum dots and organic fluorophores.

Materials	**Excitation laser source**	**Absorption cross-section (10^5 GM)**	**Ref**
Carbon-based nanomaterials			
CDs	800 nm	3.9×10^{-1}	1
Graphene oxide	760 nm, 100 fs, 80 MHz	5.084×10^{-1}	2
Nitrogen-doped graphene QDs	800 nm, 100 fs 80 MHz	4.8×10^{-1}	3
II-VI inorganic semiconductor QDs			
CdS in aqueous solution	575-875 nm, 130 fs, 1kHz	7.9×10^{-3}	4
ZnSe in n-butanol	806 nm, 100 fs, 10Hz	$4.9 \times 10^{-2} – 1.2 \times 10^{-1}$	5
ZnSe /ZnS in n-butanol	806 nm, 100 fs, 10Hz	$5.1 \times 10^{-2} – 1.4 \times 10^{-1}$	5
ZnS in methanol	532 nm, 520 nm, 28 ps, 10 Hz	$1.7 \times 10^{-2} – 2.1 \times 10^{-2}$	6
II-VI inorganic semiconductor QDs			
PbS in toluene	950 – 2400 nm, 140 fs, 1 kHz	$\sim 1.0 \times 10^{-2}$	7
PbS in toluene	1120-2500 nm, 140 fs, 1 kHz	$\sim 1.0 \times 10^{-2} – 2.1 \times 10^{-1}$	8
III-V group inorganic semiconductor QDs			
InP/ZnS NCs in toluene	800 nm; 100 fs	$3.5 \times 10^{-2} – 6.2 \times 10^{-2}$	9
InP/ZnS NCs in toluene	750 - 1100 nm; 130 fs	$\sim 1.0 \times 10^{-4} – 2.2 \times 10^{-2}$	10
VIII-IV group inorganic semiconductor QDs			
Fe_3O_4 in toluene	780 nm; 330 fs	3×10^{-1}	11
Organic Molecules			

2,1,3-benzothiadiazole (BTD)-based D–p–A–p–D-type and star-burst-type fluorescent dye in toluene	800 nm, 120 fs	$1.7 \times 10^{-4} – 8.0 \times 10^{-4}$	[12]
Bis(styryl)benzene derivatives with various structural motifs	518 - 945 nm; 100 fs	$2.1 \times 10^{-3} – 3.7 \times 10^{-2}$	[13]
Symmetrical diphenylamino - fluorenes	590 -780 nm; 150 fs	$\sim 1.0 \times 10^{-3} – 6.8 \times 10^{-2}$	[14]
Methyl-substituted ladder-type poly(p-phenylene) (MeLPPP)	718 & 800 nm; 140 fs	$1.8 \times 10^{-1} – 4.0 \times 10^{-1}$	[15]
dendrimers of 4,4'-bis (diphenylamino) stilbene (DPAS)	715 - 780 nm; ~150 fs	$3.3 \times 10^{-3} – 1.1 \times 10^{-1}$	[16]
$CsPbBr_3/SiO_2$/mPEG-DSPE NCs	800 nm; 100 fs	$5.3 \pm \times 10^{-1}$	This work

Table S2: Comparison of the 3PA cross-sections of $CsPbBr_3/SiO_2$/mPEG-DSPE NCs with the reported values from traditional semiconductor quantum dots and organic fluorophores.

Materials	Excitation laser source	Absorption cross-section (10^{-74} cm^6s^2 $photon^{-2}$)	Reference
CdSe/CdS in toluene	1300 nm; 100 fs	4.3×10^{-4}	[17]
CdSe/CdS/ZnS in toluene	1300 nm; 100 fs	2.8×10^{-3}	[17]
CdSe in hexane	160 fs; 1300 nm	$\sim 10^{-4}$	[18]
CdS in chloroform	100 fs; 900-1000 nm	$\sim 10^{-5}$	[19]
ZnS in aqueous solution	120 fs; 620-780 nm	$\sim 2.7 \times 10^{-4}$	[20]
Cu-doped ZnSe/ZnS	700-800 nm; 46 fs	3.7×10^{-3}-5.5×10^{-4}	[21]
Mn-doped ZnSe/ZnS	700-800 nm; 46 fs	4×10^{-3}-4.9×10^{-4}	[21]
ZnS	950 nm; 140 fs	$1.6 \pm 0.6 \times 10^{-5}$	[22]
Fluorene-containing ferrocene derivatives	1260-1600 nm; 160 fs	4.6×10^{-7}-8.3×10^{-7}	[23]
Ri(9,9-diethyl-9 H-fluorenyl)amine	1160-1380 nm; 100 fs	$\sim 2.1 \times 10^{-7}$ - 4.0×10^{-5}	[24]
(E)-3-(4-(2-(1-hexyl-4-methyl-1H-imidazol-5-yl)vinyl)pyridinium-1-yl)propyl sulphate (IPPS)	1197 nm; 120 fs	3.7×10^{-6}	[25]
Highly extended diphenylamino-end-capped ladder-type oligo- (p-phenylene)s	1230-1330 nm; 120 fs	2×10^{-3} - 4.6×10^{-2}	[26]
$CsPbBr_3/SiO_2$/mPEG-DSPE NCs in water	1200 nm; 100 fs	$5.0 \pm 0.4 \times 10^{-3}$	This work

Table S3: Summary of the 1PA, 2PA, and 3PA cross-sections of $CsPbBr_3/SiO_2$/mPEG-DSPE NCs in water

Excitation Laser Source	Absorption Cross-section
400 nm; 100 fs	$9.8 \pm 0.4 \times 10^{-14}$ cm^2
800 nm; 100 fs	$5.3 \pm 0.4 \times 10^4$ GM
1200 nm; 100 fs	$5.0 \pm 0.4 \times 10^{-77}$ $cm^6 s^2$ $photon^{-2}$

Table S4: Comparison of the aqueous stabilty of $CsPbBr_3/SiO_2$/mPEG-DSPE NCs with other reported works.

Perovskite Nanocrystals	Encapsulation	Size	Aqueous Stability	Water Dispersibility	Ref
$CsPbBr_3@NH_4Br$	Ammonium bromide	In micron range	Retained ~40% of PL after 3.5 h of dispersion in water	Yes	[27]
MC-$CsPbBr_3$	Polyethylene wax	~800 nm	Retained ~18.09% of PL after 90 minutes of immersion in water	No	[28]
$CsPbBr_3$ PNC@SSIP	Organosilica nanoparticles	~100 nm	Retained 80% of PL after 120 h of immersion in water	No	[29]
$CsPbBr_3@NaYF_4$ and $CsPbBr_3@ZnO$	$NaYF_4$ and ZnO	$CsPbBr_3$ @$NaYF_4$ (500 nm diameter), $CsPbBr_3$ @ZnO (length of 600 nm, width of 200 nm)	Still show green emission after 30 minutes immersion	No	[30]
$CsPbBr_3/Cs_4PbBr_6$ /$BaSO_4$	$BaSO_4$	20-100 nm	Retained 90% of PL after 1 day storage in PBS	Yes	[31]

$CsPbX_3$ NCs@MHS	Polyvinyl pyrrolidone and polystyrene	3-5 μm	Not quantified	Yes	[32]
$CsPbBr_3$@polymer fibers	polystyrene	~800 nm	Retained 70% of PL after 192 h	No	[33]
$CsPbBr_3/CsPb_2Br_5$	$CsPb_2Br_5$	~10 nm	~50% after kept in a sealed cavity with 100% humidity	No	[34]
$CsPbBr_3/SiO_2$	SiO_2	-	Retained 80% after 25 h	Yes	[35]
$CsPbBr_3/CsPb_2Br_5$	$CsPb_2Br_5$	195 nm	Retained 45% after 110 h	Yes	[36]
$CsPbBr_3/SiO_2$/ mPEG-DSPE	SiO_2 and mPEG-DSPE	~30 nm	Retained 80% after 2 weeks storage	Yes	This work

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