Supporting information for

In-situ Growth of α-CsPbI₃ Perovskite Nanocrystals on the Surface of Reduced Graphene Oxide with Enhanced Stability and Carrier Transport Quality

Qi Zhang¹, Hui Nan¹, Yangying Zhou¹, Youchen Gu¹, Meiqian Tai¹, Yaxuan Wei¹, Feng Hao^{2*}, Jianbao Li^{3,1}, Dan Oron^{4,*} and Hong Lin^{1,*}

¹ State Key Laboratory of New Ceramics & Fine Processing, School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

² School of Materials and Energy, University of Electronic Science and Technology of China, Chengdu 610054, China

³ State Key Laboratory of Marine Resource Utilization in South China Sea, Materials and Chemical Engineering Institute, Hainan University, Haikou 570228, China

⁴ Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel

Table S1. Structural parameters derived from the corresponding XRD patterns of α -CsPbI₃ NCs and α -CsPbI₃ NCs/rGO heterostructures. The crystal sizes of α -CsPbI₃ NCs are calculated by Debye-Scherrer equation. (Instrument correction value for instrumental broadening changes =0.1252 °)

	Full width at half maximum of (200) Peak, °	Calculated crystal size, nm
α-CsPbI ₃ NCs	0.87	10.64
α-CsPbI ₃ NCs/rGO	0.79	11.83



Figure S1. (A) TEM image of α -CsPbI₃ NCs/rGO heterostructures with a few Pb-nanoparticles appeared on the surface of rGO. (B) HRTEM image of α -CsPbI₃ NCs. The lattice figure of these NCs shows a value of ~3.21 Å, which is consistent with the crystal structure of α -CsPbI₃ perovskite. (C) TEM image of α -CsPbI₃ NCs/rGO heterostructures, and rGO was reduced by thermal-reduced reaction without FeI₂ treatment. Thus α -CsPbI₃ NCs showed a stochastic distribution on the surface of rGO and their shapes were not regular.

Table S2. Element contents derived from the corresponding XPS spectra of α -CsPbI₃ NCs and α -CsPbI₃ NCs/rGO. The element contents are normalized to investigate the effect of *in-situ* growth method on ligands amounts on the surface of α -CsPbI₃ NCs (Pb contents can be regarded as 1). It is clear that the amount of α -CsPbI₃ NCs obviously increased and the amount of C and O elements related to ligands significant decrease.

	α -CsPbI ₃ NCs	α-CsPbI ₃ NCs/rGO
Pb 4f	1.00	1.00
C 1s	63.11	17.37
N 1s	2.00	0.73
O 1s	25.04	1.71
I 3d	4.01	4.34
Cs 3d	1.00	1.23



Figure S2. XRD patterns of α -CsPbI₃ NCs as a function of storage times stored in dark, low temperature (~4 °C) and low humidity (~1% RH) conditions without encapsulation. The recording storage times were ~0 h, ~168 h, ~336 h and ~672 h. The standard XRD patterns of FTO, *o*-CsPbI₃ (#161480) and α -CsPbI₃ (#161481) are also listed to identify the structural transitions of as-synthesized α -CsPbI₃ NCs. These XRD patterns showed that the α -CsPbI₃ NCs can easily transform into *o*-CsPbI₃ even when stored in dark, low temperature (~4 °C) and low humidity (~1% RH) conditions without encapsulation, completely losing their ideal crystal structure of optically active materials.



Figure S3. TEM images of α -CsPbI₃ NCs as a function of storage times stored in dark, low

temperature (~4 °C) and low humidity (~1% RH) conditions without encapsulation. The recording storage times were (A) ~0 h, (B) ~168 h, (C) ~336 h and (D) ~672 h.



Figure S4. (A) UV-Visible absorption, (B) steady-state PL, and (C) time-resolved PL spectra of α -CsPbI₃ NCs as a function of storage times stored in dark, low temperature (~4 °C) and low humidity (~1% RH) conditions without encapsulation. The recording storage times were ~0 h, ~168 h, ~336 h and ~672 h. These UV-Visible absorption spectra showed a significant blue-shift of the absorption peaks when the storage time increased, thus narrowing their coverage of the visible region. Meanwhile, when the storage time increased, these PL intensities decreased and the PL bands showed a blue-shift, and their lifetime of α -CsPbI₃ NCs reduced. Particularly, when the storage time approaches ~672 h, the band gap of *o*-CsPbI₃ showed a value of ~2.64 eV, and the intensity of CsPbI₃ NCs almost disappeared, and their lifetime were hardly to detect in the testing conditions, completely losing their optoelectronic properties of optically active materials.

Table S3. Peak positions and Lifetime derived from the corresponding steady-state PL and timeresolved PL spectra of α -CsPbI₃ NCs as a function of storage times stored in dark, low temperature (~4 °C) and low humidity (~1% RH) conditions without encapsulation.

	Steady-state PL	Time-res			
	Peak position, nm	τ_1 , ns	τ_2 , ns	τ_3 , ns	τ_{aver} , ns
0h	706.00	10.09	29.59	104.92	32.10
168h	688.00	7.49	29.40	97.74	25.45
336h	680.00	2.01	14.01	51.63	10.57
672h	-	-	-	-	-



Figure S5. The color of α -CsPbI₃ NCs as a function of storage times placed in deionized water without encapsulation.

Table S4. Optoelectronic parameters derived from corresponding UV-Visible absorption, steadystate PL and time-resolved PL spectra of α -CsPbI₃ NCs and α -CsPbI₃ NCs/rGO heterostructures films.

	UV-Visible	UV-Visible Steady-state PL Time-resolved PL				
	Band gap, eV	Peak Position, nm	τ_1 , ns	τ_2 , ns	τ_3 , ns	τ_{aver} , ns
α-CsPbI ₃ NCs	1.72	706.00	10.09	29.59	104.92	32.10
α-CsPbI ₃ NCs/rGO	1.74	688.00	12.29	45.28	125.48	67.77

Table S5. Structural parameters and Lifetime derived from the corresponding XRD patterns and time-resolved PL spectra of α -CsPbI₃ NCs and α -CsPbI₃ NCs/rGO stored in ambient conditions (room temperature of ~25 °C and a humidity of ~25% RH conditions) for ~4 weeks without encapsulation. (Instrument correction value for instrumental broadening changes =0.1252 °)

	XRD	Time-resolved PL				
	Full width at half maximum of (200) Peak, °	Calculated crystal size, nm	τ_1 , ns	τ_2 , ns	τ_3 , ns	τ_{aver} , ns
α-CsPbI ₃ NCs	-	-	-	-	-	-
α-CsPbI ₃ NCs/0.2-rGO	0.74	12.83	10.93	39.01	111.17	49.39

	0 weeks, %	1 weeks, %	2 weeks, %	3 weeks, %	4 weeks, %
α-CsPbI ₃ NCs	11.22	6.11	3.53	0.86	0.18
α-CsPbI ₃ NCs/rGO	10.74	10.22	9.43	8.34	7.28

Table S6. PLQYs of α -CsPbI₃ NCs and α -CsPbI₃ NCs/rGO heterostructures films stored in ambient conditions for ~4 weeks without encapsulation.