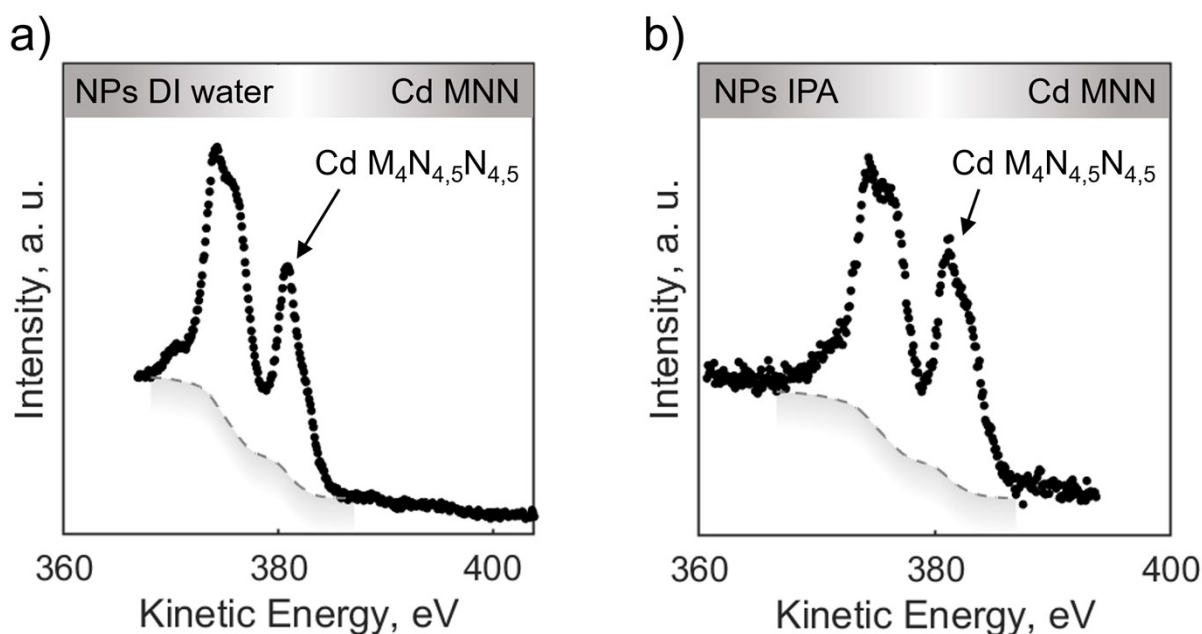


Supporting Information for

## Solvent-Directed Femtosecond Laser Ablation: Tuning Phase and Defect Engineering in Hybrid CdPS<sub>3</sub>/CdS Nanostructures

Compound Solvent	CdPS <sub>3</sub> (C2/m)			CdS (P6 <sub>3</sub> mc, Wurtzite)		
	hkl	d <sub>exp</sub> , Å	d <sub>hkl</sub> , Å	hkl	d <sub>exp</sub> , Å	d <sub>hkl</sub> , Å
DI water	1 1 -2	3.30	3.24			
	0 3 0	3.10	3.09			
	1 3 -2	2.46	2.47			
	0 2 3	2.04	2.09			
	0 6 0	1.78	1.81			
	0 0 4	1.73	1.70			
		0 2 2	2.90	2.88	1 0 0	3.62
ACN	1 3 1	2.65	2.67	1 0 1	3.17	3.19
	0 2 3	2.08	2.09	0 1 3	1.90	1.92
	1 3 -3	1.96	1.97	1 1 2	1.77	1.78
	0 6 0	1.77	1.81	1 2 3	1.15	1.17
	1 3 1	2.63	2.67	1 0 0	3.59	3.61
	1 3 -2	2.44	2.47	1 0 1	3.17	3.19
IPA	0 2 3	2.10	2.09	1 0 2	2.44	2.47
	0 6 0	1.77	1.81	0 1 3	1.90	1.92
				1 1 2	1.77	1.78

**Supplementary Table S1.** Interplanar spacings  $d_{\text{exp}}$  and calculated  $d_{\text{hkl}}$  distances determined from SAED patterns in Figs.2d,e,f of the main text.



**Supplementary Figure S1.** Measured XAES spectra Cd MNN for PLAL-produced samples in a) DI water and b) IPA. Shirley baselines denoted as dashed lines are used for the whole Cd MNN spectra.

Compound	Cd $3d_{5/2}$ Binding Energy, eV	Cd $M_4N_{4,5}N_{4,5}$ Kinetic Energy, eV	Reference
CdPS3	405.6	380.8	[S1]
CdS	405.3	381.3	[S1]
CdO	405.2	382.2	[S2]
Cd	405	384	[S1]
NPs in DI water	405.71	380.89	This study
NPs in IPA	405.46	381.15	This study
Cd <sup>0</sup> sites	405.46	383.19	This study

**Supplementary Table S2.** Cd  $d_{3/2}$  core level binding energy positions and kinetic energies of the Cd  $M_4N_{4,5}N_{4,5}$  Auger transition in cadmium compounds.

## References

- [S1] Curró, G. M., Vincenzo Grasso, and Letteria Silipigni. "X-ray photoelectron spectroscopy of CdPS3." *Journal of applied physics* 84.12 (1998): 6693-6697.
- [S2] Chastain, Jill, and Roger C. King Jr. "Handbook of X-ray photoelectron spectroscopy." Perkin-Elmer Corporation 40.221 (1992): 25.