## **Supporting Information**

## Synthesis of Nd<sup>3+</sup>/Yb<sup>3+</sup> Sensitized Upconversion Core-shell Nanocrystals with Optimized Hosts and Doping Concentrations

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**Figure S1.** Size distributions of the NaYF<sub>4</sub> NCs (seeds) and the core-shell NaYF<sub>4</sub>@NaGdF<sub>4</sub> NCs. Average diameters of the NaYF<sub>4</sub> NCs and NaYF<sub>4</sub>@NaGdF<sub>4</sub> NCs are about 32.9nm and 42.7nm, respectively. This data confirms the uniformity of these core-shell NCs and the increase in diameter after the shell growth.



**Figure S2.** XRD patterns of (A) the NaYF<sub>4</sub>:Yb,Tm seeds and (B) the prepared NaYF<sub>4</sub>:Yb,Tm@NaGdF<sub>4</sub>:Yb,Nd NCs. All diffraction peaks of the NaYF<sub>4</sub>:Yb,Tm NCs can be clearly indexed to the pure hexagonal NaYF<sub>4</sub> crystal (JCPDS No. 28-1192). XRD patterns of the NaYF<sub>4</sub>:Yb,Tm@NaGdF<sub>4</sub>:Yb,Nd NCs were much similar to those of the core NCs, albeit a little stronger in intensity. The increased intensity could have resulted from the size increase of the NCs and the similar crystal structure between NaYF<sub>4</sub> and NaGdF<sub>4</sub> (JCPDS No. 27-0699).

Element	Gd	Y	Yb	Nd	Tm	Na	F (1s)	C (1s)	0
	(3d)	(4d)	(4d)	(4d)	(4d)	(1s)			(1s)
Content	18.13	8.37	3.93	2.12	0.01	13.47	37.35	12.83	3.79
(at.%)									

TableS1.ElementcontentsofthesurfaceofNaYF4:Yb(20%),Tm(0.5%)@NaGdF4:Yb(10%),Nd(10%)determinedbyXPSanalysis.

The XPS analysis was also employed to investigate the surface information of the core-shell Y@Gd NCs. Though all the lanthanide elements could be detected in the fine-scan mode, it should be noted that the Gd element is predominant on the surface (18.13 at.%) of the core-shell NCs, which is significantly higher than the element Y (8.37 at.%). These XPS results imply that the surface of the core-shell nanocrystals mainly consists of NaGdF<sub>4</sub> shell. The signals from C and O are from the surface adsorbed oleic acids (OA) on the core-shell NCs during synthesis.



**Figure S3**. UC emission spectra of NaYF<sub>4</sub>:Yb(20%),Tm(0.5%) and NaGdF<sub>4</sub>:Yb(20%),Tm(0.5%) under 980nm excitation. This data shows that NaYF<sub>4</sub> is a better host for UC fluorescence than NaGdF<sub>4</sub> for Yb<sup>3+</sup>/Tm<sup>3+</sup> codoping when they have a similar size, doping level and the same hexagonal phases.



**Figure S4**. Comparison of UC emission intensities of NaYF<sub>4</sub>:Yb,Tm@NaYF<sub>4</sub>:Yb,Nd UCNs under 980nm excitation by changing the doping concentration of different ions: (A) Yb<sup>3+</sup> in the core; (B) Tm<sup>3+</sup> in the core; (C) Yb<sup>3+</sup> in the shell; (D) Nd<sup>3+</sup> in the shell.