Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Enhancing energy migration upconversion through migratory interlayer in core-shell-shell nanostructure towards latent fingerprinting

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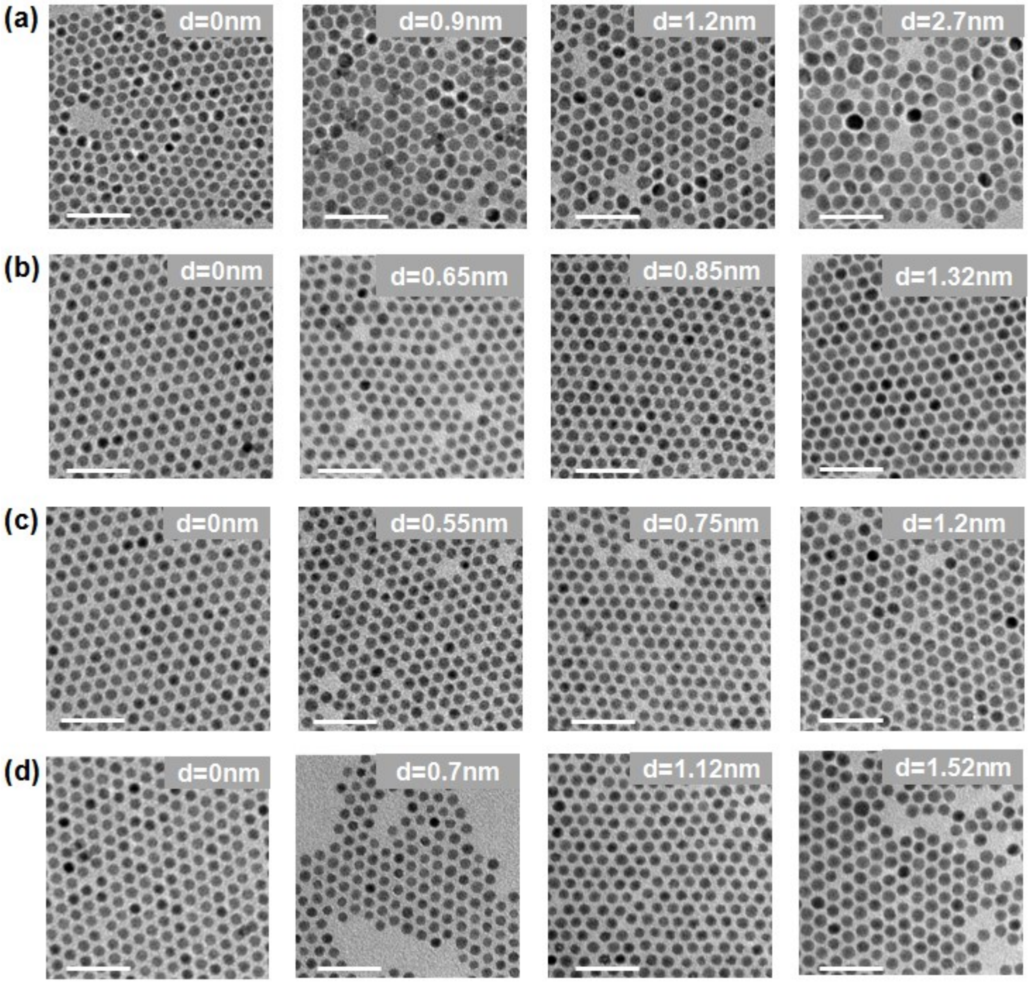

Figure S1. TEM images of the samples with a fine tuning of the $NaGdF_4$ interlayer thickness for various emitters: (a) Sm^{3+}, (b) Nd^{3+}, (c) Eu^{3+} and (d) Tb^{3+} in the outermost shell. The corresponding interlayer thickness "d" is given in the images. Scale bars, 50 nm.

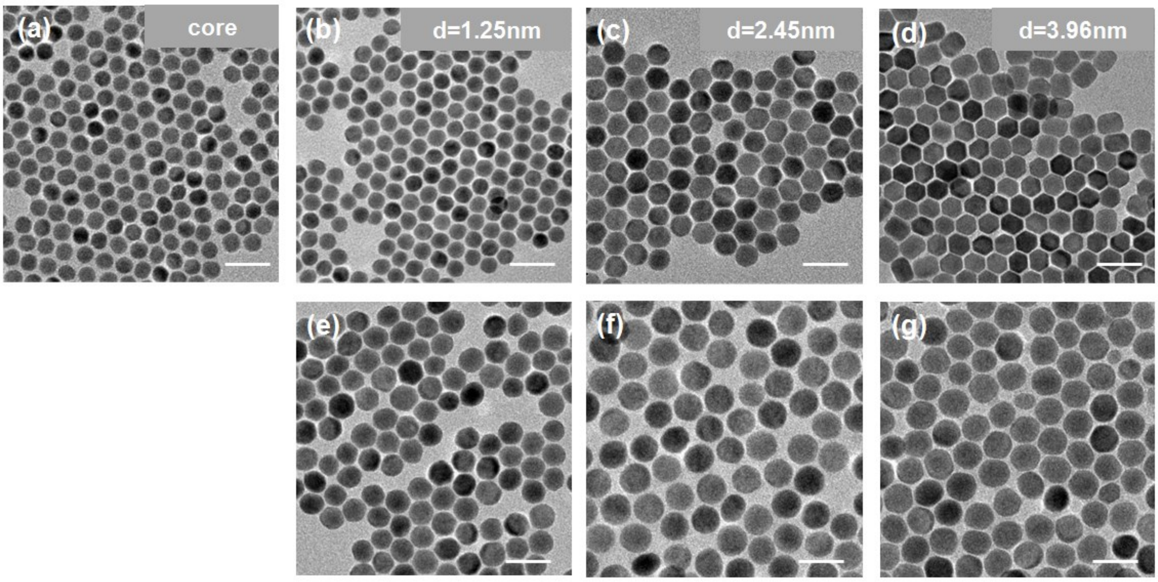

Figure S2. TEM images of $NaGdF_4$:Yb/Tm@$NaGdF_4$@$NaGdF_4$:Dy core-shell-shell nanoparticles with $NaGdF_4$ thickness of (b) 1.25 nm, (c) 2.45 nm, (d) 3.96 nm and their resultant core-shell-shell nanoparticles (e-g), respectively. Note that (a) shows the $NaGdF_4$:Yb/Tm core seeds for comparison. Scale bars, 100 nm.

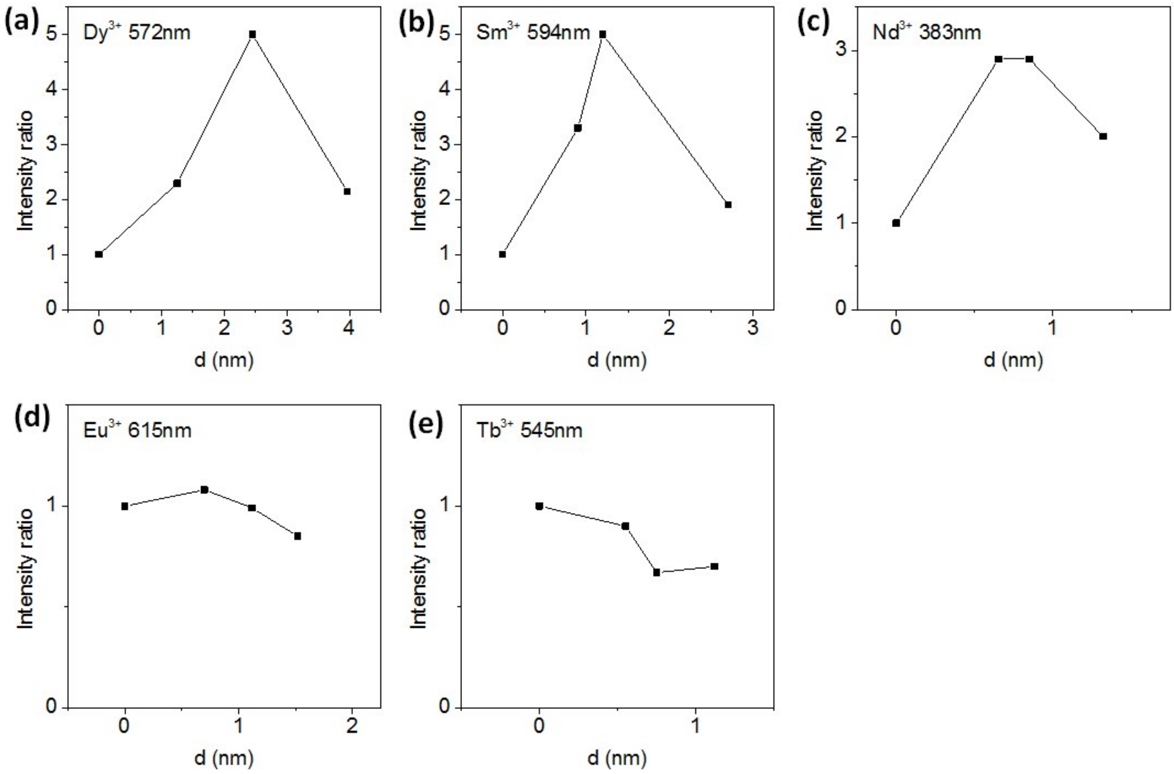

Figure S3. (a-e) Dependence of the typical upconversion emission intensity of (a) Dy^{3+} (572 nm), (b) Sm^{3+} (594 nm), (c) Nd^{3+} (383 nm), (d) Eu^{3+} (615 nm) and (e) Tb^{3+} (545 nm) on interlayer thickness for the $NaGdF_4$:Yb/Tm@$NaGdF_4$@$NaGdF_4$:A (A=Dy, Sm, Nd) core-shell-shell nanoparticles under 980 nm excitation, respectively.

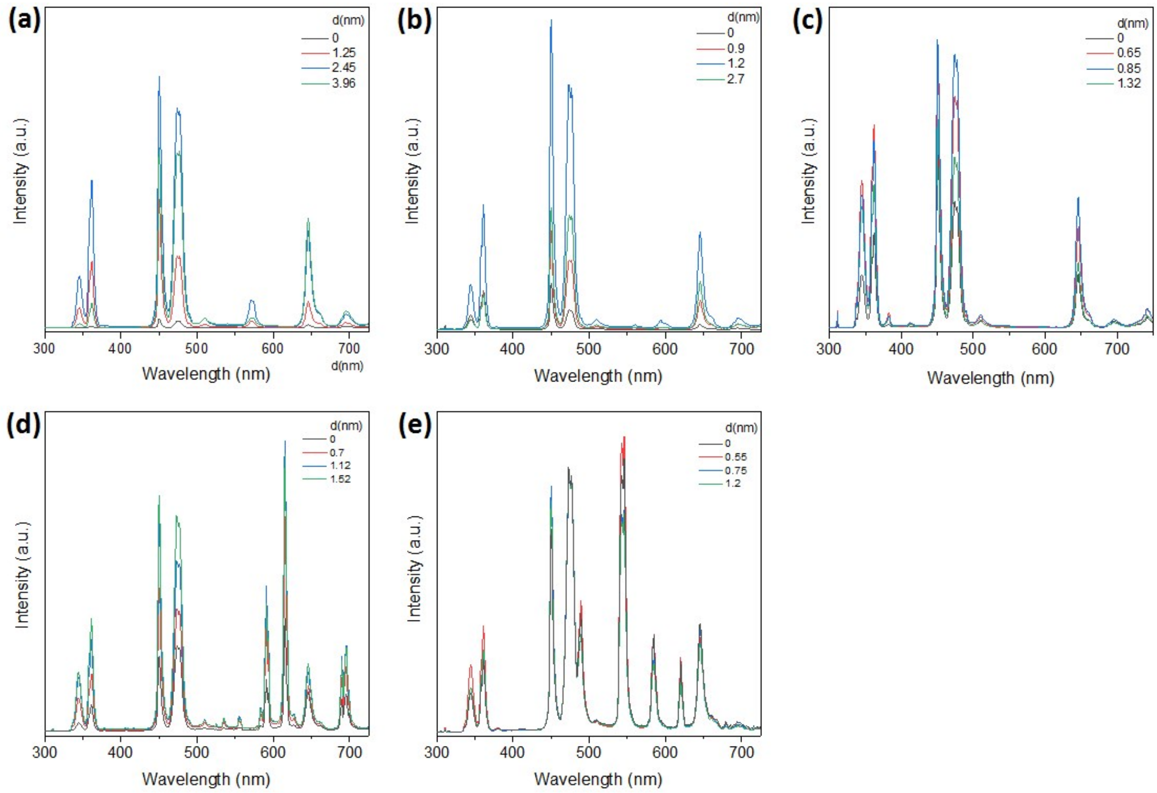

Figure S4. (a-e) Upconversion emission spectra recorded from the $NaGdF_4$:Yb/Tm@ $NaGdF_4$@$NaGdF_4$:A (A=Dy^{3+}, Sm^{3+}, Nd^{3+}, Eu^{3+}, Tb^{3+}) core-shell-shell nanoparticles under 980 nm excitation.

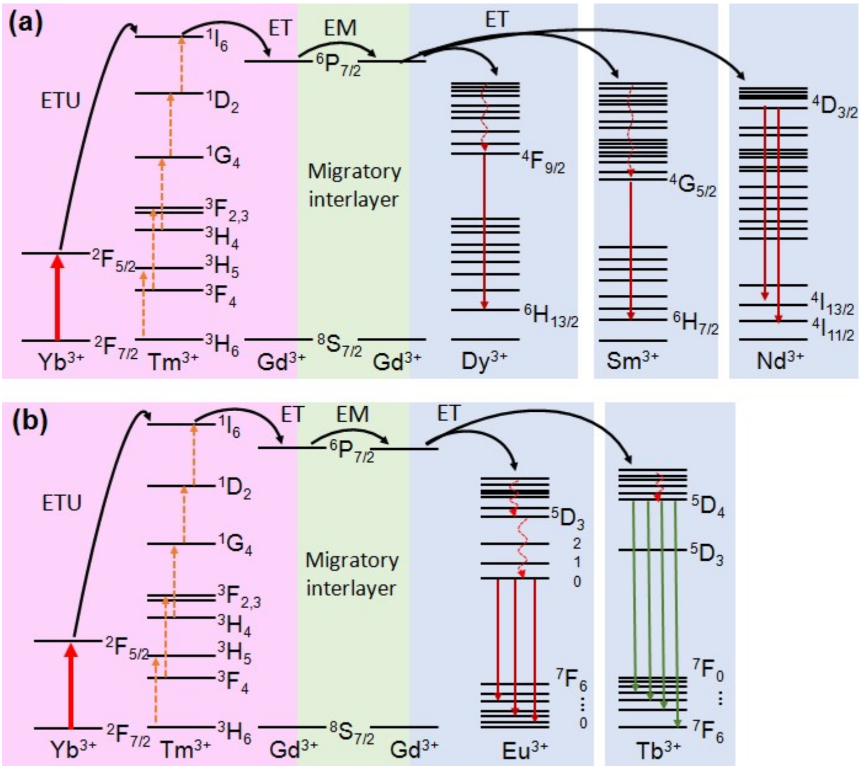

Figure S5. (a,b) Schematic of total energy transport process for photon upconversion in the $NaGdF_4$:Yb/Tm@$NaGdF_4$@$NaGdF_4$:A (A=Dy^{3+}, Sm^{3+}, Nd^{3+}, Eu^{3+}, Tb^{3+}) core-shell-shell nanoparticles under 980 nm excitation.

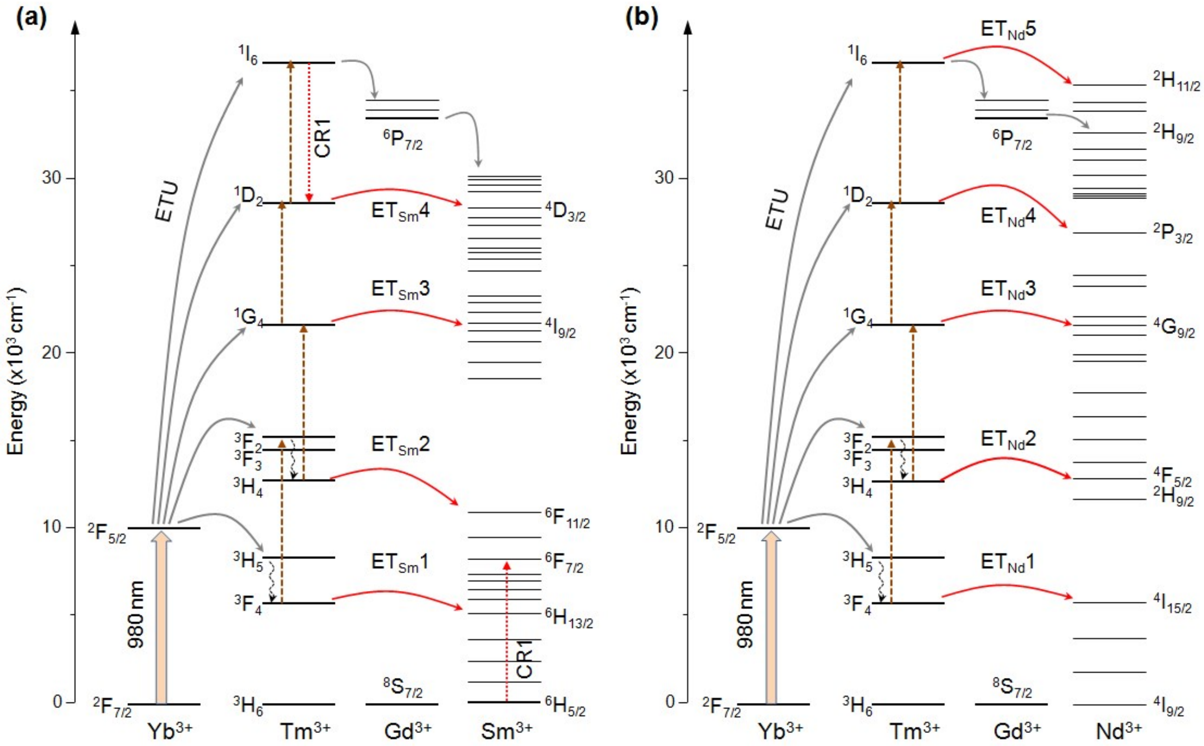

Figure S6. Schematic of possible quenching processes bewteen Tm^{3+} and the emitters of (a) Sm^{3+} and (b) Nd^{3+} at interfacial region in the $NaGdF_4$:Yb/Tm@$NaGdF_4$:A (A=Sm, Nd) core-shell nanoparticles under 980 nm excitation. Note that only the leading energy levels of the emitters are marked. $ET_{Sm}1$: Tm^{3+} ($^3F_4$) → Sm^{3+} ($^6H_{13/2}$), $ET_{Sm}2$: Tm^{3+} ($^3H_4$) → Sm^{3+} ($^6F_{11/2}$), $ET_{Sm}3$: Tm^{3+} ($^1G_4$) → Sm^{3+} ($^4I_{9/2}$), $ET_{Sm}4$: Tm^{3+} ($^1D_2$) → Sm^{3+} ($^4D_{3/2}$), CR1: [Tm^{3+} ($^1I_6$); Sm^{3+} ($^6H_{5/2}$)] → [Tm^{3+} ($^1D_2$); Sm^{3+} ($^6F_{7/2}$)]; $ET_{Nd}1$: Tm^{3+} ($^3F_4$) → Nd^{3+} ($^4I_{15/2}$), $ET_{Nd}2$: Tm^{3+} ($^3H_4$) →Nd^{3+} ($^4F_{5/2}$,$^2H_{9/2}$), $ET_{Nd}3$: Tm^{3+} ($^1G_4$) → Nd^{3+} ($^4G_{9/2}$), $ET_{Nd}4$: Tm^{3+} ($^1D_2$) → Nd^{3+} ($^2P_{3/2}$), $ET_{Nd}5$: Tm^{3+} ($^1I_6$) → Nd^{3+} ($^2H_{11/2}$).

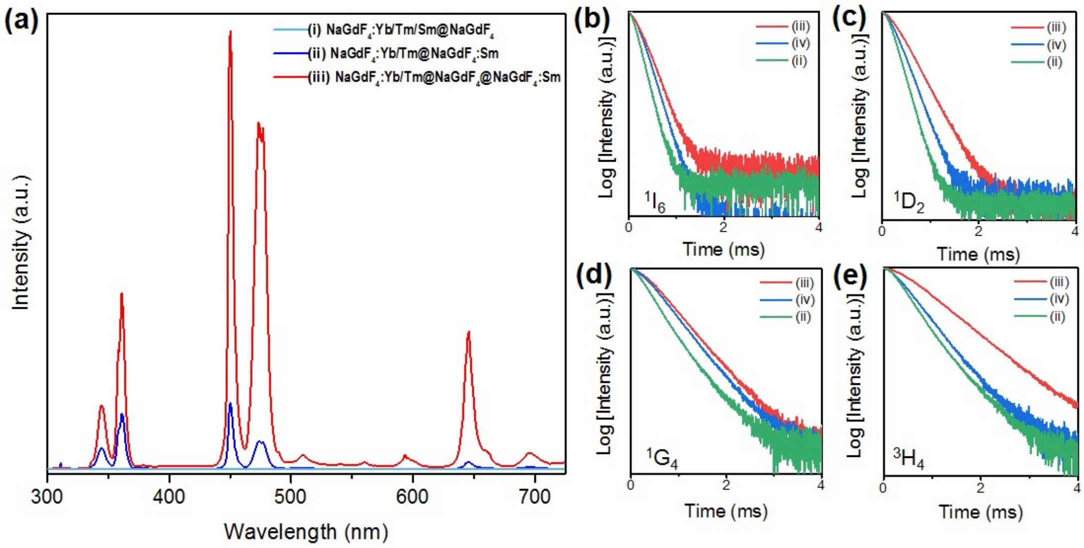

Figure S7. (a) Upconversion emission spectra of (i) $NaGdF_4$:Yb/Tm/Sm@$NaGdF_4$ and (ii) $NaGdF_4$:Yb/Tm@$NaGdF_4$:Sm core-shell and (iii) $NaGdF_4$:Yb/Tm@$NaGdF_4$@$NaGdF_4$:Sm core-shell-shell nanoparticles under 980 nm excitation. (b-e) The decay curves of Tm^{3+} at its $^1I_6$ (at 350 nm), $^1D_2$ (at 362 nm), $^1G_4$ (at 475 nm) and $^3H_4$ (at 803 nm) states for the (iv) $NaGdF_4$:Yb/Tm@$NaGdF_4$ and (ii) $NaGdF_4$:Yb/Tm@$NaGdF_4$:Sm core-shell and (iii) $NaGdF_4$:Yb/Tm@$NaGdF_4$@$NaGdF_4$:Sm core-shell-shell nanoparticles under pulsed 980 nm excitation.