Supporting information

Inverse thermal quenching effect in lanthanide-doped upconversion nanocrystals for anti-counterfeiting

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Figure S1-S14

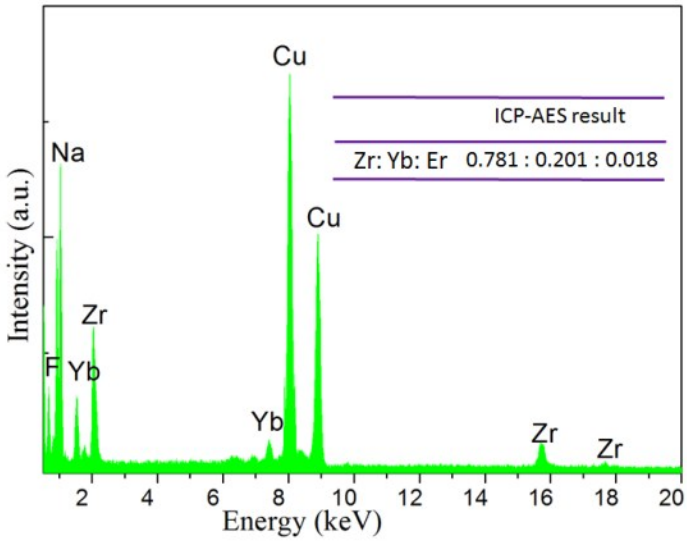

Figure S1 EDS spectra of 20Yb/2Er: Na_3ZrF_7 NCs, Cu signals come from copper grid; Inset is the corresponding ICP-AES measured result.

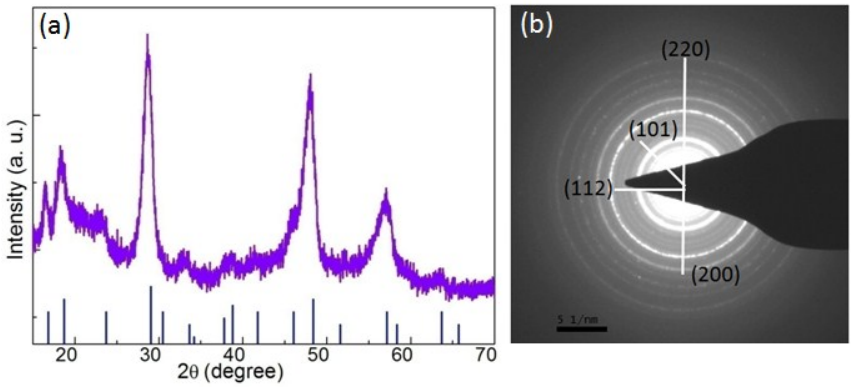

Figure S2 (a) and (b) are XRD pattern and SAED image of 20Yb/2Er: Na_3ZrF_7 NCs, respectively. Bars represent standard tetragonal Na_3ZrF_7 (JCPDS 12-0562) crystal data.

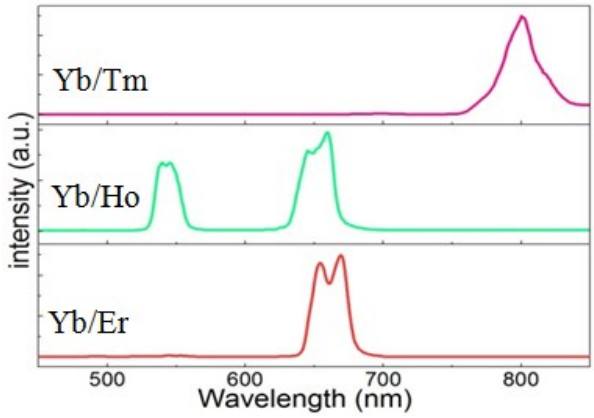

Figure S3 (c) Room-temperature UC emission spectra of 20Yb/2Er (2Ho or 1Tm): Na_3ZrF_7 NCs.

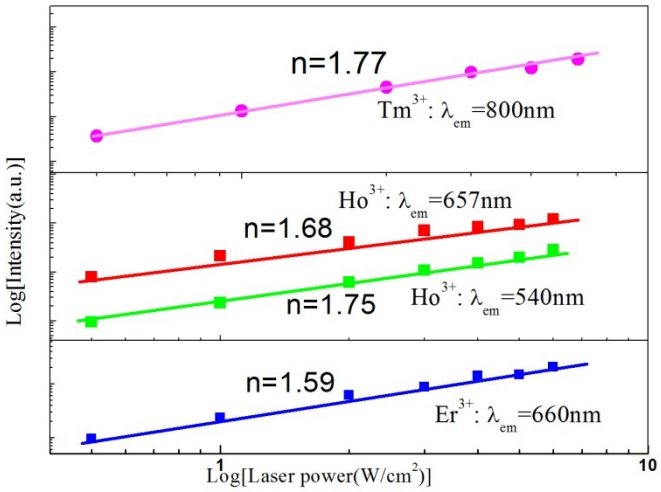

Figure S4 Log-log plots of UC emission intensity versus pumping power for the 20Yb/Ln: Na_3ZrF_7 (Ln = 2Er, 2Ho, 1Tm) NCs.

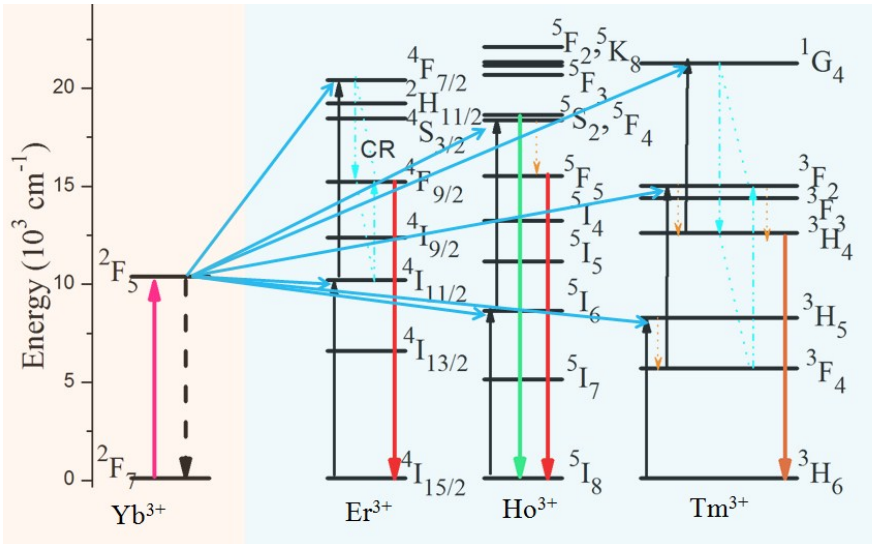

Figure S5 Proposed mechanisms for the energy transfer from Yb^{3+} to Er^{3+}, Ho^{3+} and Tm^{3+}.

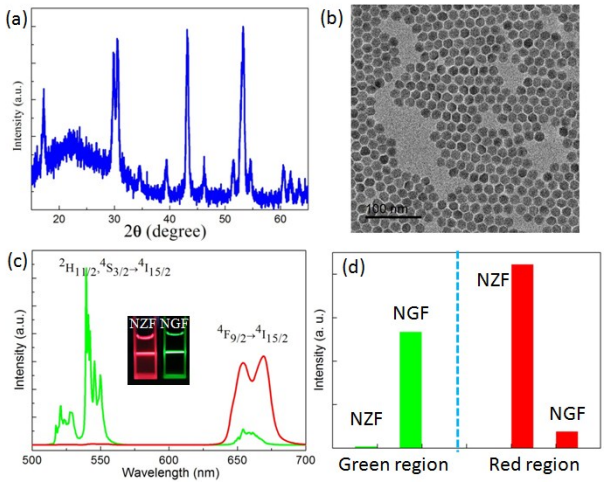

Figure S6 (a) and (b) are the XRD pattern and TEM image of 20Yb/2Er: $NaGdF_4$ NCs (NGF), respectively; (c) UC emission spectra of 20Yb/2Er: $NaGdF_4$ and 20Yb/2Er: Na_3ZrF_7 (NZF) NCs; (d) Histograms of the integral emission intensity of the green and red region for the NGF and NZF samples.

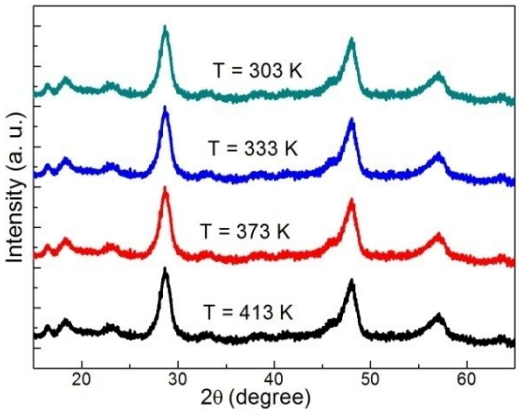

Figure S7 Temperature-dependent XRD patterns of 20Yb2Er: Na_3ZrF_7 NCs.

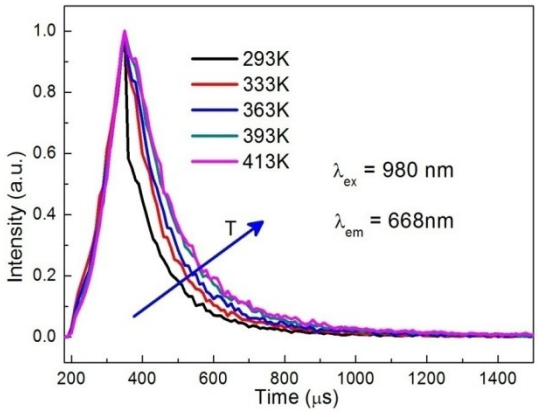

Figure S8 Temperature-dependent lifetimes of $^4F_{9/2}$ excited state of Er^{3+} in 20Yb2Er: Na_3ZrF_7 NCs under 980 nm excitation.

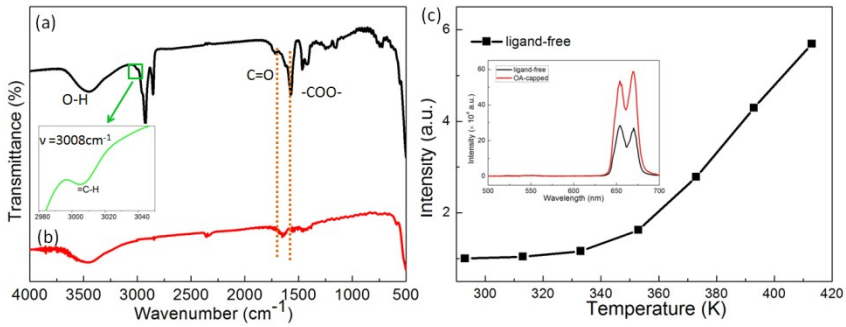

Figure S9 FTIR spectra of the OA-capped (a) and ligand-free (b) 20Yb/2Er: Na_3ZrF_7 NCs; (c) Dependence of the Integral UC emission intensity on temperature for the ligand-free 20Yb/2Er: Na_3ZrF_7 NCs under 976 laser excitation. Insert of (c) are the UC emission spectra of the OA-capped and ligand-free samples under same characterization conditions.

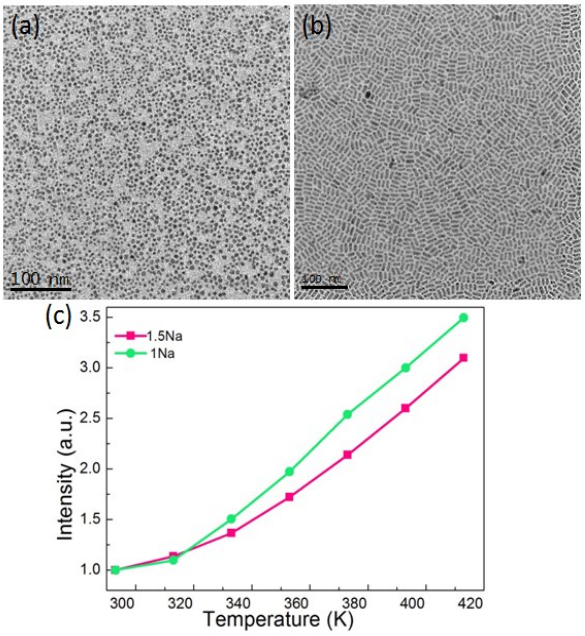

Figure S10 TEM images of 20Yb/2Er: Na_3ZrF_7 NCs prepared with different Na^+ concentration (a: 1mmol and b: 1.5mmol), (c) Dependence of the Integral UC emission intensity on temperature for (a) and (b).

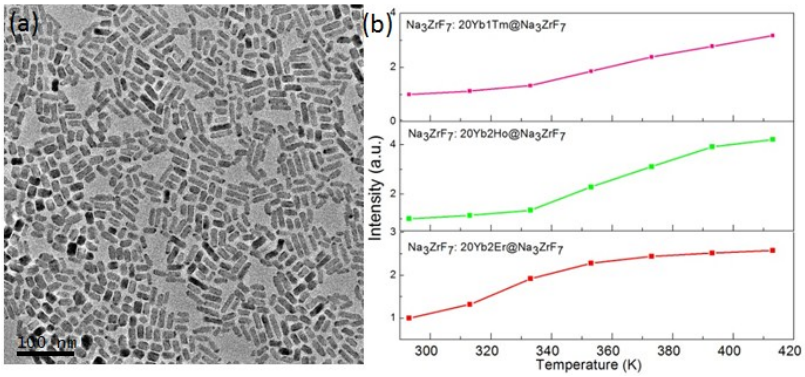

Figure S11 TEM image of 20Yb/2Er: Na_3ZrF_7@Na_3ZrF_7 NCs, (b) Dependence of the Integral UC emission intensity on temperature for 20Yb/2Er: Na_3ZrF_7@Na_3ZrF_7, 20Yb/2Ho: Na_3ZrF_7@Na_3ZrF_7 and 20Yb/1Tm: Na_3ZrF_7@Na_3ZrF_7 NCs.

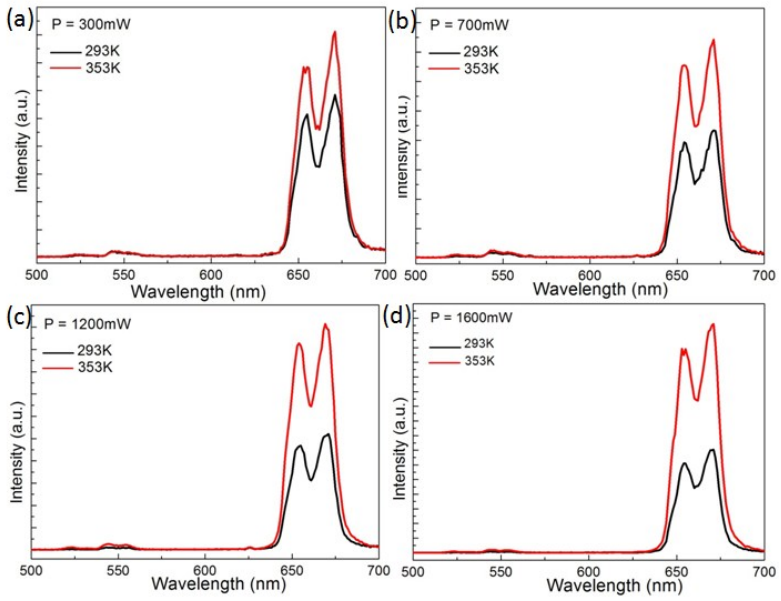

Figure S12 Temperature-dependent photon UC emission spectra of 20Yb/2Er: Na_3ZrF_7 under different pumping power.

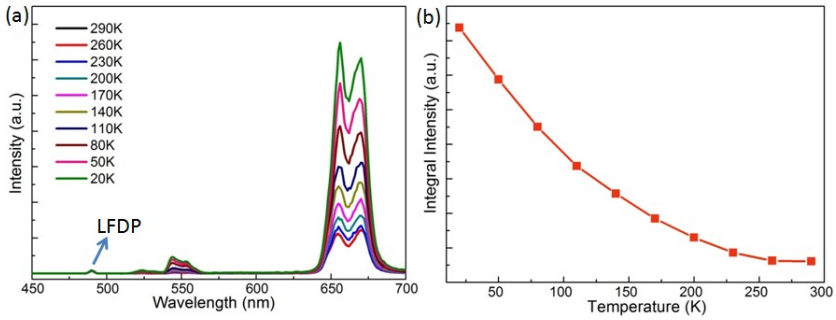

Figure S13 (a) Low-temperature dependent photon UC emission spectra of 20Yb/2Er: Na_3ZrF_7 NCs under 980 nm laser exciation. LFDP represents the laser frequency doubling peak. (b) Dependence of the Integral UC emission intensity on temperature (between 20 and 290 K) for 20Yb/2Er: Na_3ZrF_7 NCs.

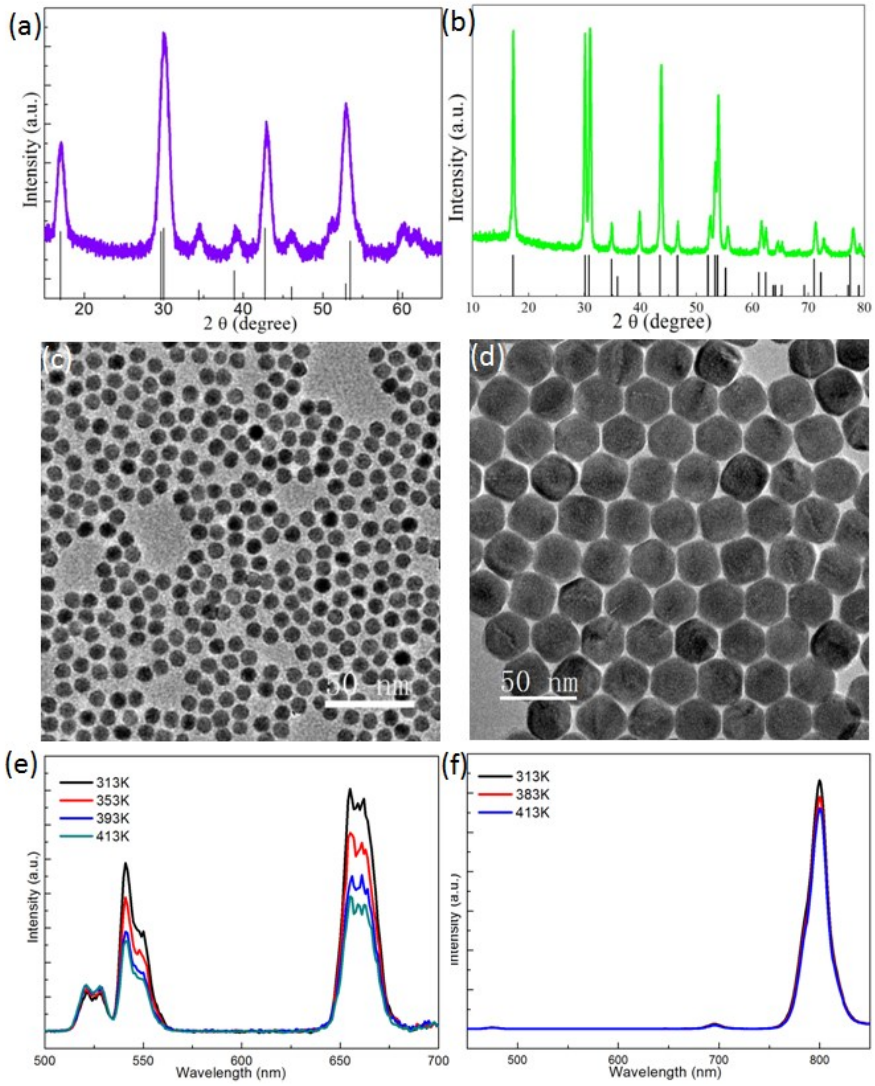

Figure S14 (a) and (b) are the XRD patterns of 20Yb/2Er: $NaGdF_4$ and 49Yb/2Tm: $NaYF_4$ NCs, respectively, bars represent standard hexagonal $NaGdF_4$ (JCPDS 27-0699) and $NaYF_4$ (JCPDS 16-0334) crystal data. (c) and (d) are their corresponding TEM image of (a) and (b), respectively. (e) and (f) are their corresponding temperature dependent UC emission spectra.

References

[S1] Segall, P. J. D. L. M. J. Probert, C. J. Pickard, P. J. Hasnip, S. J. Clark and M. C. Payne, J. Ph ys. Condens. Matter, 2002, 14, 2717.

[S2] J. P. Perdew, K. Burke and M. Ernzerhof, Phys. Rev. Lett., 1996, 77, 3865.

[S3] D. R. Hamann, M. Schlüter and C. Chiang, Phys. Rev. Lett., 1979, 43, 1494.