

## Supporting Information

### **Broadening temperature sensitivity range with core-shell YbEr@YbNd double optical nanothermometer**

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#### ***Characterization***

X-ray powder diffraction (XRD) patterns were recorded with the X'Pert PRO X-ray diffractometer with PIXcel ultrafast line detector, focusing mirror, Soller slits for Cu K $\alpha$  radiation. Transmission electron microscopy (TEM) and selected area electron diffraction (SAED) were performed on the FEI Tecnai G2 20 X-TWIN microscope operating at 200 kV. Vis part of luminescence spectra were collected with Ocean Optics spectrophotometer (model HR4000,  $\delta\lambda=0.3$  nm) while NIR part were collected using Silver-Nova CCD Spectrometers and 808 nm of CW laser diode photo-excitation.

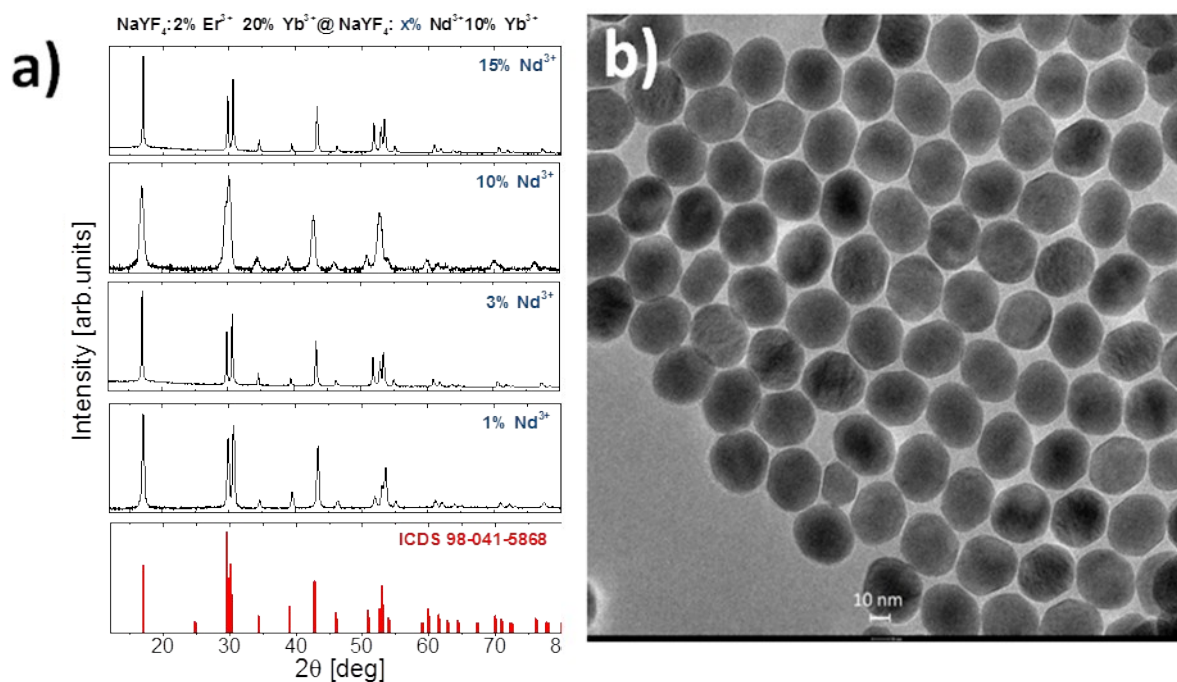


Fig.S1. Structural and morphological characterization of  $\beta$ -NaYF<sub>4</sub>:Yb<sup>3+</sup>, Er<sup>3+</sup>/ $\beta$ -NaYF<sub>4</sub>:Yb<sup>3+</sup>, Nd<sup>3+</sup> nanocrystals: XRD patterns for different Nd<sup>3+</sup> concentration-a; Representative TEM image of the NPs containing 10% of Nd<sup>3+</sup>-b

### ***Spectral characterisation***

The spectral properties have been measured with a home built system. The proper combination of dichroic filters allowed to excite the sample with  $\sim 808$  nm (Spectra-Laser Poland), and record the spectra with two OceanOptics (HR4000) spectrometers sensitive in the Vis-NIR region. One of them was used to record green Er<sup>3+</sup> Upconversion, while the second one was used for Nd<sup>3+</sup> and Yb<sup>3+</sup> emission. The sample (colloidal suspension) was dried at the thin (0.17mm) glass plate and was placed within Linkam TMS94 cryostat, as presented in the Figure S2.

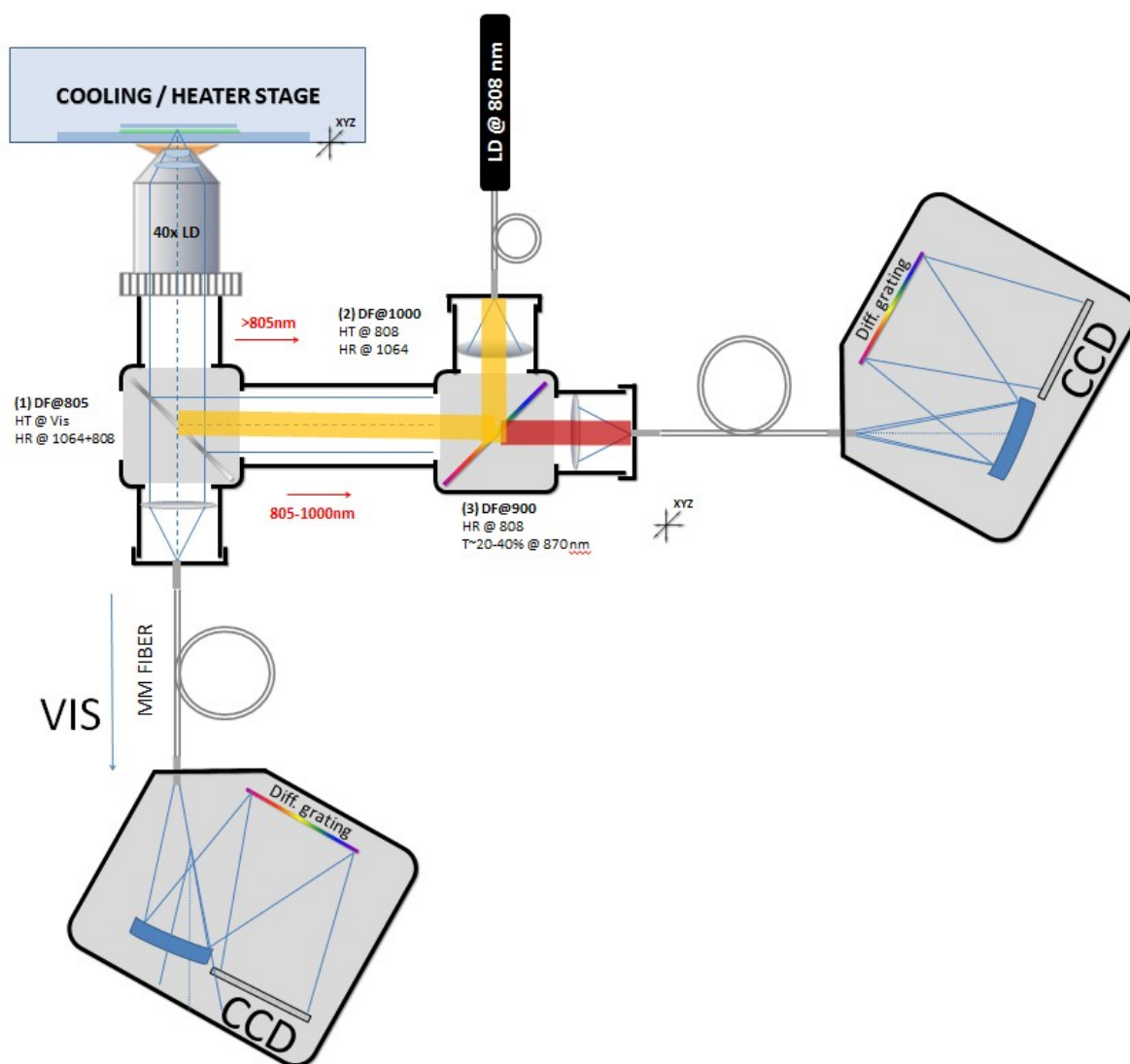


Fig.S2. The measurement setup allowed to excite the samples with 808 and watch both visible and NIR emission with two CCD spectrophotometers.

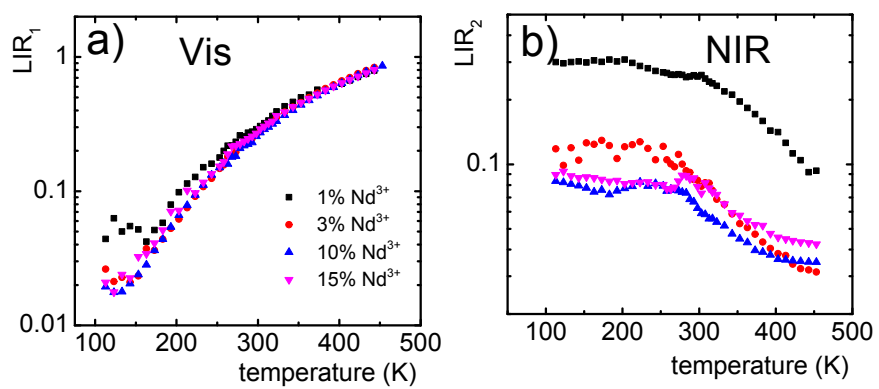


Fig. S3. The temperature dependence of LIR<sub>1</sub> –a; and LIR<sub>2</sub>–b for different Nd<sup>3+</sup> concentration; LIR<sub>1</sub> corresponds to NIR and LIR<sub>2</sub> corresponds to VIS.

As one may find in numerous studies (1-3), the best performance of such nanoscale luminescent thermometers require optimised composition, since a proper balance between respective radiative/non-radiative energy transfers is of key importance here. This is the same in our case, i.e. for higher Nd<sup>3+</sup> content the absorption cross section at 808 nm rises up and the Nd-Yb distance decreases (facilitating Nd→Yb energy transfer), while the parasitic (<sup>4</sup>F<sub>3/2</sub>; <sup>4</sup>I<sub>9/2</sub>)→(<sup>4</sup>I<sub>15/2</sub>; <sup>4</sup>I<sub>15/2</sub>) cross relaxation between neighbour Nd<sup>3+</sup>-Nd<sup>3+</sup> pairs decreases the absolute Nd emission and ultimately Nd→Yb energy transfer, and thus reduces the Yb/Er up-conversion. These properties will of course strongly depend on the dopant composition and host material (owing to available phonons and average distances between active ions in the crystalline matrix). Having 1%, 3%, 10% and 15% Nd<sup>3+</sup> doped samples, we expected to test a relatively versatile range of Nd<sup>3+</sup> dopants, meeting the optimisation requirements described shortly above (Fig.S4). Although, the signal to noise ratio of Yb and Nd emission in NIR improves for rising Nd<sup>3+</sup> concentration, one may note that indeed the <sup>4</sup>F<sub>3/2</sub>→<sup>4</sup>I<sub>11/2</sub> Nd<sup>3+</sup> emission (and thus sensitivity of the thermometer) drops down with rising Nd<sup>3+</sup> concentration. In the same time, the Er<sup>3+</sup> up-conversion stays not affected by Nd<sup>3+</sup> concentration.

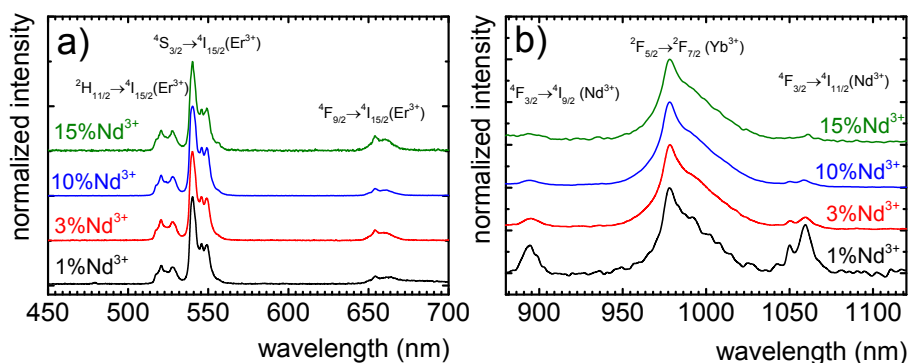


Fig. S4. The comparison of up-conversion –a; and down-shifting –b spectra of β-NaYF<sub>4</sub>:Yb<sup>3+</sup>Er<sup>3+</sup>/β-NaYF<sub>4</sub>:Yb<sup>3+</sup>/Nd<sup>3+</sup> for different Nd<sup>3+</sup> concentration.

1. Rocha, U., et al., *Nd<sup>3+</sup> doped LaF<sub>3</sub> nanoparticles as self-monitored photo-thermal agents*. Applied Physics Letters, 2014. **104**(5): p. 053703.
2. Balabhadra, S., et al., *Boosting the sensitivity of Nd<sup>3+</sup>-based luminescent nanothermometers*. Nanoscale, 2015. **7**(41): p. 17261-17267.
3. Benayas, A., et al., *Nd:YAG Near-Infrared Luminescent Nanothermometers*. Advanced Optical Materials, 2015. **3**(5): p. 687-694.