**Supporting Information**

**Phase and Morphology Selective Interface-Assisted Synthesis of Highly Luminescent Ln$^{3+}$-doped NaGdF$_4$ Nanorods**

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**Experimental Details**

**Synthesis of 1-butyl-3-methylimidazolium chloride ([C$_4$mim][Cl])**

36 mL 1-Methylimidazole (purchased from Sigma. Aldrich) and 70 mL 1-chlorobutane (purchased from Sigma Aldrich) were mixed and then heated under reflux at 60 °C for 72 h. The solvent was evaporated and the resulting liquid was dissolved in dichloromethane and subsequently crystallized from toluene at -50 °C. Afterwards, the white crystalline reaction product was washed in toluene under argon atmosphere and finally dried in vacuum at 60 °C.

**1-Butyl-3-methylimidazolium tetrafluoroborate ([C$_4$mim][BF$_4$])**
A solution of [C₄mim][Cl] (75 g) and sodium tetrafluoroborate (48 g, purchased from Sigma. Aldrich) in 150 mL acetone was reacted at room temperature for 72 h. After removing of the precipitated sodium chloride, the remaining solution was stirred with activated charcoal. Then the solvent was evaporated. The remaining liquid was dissolved in dichloromethane and washed with several amounts of de-ionized water in order to remove any remaining chloride (silver nitrate test). Subsequently, the solvent was boiled off and the obtained colourless liquid was dried in vacuum at 80 °C for 48 h.

**Synthesis of NaGdF₄: Ln³⁺ NCs**

In general, 0.5 mmol sodium oleate and a total amount of 0.5 mmol Ln-trihalide hydrate (Ln=Gd, Yb, Er, Tm, Ce, and Tb) (these chemicals were purchased from ABCR GmbH) were added to 5 mL oleic acid and 15 mL octadecene in a three-neck flask (for thermometer, cooling tube, and separatory funnel). The mixture were firstly heated to 150 °C under argon flow with constant stirring for 90 min to form a clear solution. After addition of 4ML [C₄mim][BF₄], the solution was heated to 300–310 °C under argon flow for 60 min, and then allowed to cool to room temperature. The obtained NCs were precipitated by addition of 15 mL acetone, collected by centrifugation, washed with ethanol several times, and dried at 60 °C for 24 h.

**Characterization.**

X-ray diffraction (XRD) measurements were performed on a Huber G70 diffractometer (Rimsting, Germany) using MoKα radiation (λ = 0.07107 nm). TEM measurements were performed using a
JEOL-2010 TEM equipped with the energy dispersive X-ray spectrometer (EDX). Infrared spectra were recorded on an Alpha FTIR spectrometer (Bruker Optics, in ATR mode with a diamond crystal). The excitation, emission spectra and decay time of all samples were measured on a Fluorolog 3 (Horiba Jobin Yvon) luminescence spectrometer, equipped with steady and pulsed Xe lamps for sample excitation and a photomultiplier for signal detection. Upconversion luminescence spectra were carried out upon 980 nm excitation provided by a mode-locked picosecond Ti:sapphire laser (700-1000 nm, pulse width ≤ 1.5 ps, Tsunami, Spectra-Physics). All photoluminescence studies were carried out at room temperature.