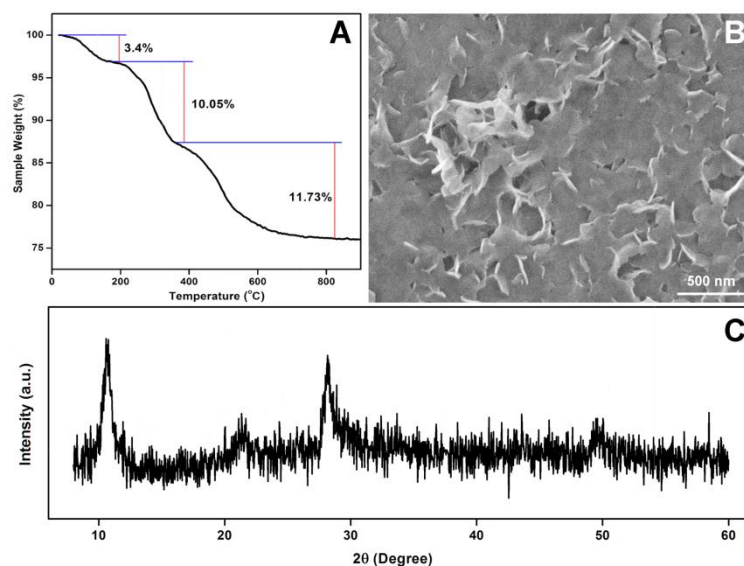


## Two-step synthetic route to GdOF:Ln<sup>3+</sup> Nanocrystals with multicolor Luminescence Properties

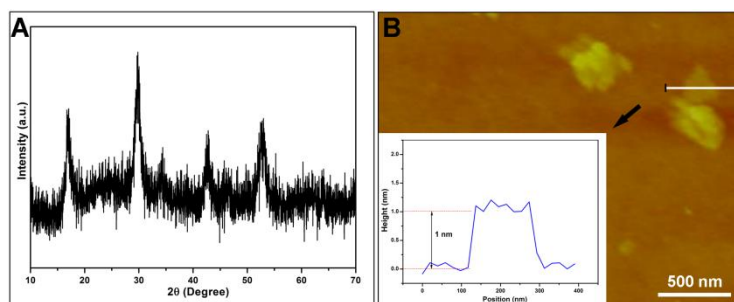
Baiqi Shao,<sup>a,b</sup> Yang Feng,<sup>ab</sup> Mengmeng Jiao,<sup>a,b</sup> Wei Lü,<sup>a</sup> and Hongpeng You<sup>\*a</sup>

State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry, University of Chinese Academy of Sciences, Changchun 130022, and Graduate University of the Chinese Academy of Sciences, Beijing 100049, P. R. China

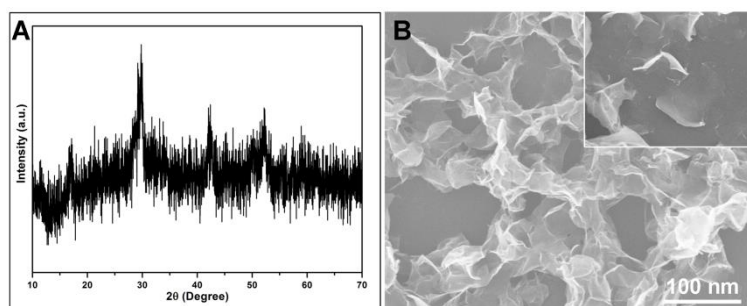


**Figure S1.** TG curve (A), SEM image (B), and XRD pattern (C) of the LGdH precursor.

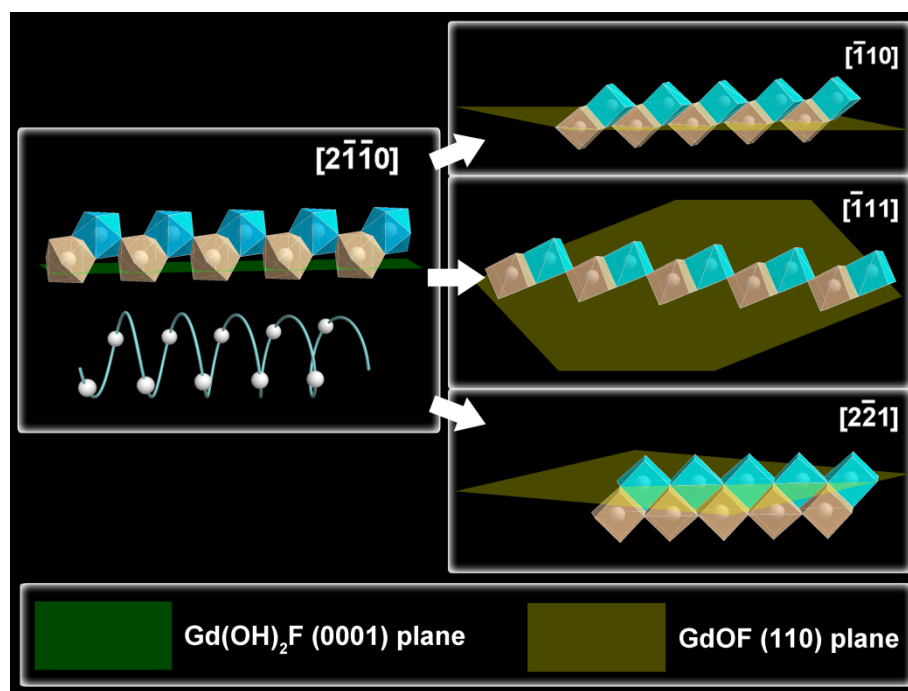
The TG curve shows three distinct mass losses, which is consistent with the results of other layered hydroxide systems. The initial mass loss of 3.4% before 150 °C ascribes to the removal of the intercalated water from the interlayer galleries. The following mass loss of 10.05 % at 290°C corresponds to the partial decomposition of the layers to Gd<sub>2</sub>O<sub>2</sub>(OH)NO<sub>3</sub>. The complete decomposition to Gd<sub>2</sub>O<sub>3</sub> at 490 °C leads to a further mass loss of 11.73%. The observed whole mass loss of 24% is consistent with the calculated mass loss value (24.12%) of



**Figure S2.** The XRD pattern (A) and AFM image (B) of sample with reaction time of 5 min, inset in panel B is the corresponding height profile of the nanosheet.

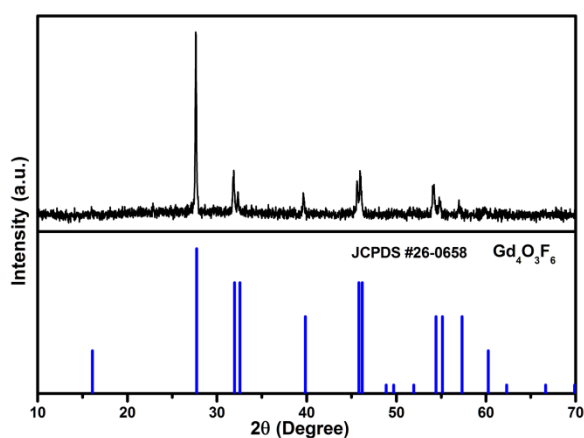


**Figure S3.** The XRD pattern (A) and SEM image (B) of the sample with reaction time of 10 min at ambient temperature.

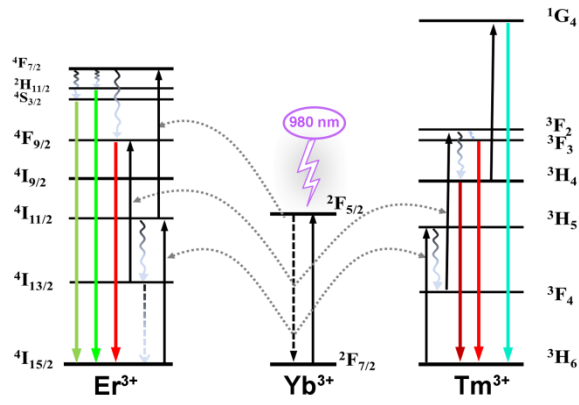


**Figure S4** The detailed structure resemblances between the  $Gd^{3+}$  atomic arrangements in the  $Gd(OH)_2F$  (0001) and  $GdOF$  (110) planes, respectively .

Structurally, the  $Gd^{3+}$  atomic arrangements in the  $Gd(OH)_2F$  (0001) and  $GdOF$  (110) planes are topologically similar. On the one hand, the angles between the  $Gd^{3+}$  atomic arrangements along the three basic crystallographic directions in the  $Gd(OH)_2F$  (0001) plane is approximate to those in the  $GdOF$  (110) plane. In the  $Gd(OH)_2F$  (0001) plane, the angles among the three equivalent directions are  $60^\circ$  ( $\angle[2\bar{1}10]$  &  $[1\bar{1}20]$ ) and  $120^\circ$  ( $\angle[2\bar{1}10]$  &  $[\bar{1}2\bar{1}0]$ ), respectively. Meanwhile, the counterpart angles in the  $GdOF$  (110) plane are  $55^\circ$  ( $\angle[2\bar{2}1]$  &  $[\bar{1}11]$ ) and  $128^\circ$  ( $\angle[2\bar{2}1]$  &  $[\bar{1}10]$ ). On the other hand, the  $Gd^{3+}$  atomic arrangements along the three basic directions in the  $Gd(OH)_2F$  (0001) and  $GdOF$  (110) planes are all analogous screw-axis structures. In the  $Gd(OH)_2F$  (0001) lattice plane, the edge-edge-sharing  $Gd(O/F)_9$  polyhedra constitute zigzag chains with a screw-axis structure along the three equivalent directions. While, the the edge-edge-sharing  $Gd(O/F)_8$  polyhedra show a similar arrangement along the counterpart directions in the  $GdOF$  (110) lattice plane despite that they intuitively exhibit line chains along the  $[\bar{1}11]$  and the  $[\bar{1}10]$  directions when viewed along  $[110]$  direction.



**Figure S5** XRD pattern of the final product when the  $NH_4F$  feeding amount was increased to 2 mmol.



**Figure S5** Schematic energy level diagrams for the UC emissions of  $\text{GdOF}:0.19\text{Yb}^{3+},0.01\text{Er}^{3+}$  and  $\text{GdOF}:0.05\text{Yb}^{3+},0.01\text{Tm}^{3+}$  samples under 980 nm laser.