### Enhanced Photoluminescence of LEuH Nanosheets: 2D Photonic

## Crystals Self-Assembled by Core-Shell SiO<sub>2</sub>@LEuH Spheres

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#### The discussion of the XRD patterns

We could not compare the XRD profile of LEuH with corresponding ICDD card data, because of the ICDD card data for LEuHs were not available at present. We confirmed the LEuH phase by comparing the XRD pattern with those in the literatures which contain the structure information of LEuH, such as "Eur. J. Inorg. Chem., 2009, 2009, 929-936", "Inorg. Chem., 2009, 48, 6724-6730" and "RSC Advances, 2018, 8, 3592-3598".

Song-Ho Byeon et al. reported the synthesis of  $NO_3^-$ -LEuHs for the first time via co-precipitation method (Eur. J. Inorg. Chem., 2009, 2009, 929-936). Then, Fengxia Geng et al. reported the synthesis of  $NO_3^-$ -LEuHs for the first time through homogeneous precipitation of  $Ln(NO_3)_3 \cdot xH_2O$  with hexamethylenetetramine (Inorg. Chem., 2009, 48, 6724-6730). And the above two works both pointed out that  $Ln(NO_3)_3 \cdot xH_2O$  was of monoclinic phase and Geng's work gave the basal spacing of  $NO_3^-$ -LEuHs with 8.25Å. Then, in our previous work (RSC Advances, 2018, 8, 3592-3598), the  $NO_3^-$ -LEuHs was indexed and the basal spacing was of 8.31 Å, which was consistent with the reported. In the present case, the XRD pattern of  $NO_3^-$ -LEuHs was consistent with the results of our previous work, expect for the larger basal spacing of *(001)* planes due to the wet state of the sample.

# The hybridization mechanism between TSPMP and silica microspheres

The hybridization mechanism between TSPMP and silica microspheres is ascribed to the dehydration condensation reaction of the hydroxyls between TSPMP and silica microspheres. The mechanism is as follows:





Figure S1. The zeta-potentials for A) the phosphate-modified silica spheres, B) the nanosheets, and C) the hybridization samples.

The zeta-potentials of the LEuH nanosheets, the phosphate-modified silica spheres and the hybridization sample at the experimental pH conditions have been reinvestigated and measured to be 27.7 mV (pH = 6.11), -55.7 mV (pH = 9.31) and -4.32 mV, (pH = 7.08) respectively, which indicated the positive charge surface of LEuH nanosheets and the negative charged surface of phosphate-modified silica spheres and verified the electrostatic interaction between them.

## The characterization of the nanosheets



Figure S2. AFM images of the nanosheets after ultra-sonication.

The thickness of the LEuH nanosheets was measured by AFM, and the result was shown in Figure S2. According to the crystal structure data of LRHs, its crystallographic thickness can be estimated to be 0.93 nm (L. Hu, R. Ma, T. C. Ozawa and T. Sasaki, Chem. Asian J., 2010, 5, 248-251). The thickness of the nanosheets in this work was measured as 5.48 nm, which indicates that the nanosheets are few-layers.

### The process of self-assembly

Firstly, the substrate was immersed into the breaker vertically, the liquid around the substrate rose to form a liquid membrane and the thickness of the membrane decreased with the increase of the height. As the evaporation of the solvent, the particles transfered from the bulk of the suspension to the membrane constantly; Secondly, under the actions of the capillary force and the electrostatic force, the spheres achieved the thermodynamics steady state and self-assembled into periodic arrays.



Figure S3. SEM images of the arrays of SiO<sub>2</sub>@LEuHs with different concentration, (a) for 0.0002; (b) for 0.002; (c) for 0.004; (d) for 0.006; (e) for 0.008, (f) for 0.01 and (g) for 0.012wt%, respectively.

# The PL lifetime and PL quantum yield of the nanosheets



Figure S4. Luminescence decay curves of the nanosheets, (a) the nanosheets before ultra-sonication, (b) the nanosheets after ultra-sonication.

We measured the PL lifetime and PL quantum yield of the nanosheets before and after ultra-sonication. The Luminescence decay curves of the nanosheets were shown in the Figure S4. The results showed that the PL Lifetime of the nanosheets was slightly elongated after exfoliation, and the phenomenon was consistent with that of our previous work (P. Feng, X. Wang, Y. Zhao, D.-C. Fang and X. Yang, RSC Advances, 2018, 8, 3592-3598), indicating that no direct correlation processed between the PL lifetime and the surface defects of the nanosheets in our study.

The PL quantum yields were also measured for the samples, but the results were shown too small due to the presence of a large amount of hydroxyl groups of LRHs.