Electronic Supplementary Material (ESI) for Chemical Communications.

Supplementary Information

A groundbreaking strategy for fabricating YAG:Ce^{3+} transparent ceramic film *via* sintering LRH nanosheets on sapphire

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Experimental section

Layered rare earth hydroxides of $(Y_{0.99-x}Gd_xCe_{0.01})_2(OH)_5NO_3{\cdot}nH_2O$ were facile synthesized *via* hydrothermal reaction. The LRH crystals were exfoliated into nanosheets by hydrothermal anion exchange followed by mechanical agitation in formamide. Then a nanosheets film was prepared by spin-coating the exfoliated nanohseets on a sapphire substrate using a similar process that described previously [1]. The YAG ceramic films were obtained by calcining the nanosheet film at 1450 °C oxygen gas for 2 h followed by reduction in hydrogen gas at 1200 ^{o}C for 2 h.

Phase identification was performed by X-ray diffraction (XRD, Model Smart Lab, Rigaku, Tokyo, Japan), operating at 40 kV/40mA using nickel-filtered Cu $K\alpha$ radiation and a scanning speed of 6.0° 2θ/min. The product morphology was analyzed by field emission scanning electron microscopy (FE-SEM, Model JSM-7001F, JEOL, Tokyo) and transmission electron microscopy (TEM, Model JEM-2000FX, JEOL, Tokyo). A Nanosurf easyScan 2 atomic force microscope (AFM, Switzerland) was employed to obtain topographical images of the nanosheets. Fourier transform infrared spectroscopy experiments (FT-IR, Nicolet iS5, Thermal Fisher Scientific, USA) were under taken using the standard KBr method. The photoluminescence of the samples were analyzed with an FP-8600 fluorospectrophotometer (Jasco,Tokyo).

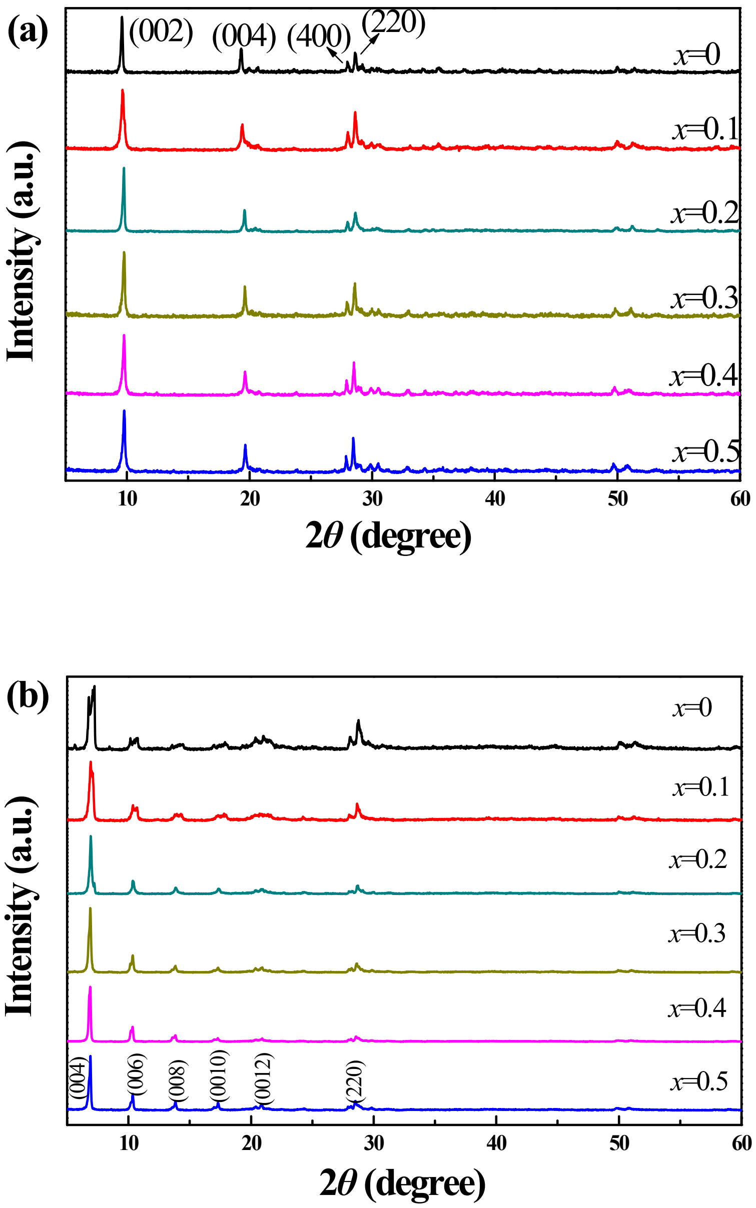

Figure S1. XRD patterns of (a) $(Y_{0.99-x}Gd_xCe_{0.01})_2(OH)_5NO_3\cdot nH_2O$ (LRH) and (b) their intercalated products LRH-DS.

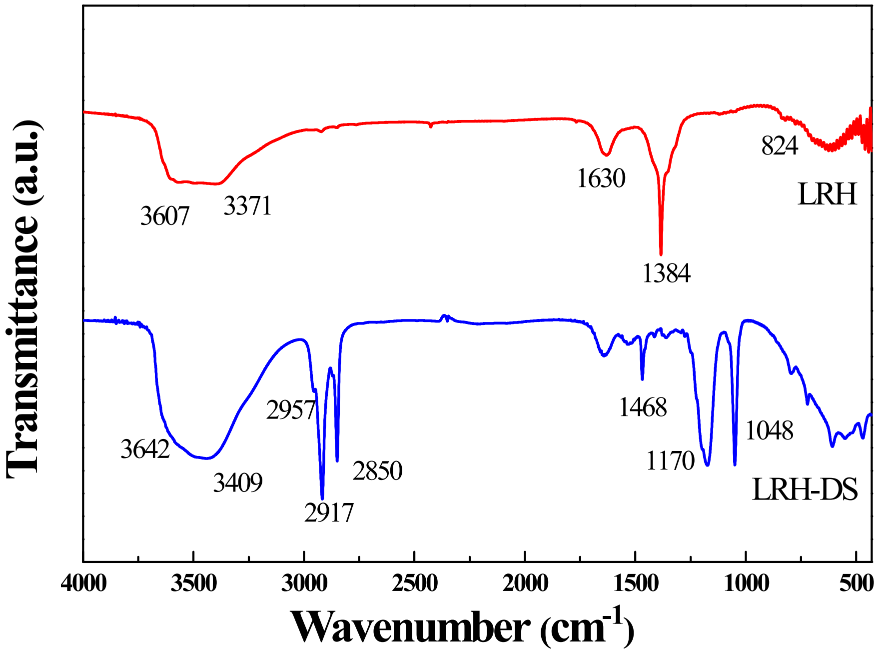

Figure S2. FT-IR spectra of LRH and LRH-DS samples. After anion exchange, the vibration of nitrate anion at ~1384 cm^{-1} is no longer observed. Instead, two intense bands at around ~1170 and ~1048 cm^{-1} are assigned to the stretching mode of the sulfate (OSO_3^-) [2,3]. The strong absorption peaks at ~2917 and ~2850 cm^{-1} are due to the asymmetric and symmetric CH_2 stretching vibrations respectively, in the alkyl chain of dodecylsulfate, whereas the weak band at ~2957 cm^{-1} is assigned to the stretching mode of the terminal CH_3 group of the hydrocarbon tail [2,3]. The above results confirmed a complete replacement of nitrate anion by the DS$^-$ units in the gallery of LRH.

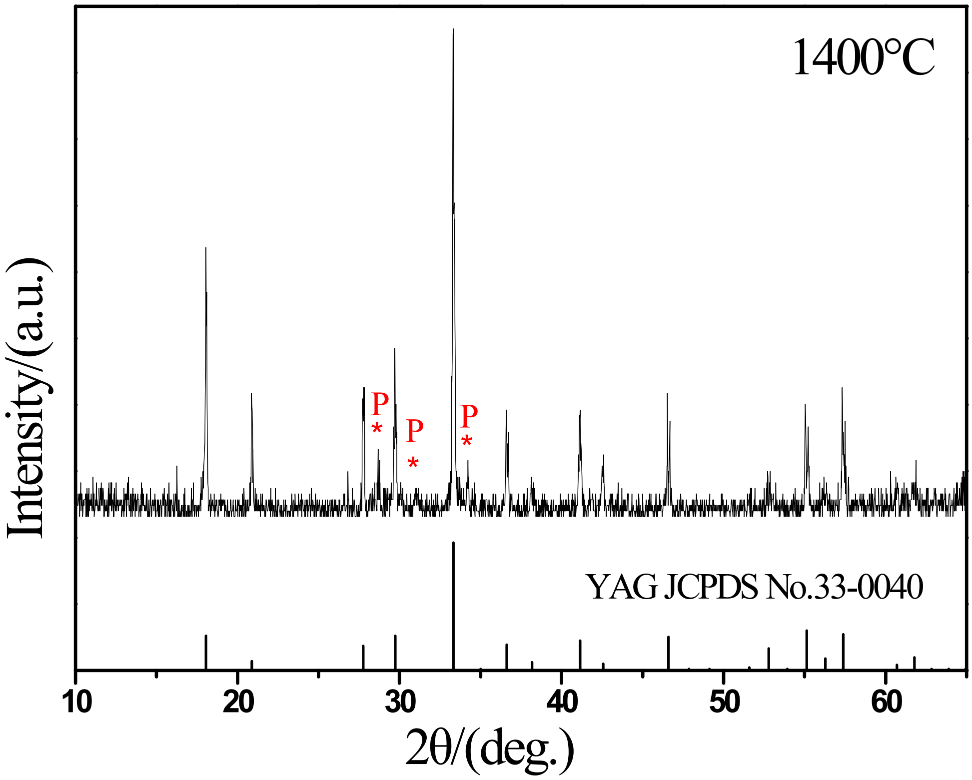

Figure S3. XRD patterns of the ceramic film (x=0.5 sample) calcined at 1400 ^{o}C. The diffraction peaks marked by asterisk are the perovskite impurity (YAP).

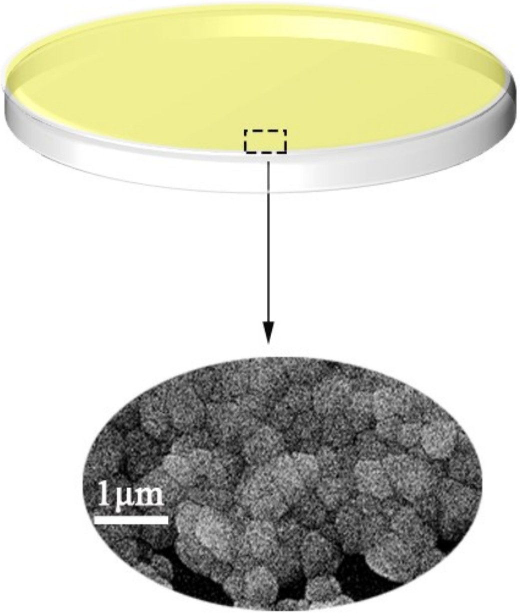

Figure S4. FE-SEM micrograph of ($Y_{0.49}Gd_{0.50}Ce_{0.01}$)AG ceramic film at the edge, indicating that the film is composed of one layer of grains with the thickness around 0.5 μm.

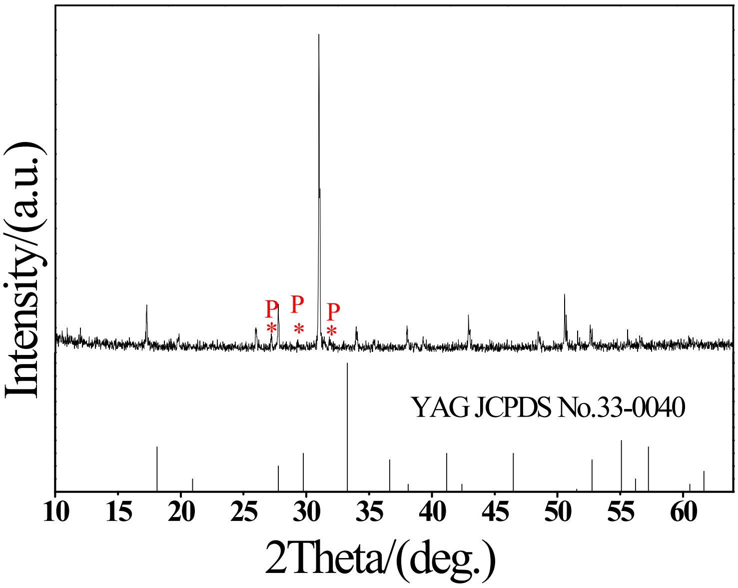

Figure S5. XRD patterns of the film calcined at 1450 °C from the LRH nanosheets film (*x*=0.5 sample) prepared by twice spin-coating processing. The diffraction peaks marked by asterisk are the perovskite impurity (YAP). This indicates that calcining thicker LRH nanosheets film on sapphire substrate cannot yield YAG film in pure phase.

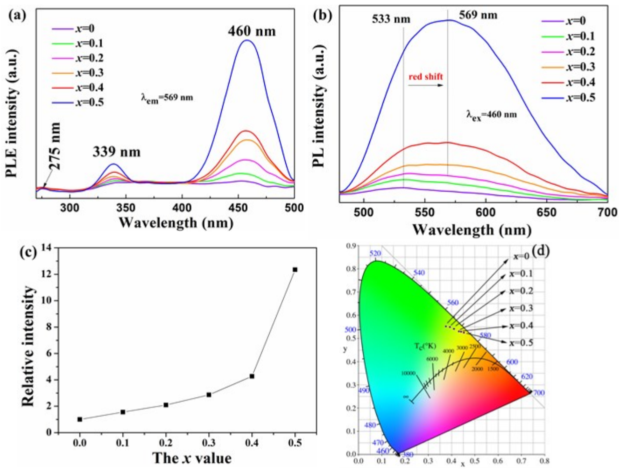

Figure S6. (a and b) PLE/PL spectra, (c) relative integral intensity and (d) CIE chromaticity coordinates of ($Y_{0.99-x}Gd_xCe_{0.01}$)AG ceramic films.

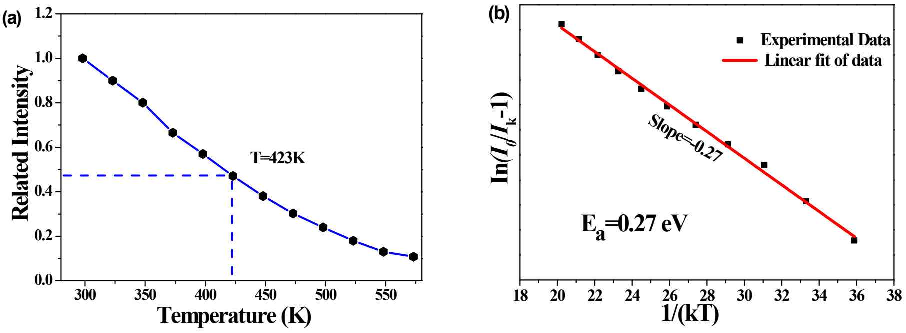

Figure S7. (a) Relative emission intensity of PL spectra at the temperatures ranging from 298 to 573 K, and (b) activation energy of thermal quenching for emission bands.

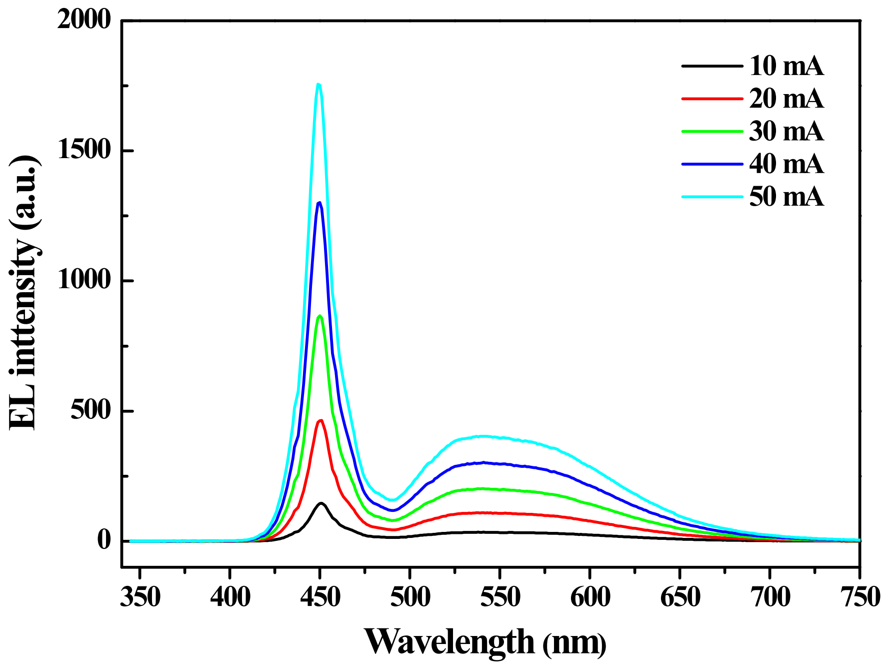

Figure S8. Electroluminescence spectra of YAG:Ce^{3+} ceramic film driven by a blue-LED chip with the wavelength of 460 nm, under a voltage of 2.0 V.

References:

[1] Q. Zhu, J.-G. Li, X. D. Li, X. D. Sun, Y. Qi, M. Y. Zhu, Y. Sakka, *Sci. Technol. Adv. Mater.*, 2014, **15**, 014203.

[2] K. Nakamoto, *Infrared Spectra of Inorganic and Coordination Compounds*, John Wiley & Sons, New York, 1963.

[3] J. A. Gadsden, *Infrared Spectra of Minerals and Related Inorganic compounds*, Butterworths, London, 1975.