Supplementary Information (SI) for Inorganic Chemistry Frontiers.

Gd/Tb/Eu alloyed double-perovskite lanthanide halide for color tunable photoluminescence and robust scintillation performance

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EXPERIMENTAL SECTION

Chemicals and Materials: CsCl (99.99%), NaCl (99.99%), $EuCl_3$ (99.99%), $GdCl_3$ (99.99%), $TbCl_3$ (99.99%), (3-Aminopropyl) triethoxysilane (APTES) (> 95%), Ethyl acetate (AR, ≥99.5%) were purchased from Aladdin. Ethanol (≥99.7%) was purchased from Huzhou Shuanglin chemical Technology Co., Ltd. All the chemical reagents were used as received without further purification.

Synthesis of submicron $Cs_2NaLnCl_6$ (Ln=Gd, Tb, Eu): Take $Cs_2NaGdCl_6$ as a typical example. 0.2 mmol CsCl, 0.1 mmol NaCl, 0.1 mmol $GdCl_3$ were dissolved in 500 μL DI-water to obtain the salt solvent as precursor. The anti-solvent was achieved by mixing 10 mL ethyl acetate and 10 mL absolute ethyl alcohol dissolving 100 μL APTES. Then, the precursor was added into the anti-solvent drop by drop under vigorous tiring. Suddenly, the white powers were separated out from solvent and further be collected by centrifugation. Prepared were drying in oven at 60°C for three hours. For Tb^{3+} and Eu^{3+} ions incorporation, given stoichiometric ions are utilized to replace Gd^{3+} ions.

Characterizations: X-ray diffraction (XRD) analysis was carried out by a powder diffractometer (Bruker D8 Advance) with a Cu-Kα (λ = 1.5405 Å) radiation. TESCAN VEGA3 S-4300 field-emission electron microscope was executed by a Scanning electron microscope (SEM), with electron energy of 30 keV. PL spectra were recorded on an Edinburgh Instruments (EI) FLS1000 spectrofluorometer equipped with a continuous (450 W) xenon lamp. PLQYs ere measured by the spectrofluorometer (FS1000) equipped with an integrating sphere. XEOL spectra were measured by a spectrometer (OmniFluo960) with an X-ray tube (target material: W, voltage 50 kV, tube current 200 μA) and a photon counter (DCS210PC-9S). X-ray imaging was performed using a home-made setup comprising a miniature X-ray tube and camera (ORCA-Fusion BT, C15440-20UP).

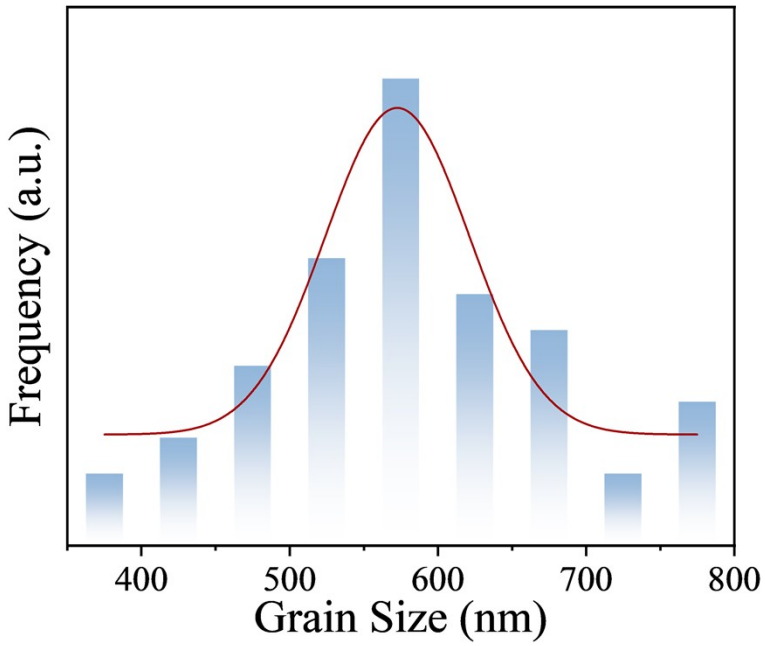

Fig. S1. Statistical histogram of grain size of $Cs_2NaTbCl_6$.

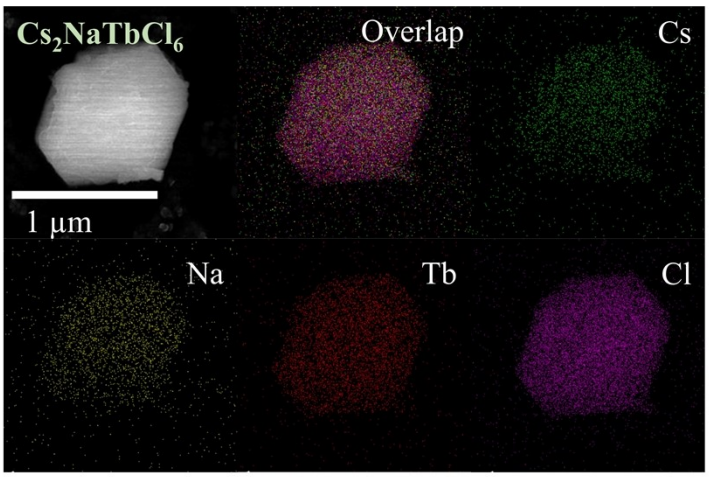

Fig. S2. EDS elemental mapping on $Cs_2NaTbCl_6$.

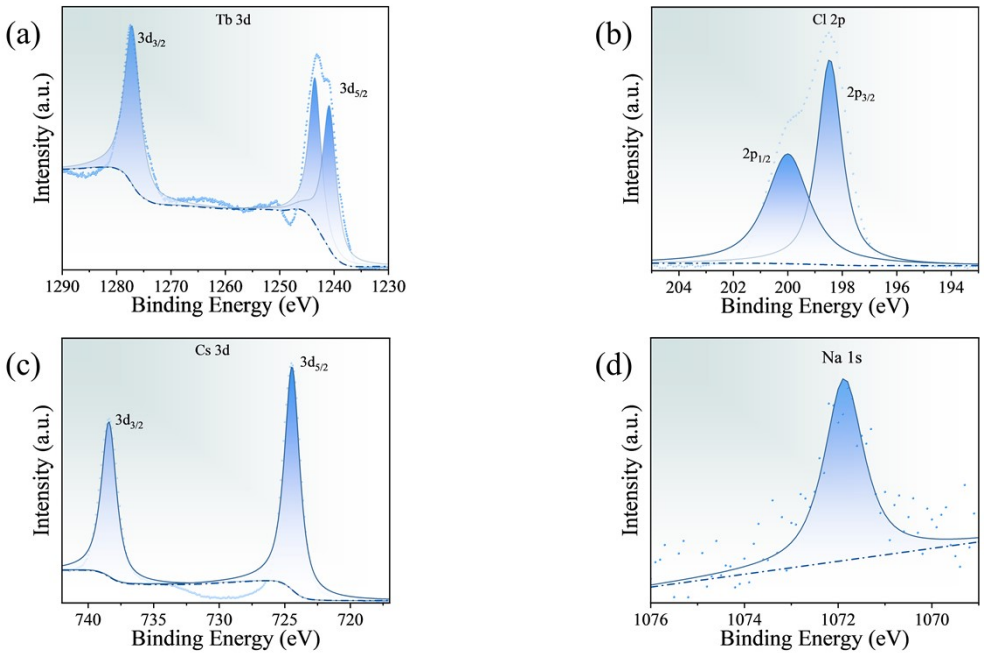

Fig. S3. High-resolution XPS spectra of Tb (a), Cl (b), Cs (c) and Na (d).

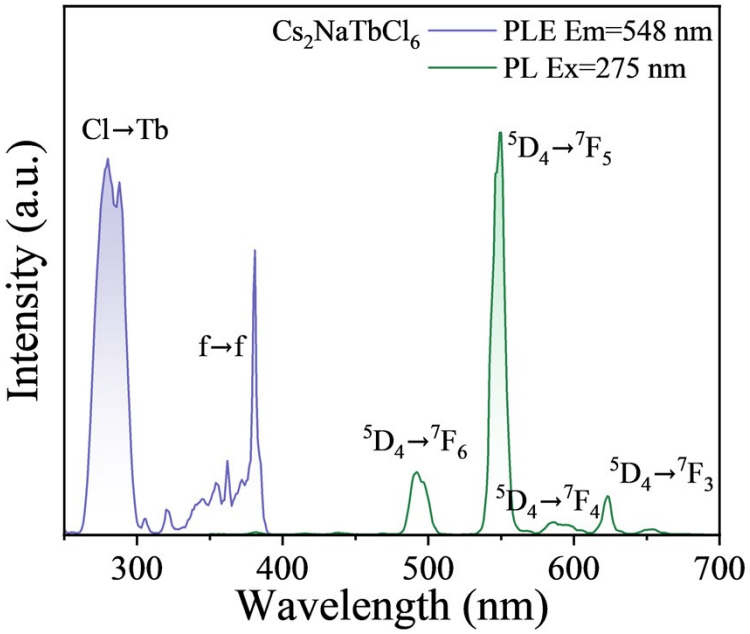

Fig. S4. PLE (Em=548 nm) and PL spectra (Ex=275 nm) of $Cs_2NaTbCl_6$.

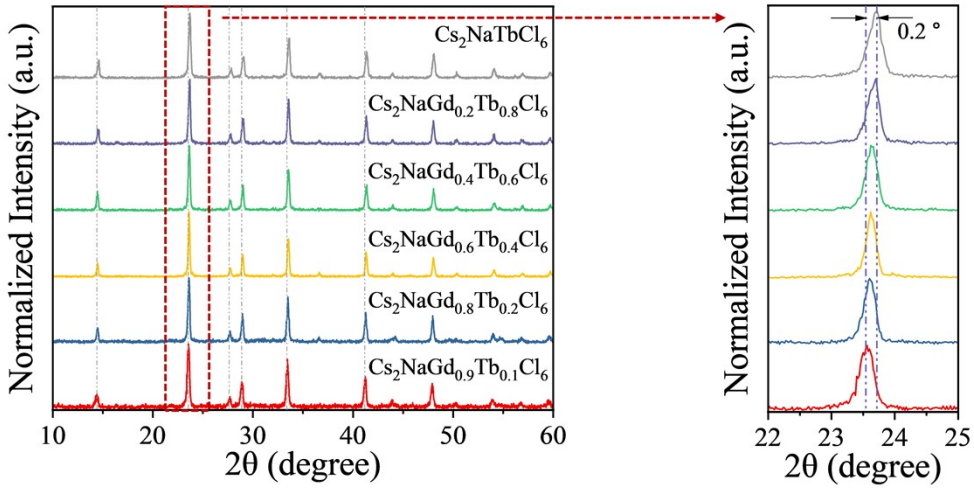

Fig. S5. XRD patterns of a series of Cs2NaGd1-xTbxCl6, and high-resolution spectra of (2 2 0).

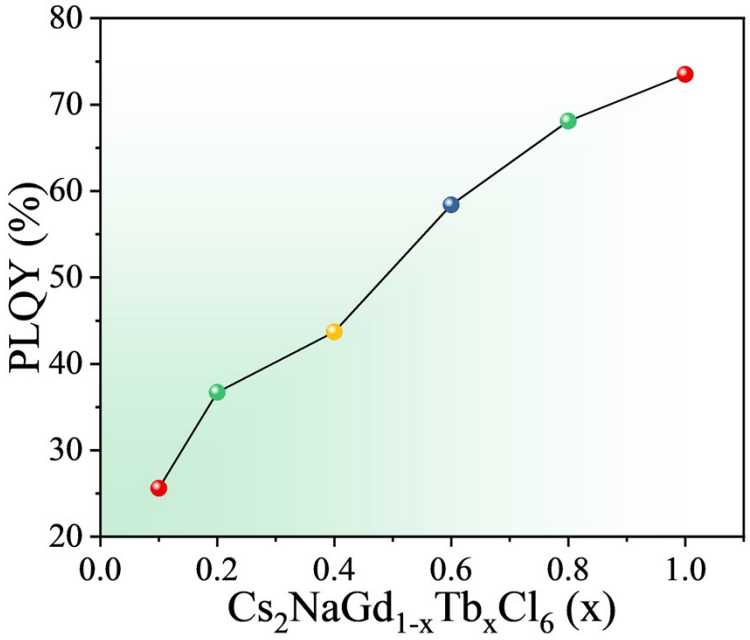

Fig. S6. PLQY of $Cs_2NaGd_{1-x}Tb_xCl_6$ with various Tb concentration (Ex=275 nm).

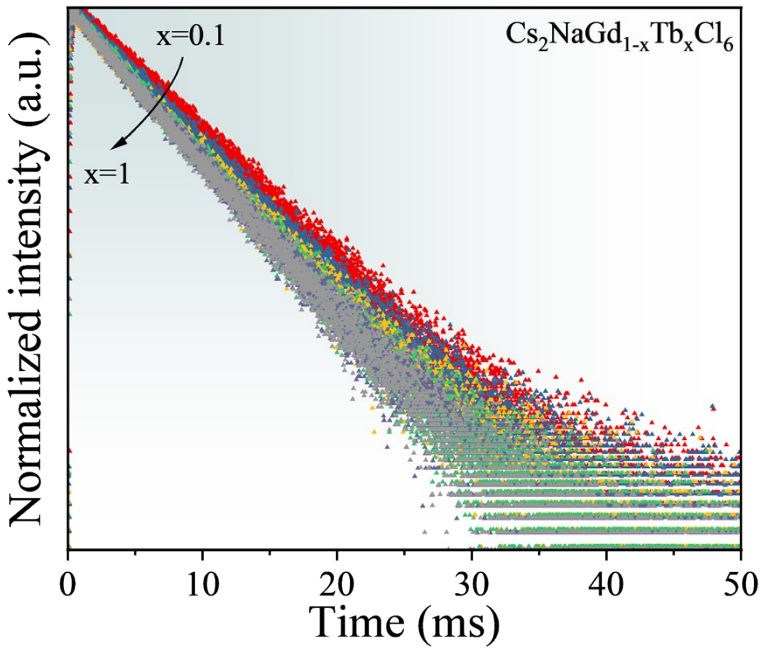

Fig. S7 Photoluminescence decay curves for the $^5D_4$ level (548 nm).

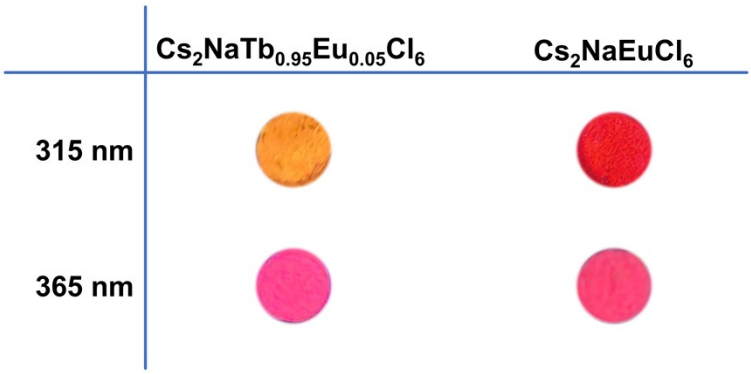

Fig. S8. Luminescence images of $Cs_2NaTb_{0.95}Eu_{0.05}Cl_6$ and $Cs_2NaEuCl_6$ under 315 nm and 365 nm excitation.

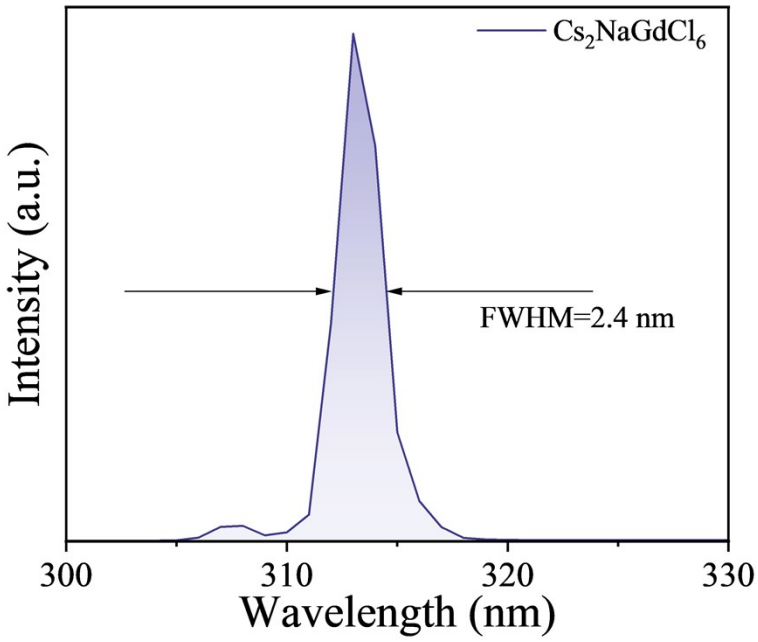

Fig. S9. XEOL spectrum of $Cs_2NaGdCl_6$ showing narrow band emission peak.

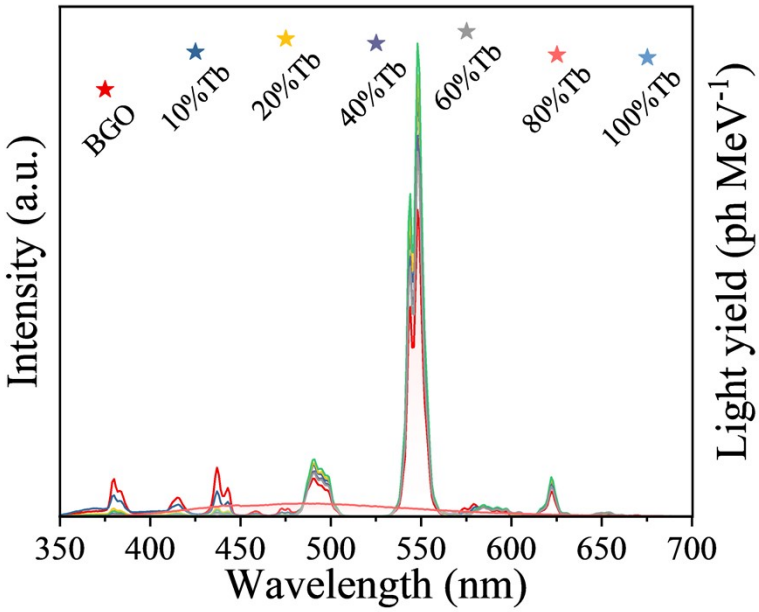

Fig. S10. Quantitative XEOL spectra and corresponding LY with BGO as reference.

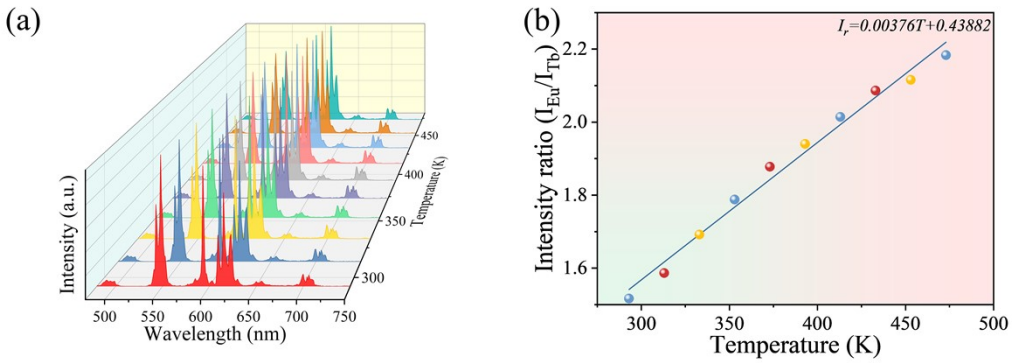

Fig. S11. (a) Temperature dependent XEOL spectra of $Cs_2NaGd_{0.35}Tb_{0.6}Eu_{0.}05Cl_6$. (b) intensity ratio between the Eu and Tb under various temperature.

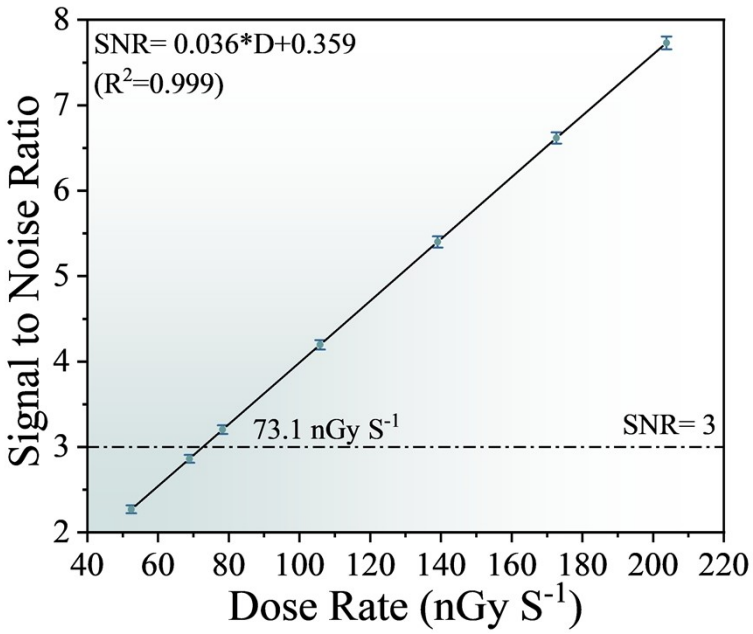

Fig. 12. SNR values of $Cs_2NaGd_{0.35}Tb_{0.6}Eu_{0.05}Cl_6$@PMMA films as a function of dose ratio.

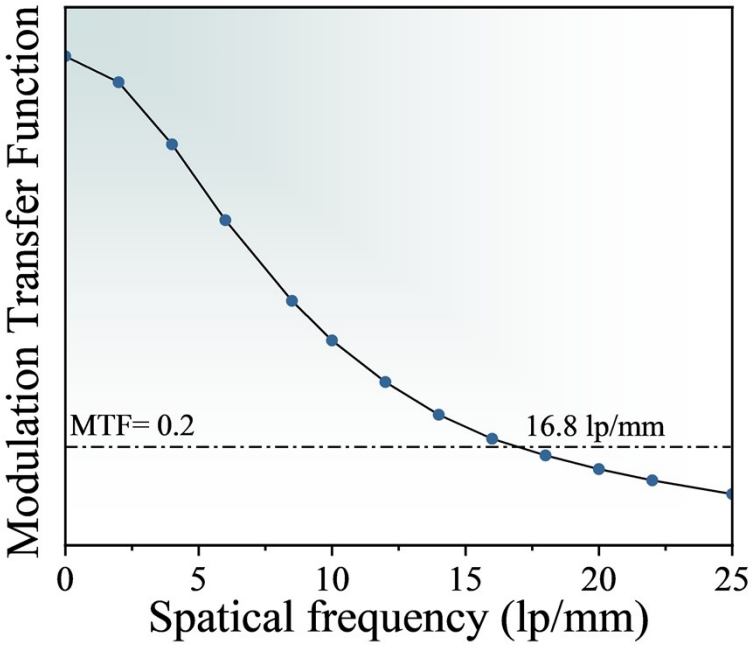

Fig. 13. The MTF plot of $Cs_2NaGd_{0.35}Tb_{0.6}Eu_{0.05}Cl_6$@PMMA film.

Table S1: The lifetime of 437 nm and 548 nm of $Cs_2NaGd_{1-x}Tb_xCl_6$.

Ex=275 nm	Em=437 nm ($^5D_3 \rightarrow {}^7F_4$)	Em=548 nm ($^5D_4 \rightarrow {}^7F_5$)
$CsNaGd_{0.9}Tb_{0.1}Cl_6$	2.52 ms	7.16 ms
$CsNaGd_{0.8}Tb_{0.2}Cl_6$	1.42 ms	6.63 ms
$CsNaGd_{0.6}Tb_{0.4}Cl_6$	106.6 μs	6.29 ms
$CsNaGd_{0.4}Tb_{0.6}Cl_6$	40.3 μs	6.02 ms
$CsNaGd_{0.2}Tb_{0.8}Cl_6$	27.0 μs	5.98 ms
$CsNaTbCl_6$	19.6 μs	5.92 ms