## Electronic Supplementary Information

## Broad bandwidth emission and in situ electric field modulation of photoluminescence in Nd-doped ferroelectric crystals

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## **Experimental Section:**

The Nd-doped ferroelectric crystals ( $Pb_{0.97}Nd_{0.02}$ )[( $Mg_{1/3}Nb_{2/3}$ )<sub>0.61</sub>Ti<sub>0.39</sub>]O<sub>3</sub> [Nd:PMNT] with tetragonal perovskite structure were grown by top-seeded solution-growth method.<sup>[1]</sup> The plates were sliced along the <001> direction for electrical, photoluminescence performance measurements. The transparent indium tin oxide (ITO) was deposited on both sides of samples using magnetron sputtering system for photoluminescence response measurement. The polarization-electric field (*P-E*) hysteresis loop and current vs electric field (*I-V*) curve were measured using a ferroelectric analyzer (TF2000, aix-ACCT, Germany). The piezoresponse images before and after poling through an explanted direct current power source were observed by a piezoresponse force microscopy (PFM, Cypher ES, Asylum Research, USA) using conductive proves (ASYELEC-01, Asylum Research, USA) at room temperature. A UV/V/NIR fluorescence spectrometer (FLS980, Edinburgh Instruments, England) was used to measure photoluminescence (PL) response.



**Fig. S1** The ferroelectric polarization-electric field (*P-E*) hysteresis loop and the current-electric field (*I-E*) curve of Nd:PMNT single crystal. The values of the remanent polarization (*P<sub>r</sub>*) and coercive field (*E*<sub>C</sub>) were 36.7  $\mu$ C/cm<sup>2</sup> and 4.91 kV/cm, respectively.



Fig. S2 Schematic diagram of the interaction between  $Nd^{3+}$  ions and B-site ions moving from a B<sub>1</sub> position to a B<sub>2</sub> position, when the 90° domains switching induced by an external electric field.



**Fig. S3** The PFM phase image of a Nd:PMNT crystal under (a) zero field (10  $\mu$ m×10  $\mu$ m) and (b) a coercive field along the *z* direction (5  $\mu$ m×5  $\mu$ m). The schematic diagram is the distribution of the 180° domain (c) in the virgin sample and (d) under the coercive electric field along the *z* direction.



**Fig. S4** Transmittance of Nd:PMNT crystals under the electric field of 0 kV/cm,  $E_{\rm C}$  and  $2E_{\rm C}$ . The transmittance was increased by about 6.4% from 0 kV/cm to  $E_{\rm C}$ , and by about 0.6% from  $E_{\rm C}$  to  $2E_{\rm C}$ .

## Reference

[1] X. Long, Z.-G. Ye, Acta Mater., 2007, **55**, 6507-6512.