## Spectral characteristics of Nd<sup>3+</sup>/Yb<sup>3+</sup>:YPO<sub>4</sub> single crystal with strong multi-wavelength emission

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† Electronic supplementary information (ESI) available: The reaction formula, calculation of emission cross-sections,  $I_{sat}$ ,  $\beta_{min}$  and  $I_{min}$ . See DOI: 10.1039/x0xx00000x

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Eqn. S1 PbHPO<sub>4</sub> decomposes chemical reaction equation. Eqn. S2 Nd<sub>x</sub>:Yb<sub>y</sub>:Y<sub>(1-x-y)</sub>PO<sub>4</sub> synthesis chemical reaction equation. Calculations of emission cross-sections. (Equations S3-S5) Calculations of  $I_{sat}$ ,  $\beta_{min}$  and  $I_{min}$ . (Equations S6-S8) Eqn. S1 PbHPO<sub>4</sub> decomposes chemical reaction equations.

$$2PbHPO_4 = Pb_2P_2O_7 + H_2O \qquad (S1)$$

**Eqn. S2**  $Nd_x: Yb_y: Y_{(1-x-y)}PO_4$  synthesis chemical reaction equation.

$$Pb_{2}P_{2}O_{7} + (1-x-y)Y_{2}O_{3} + xNd_{2}O_{3} + yYb_{2}O_{3} = 2Nd_{x}:Yb_{y}:Y_{(1-x-y)}PO_{4} + 2PbO$$
 (S2)  
Where x=0.8 at.%, y=2.0 at.%.

## Calculations of emission cross-sections.

We calculated the emission cross section of  $Nd^{3+}$  and  $Yb^{3+}$  ions by RM method formula (**S3**) and F-L method formula (**S5**), respectively.

$$\sigma_{em}^{i}(\lambda) = \frac{3\sigma_{abs}^{i}\exp(-hc/kT\lambda)}{8\pi n^{2}\tau_{rad}c\sum_{j}\lambda^{-4}\sigma_{abs}^{j}(\lambda)\exp(-hc/(kT\lambda))d\lambda}$$
(83)

*h* is the Planck constant, *c* is the speed of light in vacuum, *k* is the Boltzmann constant, *T* is absolute temperature, *n* is the refractive index,  $\tau_{rad}$  is the radiation lifetime, which can be calculated by the following formula.

$$\tau_{\rm rad} = \frac{3}{8\pi n^2 c} \frac{Z_l}{Z_u} \frac{\exp(-hc/(k_B T\lambda))}{\sum_j \lambda^{-4} \sigma_{abs}^j(\lambda) \exp(-hc/(k_B T\lambda)) d\lambda}$$
(84)

 $Z_u$  is the high energy level group,  $Z_l$  is the low energy level group, where  $g_k$  is the degeneracy of each sub level,  $k_B$  is Boltzmann constant.

$$\sigma_{em}^{F-L}(\lambda) = \frac{\lambda^5}{8\pi n^2 c} \frac{1}{\tau_{rad}} \frac{I(\lambda)}{\int \lambda I(\lambda) d\lambda}$$
(85)

 $I(\lambda)$  represents the optical density.

## Calculations of $I_{sat}$ , $\beta_{min}$ and $I_{min}$ .

 $I_{sat}$  represents the pump saturation intensity, which is a measure of the ease with which the Yb<sup>3+</sup> ions population must be bleached to overcome the ground state absorption, is given by the equation:

$$I_{sat} = hc/[\lambda_p \sigma_{abs}(\lambda_p)\tau_f]$$
 (S6)

Where  $\lambda_p$  is the pump wavelength, *h* is the Planck constant,  $\sigma_{abs}(\lambda_p)$  is absorption cross-sections at the pump wavelength  $\lambda_p$ .

The minimum fraction of Yb<sup>3+</sup> ions that must be excited such that the ground state absorption and the gain exactly balance, and there is net transparency at  $\lambda_{ext}$ . And the equation can be expressed by

$$\beta_{min} = \sigma_{abs}(\lambda_{ext}) / [\sigma_{abs}(\lambda_{ext}) + \sigma_{ext}(\lambda_{ext})]$$
(S7)

 $I_{min}$  represents the minimum absorbed pump intensity required to reach the threshold. It can be obtained from the following equation:

$$I_{min} = \beta_{min} \times I_{sat}$$
 (S8)