

## **Supporting Information**

### **Structure modification and luminescence regulation in new violet light excitable $\text{Sr}_{(2-y)}\text{Ba}_y\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{xEu}^{2+}$ phosphors via cation substitution**

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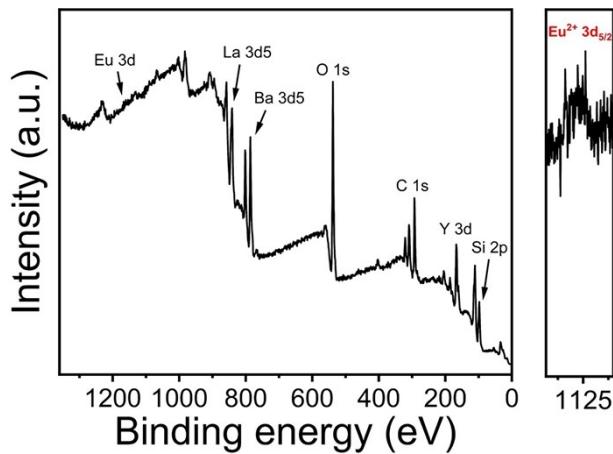
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**Table S1.** Crystallographic data and refined parameters for the  $\text{Sr}_{(2-y)}\text{Ba}_y\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{Eu}^{2+}$  ( $0.5 \leq y \leq 2$ ) samples.

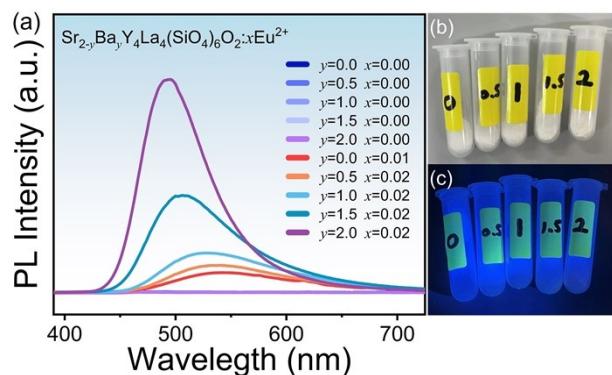
| Sample      | $\text{Sr}_2$ | $\text{Sr}_{1.5}\text{Ba}_{0.5}$ | $\text{SrBa}$ | $\text{Sr}_{0.5}\text{Ba}_{1.5}$ | $\text{Ba}_2$ |
|-------------|---------------|----------------------------------|---------------|----------------------------------|---------------|
| Space group | $P6_3/m$      | $P6_3/m$                         | $P6_3/m$      | $P6_3/m$                         | $P6_3/m$      |
| Symmetry    | Hexagonal     | Hexagonal                        | Hexagonal     | Hexagonal                        | Hexagonal     |
| a, Å        | 9.568431(5)   | 9.582288(8)                      | 9.617445(2)   | 9.627483(2)                      | 9.650372(3)   |
| b, Å        | 9.568431(2)   | 9.582288(7)                      | 9.617445(3)   | 9.627483(5)                      | 9.650372(2)   |
| c, Å        | 7.032033(4)   | 7.038839(2)                      | 7.059281(4)   | 7.063548(7)                      | 7.094781(5)   |
| V, Å        | 557.562       | 559.719                          | 565.471       | 566.995                          | 572.213       |
| Z           | 6             | 6                                | 6             | 6                                | 6             |
| $R_p$       | 6.84          | 6.97                             | 7.63          | 9.21                             | 5.24          |
| $R_{wp}$    | 9.35          | 9.83                             | 10.65         | 11.54                            | 7.71          |
| $\chi^2$    | 1.373         | 1.372                            | 1.177         | 1.249                            | 1.273         |

**Table S2.** Wyckoff Positions of Group  $P63/m$  (No. 176).

| Multiplicity | Wyckoff letter | Site symmetry | Coordinates  |
|--------------|----------------|---------------|--|
| y            |                |               |  |
|              |                |               | $(x,y,z) (-y,x-y,z) (-x+y,-x,z) (-x,-y,z+1/2)$   |
| 12           | i              | 1             | $(y,-x+y,z+1/2) (x-y,x,z+1/2) (-x,-y,-z)$<br>$(y,-x+y,-z) (x-y,x,-z) (x,y,-z+1/2)$<br>$(-y,x-y,-z+1/2) (-x+y,-x,-z+1/2)$ |
| 6            | h              | m..           | $(x,y,1/4) (-y,x-y,1/4) (-x+y,-x,1/4)(-x,-y,3/4)$<br>$(y,-x+y,3/4) (x-y,x,3/4)$  |
| 6            | g              | -1            | $(1/2,0,0) (0,1/2,0) (1/2,1/2,0) (1/2,0,1/2)$<br>$(0,1/2,1/2) (1/2,1/2,1/2)$   |
| 4            | f              | 3..           | $(1/3,2/3,z) (2/3,1/3,z+1/2) (2/3,1/3,-z)$<br>$(1/3,2/3,-z+1/2)$   |
| 4            | e              | 3..           | $(0,0,z) (0,0,z+1/2) (0,0,-z) (0,0,-z+1/2)$  |
| 2            | d              | -6..          | $(2/3,1/3,1/4) (1/3,2/3,3/4)$  |
| 2            | c              | -6..          | $(1/3,2/3,1/4) (2/3,1/3,3/4)$  |
| 2            | b              | -3..          | $(0,0,0) (0,0,1/2)$  |
| 2            | a              | -6..          | $(0,0,1/4) (0,0,3/4)$  |



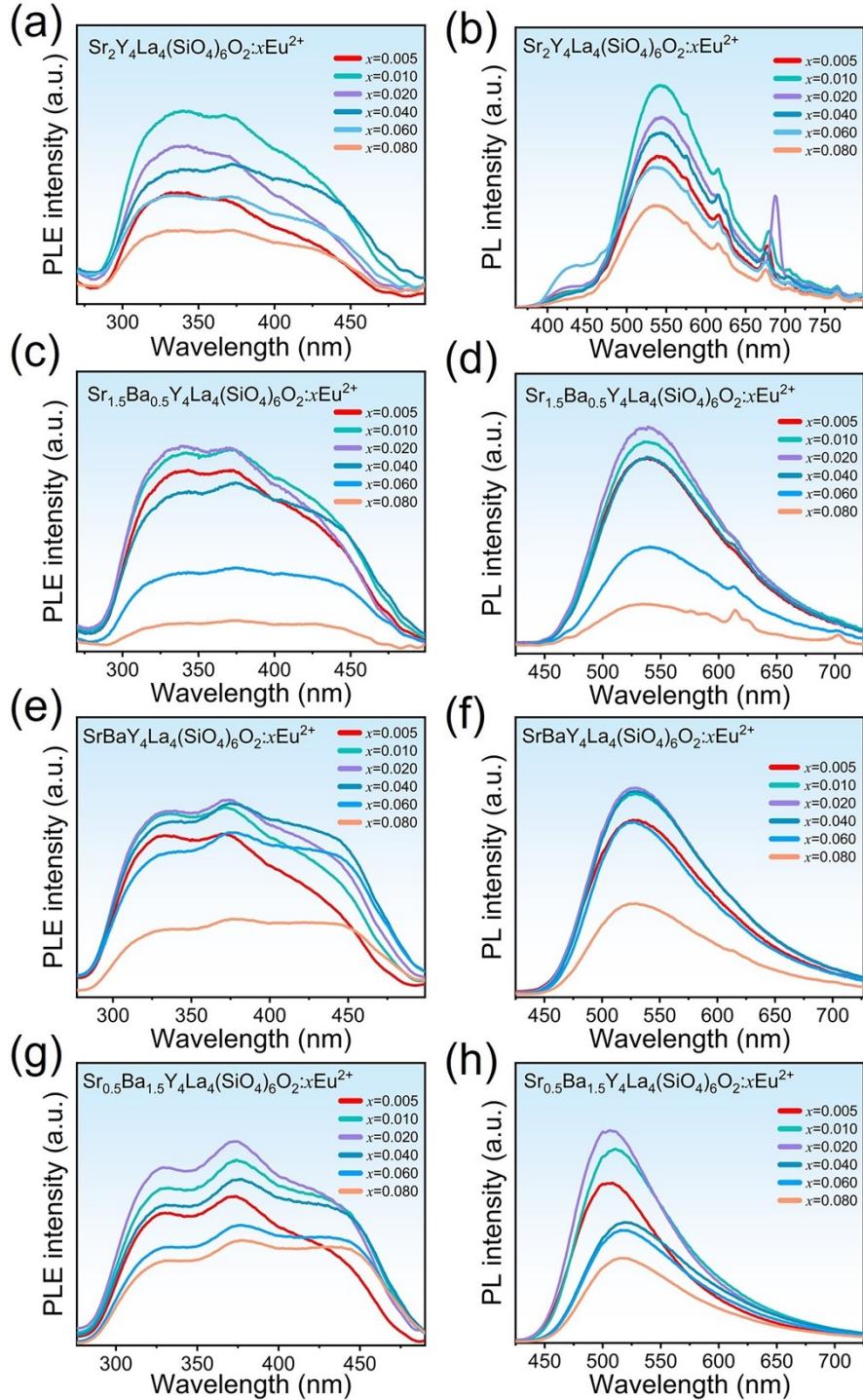
**Figure S1.** X-ray photoelectron spectroscopy (XPS) of  $\text{Ba}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.02\text{Eu}^{2+}$  phosphor, Figure on the right shows the high-resolution XPS spectrum of the Eu 3d core level of the sample.



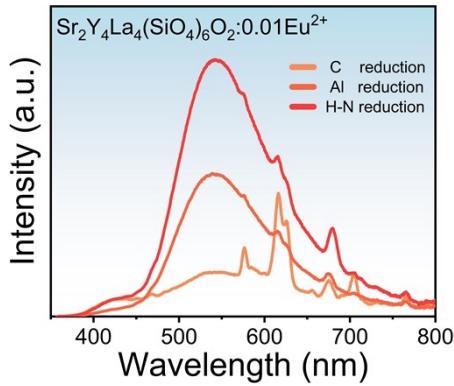
**Figure S2.** (a) Photoluminescence spectra of undoped  $\text{Eu}^{2+}$  samples and doped  $\text{Eu}^{2+}$ . (b) Images of the undoped  $\text{Eu}^{2+}$  sample under daylight illumination. (c) Images of the undoped  $\text{Eu}^{2+}$  samples under 365 nm UV light.

**Table S3.** Bond lengths in the Ba<sub>2</sub>Y<sub>4</sub>La<sub>4</sub>(SiO<sub>4</sub>)<sub>6</sub>O<sub>2</sub>:0.02Eu<sup>2+</sup> compound.

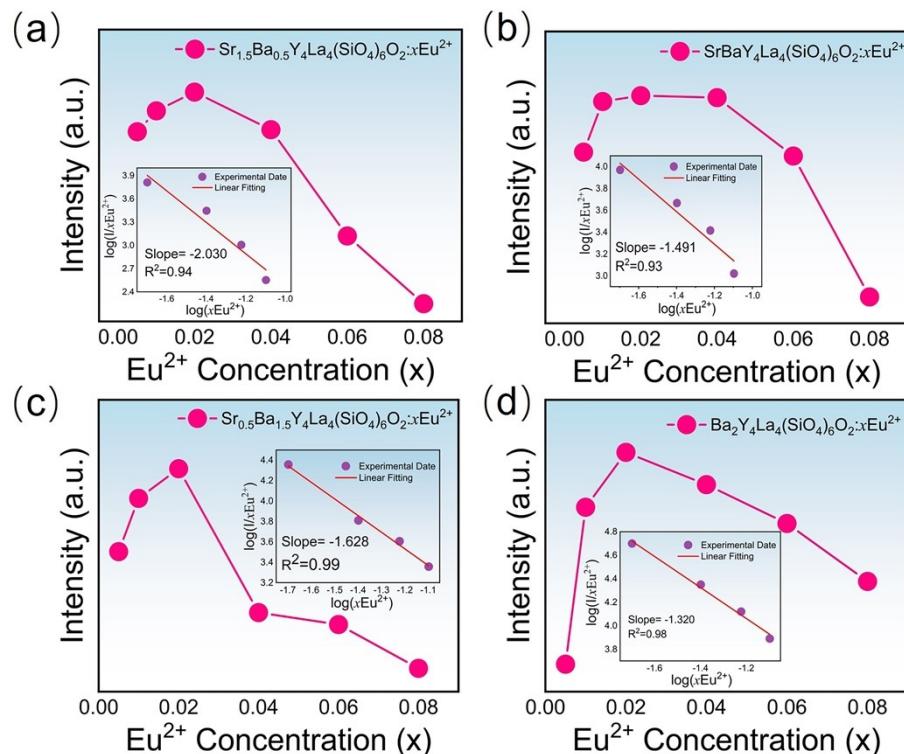
| Selected atoms     | Bond length (Å) | Selected atoms     | Bond length (Å) |
|--------------------|-----------------|--------------------|-----------------|
| Ba1-O1             | 2.469(7)        | Ba2-O1             | 2.691(5)        |
| Ba1-O1             | 2.469(7)        | Ba2-O2             | 2.467(5)        |
| Ba1-O1             | 2.468(7)        | Ba2-O3             | 3.402(5)        |
| Ba1-O2             | 2.526(6)        | Ba2-O3             | 2.545(4)        |
| Ba1-O2             | 2.527(6)        | Ba2-O3             | 2.4142(16)      |
| Ba1-O2             | 2.526(6)        | Ba2-O3             | 3.402(5)        |
| Ba1-O3             | 2.8749(15)      | Ba2-O3             | 2.4142(16)      |
| Ba1-O3             | 2.8739(15)      | Ba2-O3             | 2.545(4)        |
| Ba1-O3             | 2.8746(15)      | Ba2-O4             | 2.287(5)        |
| Y1-O1              | 2.469(7)        | Y2-O1              | 2.691(5)        |
| Y1-O1              | 2.469(7)        | Y2-O2              | 2.467(5)        |
| Y1-O1              | 2.468(7)        | Y2-O3              | 2.4142(16)      |
| Y1-O2              | 2.526(6)        | Y2-O3              | 2.545(4)        |
| Y1-O2              | 2.527(6)        | Y2-O3              | 2.4142(16)      |
| Y1-O2              | 2.526(6)        | Y2-O3              | 2.545(4)        |
| Y1-O3              | 2.8749(15)      | Y2-O4              | 2.287(5)        |
| Y1-O3              | 2.8739(15)      | La2-O1             | 2.691(5)        |
| Y1-O3              | 2.8746(15)      | La2-O2             | 2.467(5)        |
| La1-O1             | 2.469(7)        | La2-O3             | 2.545(4)        |
| La1-O1             | 2.469(7)        | La2-O3             | 2.4142(16)      |
| La1-O1             | 2.468(7)        | La2-O3             | 2.545(4)        |
| La1-O2             | 2.526(6)        | La2-O3             | 2.4142(16)      |
| La1-O2             | 2.527(6)        | La2-O4             | 2.287(5)        |
| La1-O2             | 2.526(6)        |                    |                 |
| La1-O3             | 2.8749(15)      |                    |                 |
| La1-O3             | 2.8739(15)      |                    |                 |
| La1-O3             | 2.8746(15)      |                    |                 |
| Average length (Å) | 2.6232          | Average length (Å) | 2.5606          |



**Figure S3.** Concentration gradient (a) PLE and (b) PL spectra of  $\text{Sr}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{xEu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ). Concentration gradient (c) PLE and (d) PL spectra of  $\text{Sr}_{1.5}\text{Ba}_{0.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{xEu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ). Concentration gradient (e) PLE and (f) PL spectra of  $\text{SrBaY}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{xEu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ). Concentration gradient (g) PLE and (h) PL spectra of  $\text{Sr}_{0.5}\text{Ba}_{1.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{xEu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ).



**Figure S4.** Photoluminescence spectra of  $\text{Sr}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.01\text{Eu}^{2+}$  samples under various reducing atmospheres.



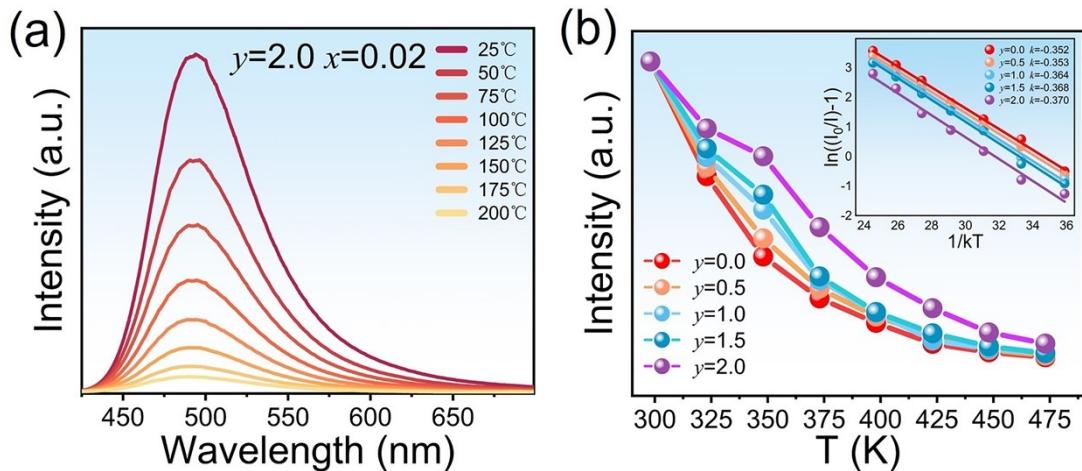
**Figure S5.** Emission intensity varies with increasing  $\text{Eu}^{2+}$  concentration  $x$ , inset shows the linear fitting of  $\log(x)$  versus  $\log(I/x)$  for phosphors with  $x\text{Eu}^{2+}$  concentrations exceeding the quenching concentration of (a)  $\text{Sr}_{1.5}\text{Ba}_{0.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}(0.005 \leq x \leq 0.080)$ , (b)  $\text{SrBaY}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}(0.005 \leq x \leq 0.080)$ , (c)  $\text{Sr}_{0.5}\text{Ba}_{1.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}(0.005 \leq x \leq 0.080)$ , (d)  $\text{Ba}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}(0.005 \leq x \leq 0.080)$ .

**Table S4.** Slope obtained from the linear fitting of  $\log(x)$  versus  $\log(I/x)$  along with the calculated  $\theta$  value for  $\text{Sr}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ),  $\text{Sr}_{1.5}\text{Ba}_{0.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ),  $\text{SrBaY}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ),  $\text{Sr}_{0.5}\text{Ba}_{1.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ),  $\text{Ba}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.080$ ).

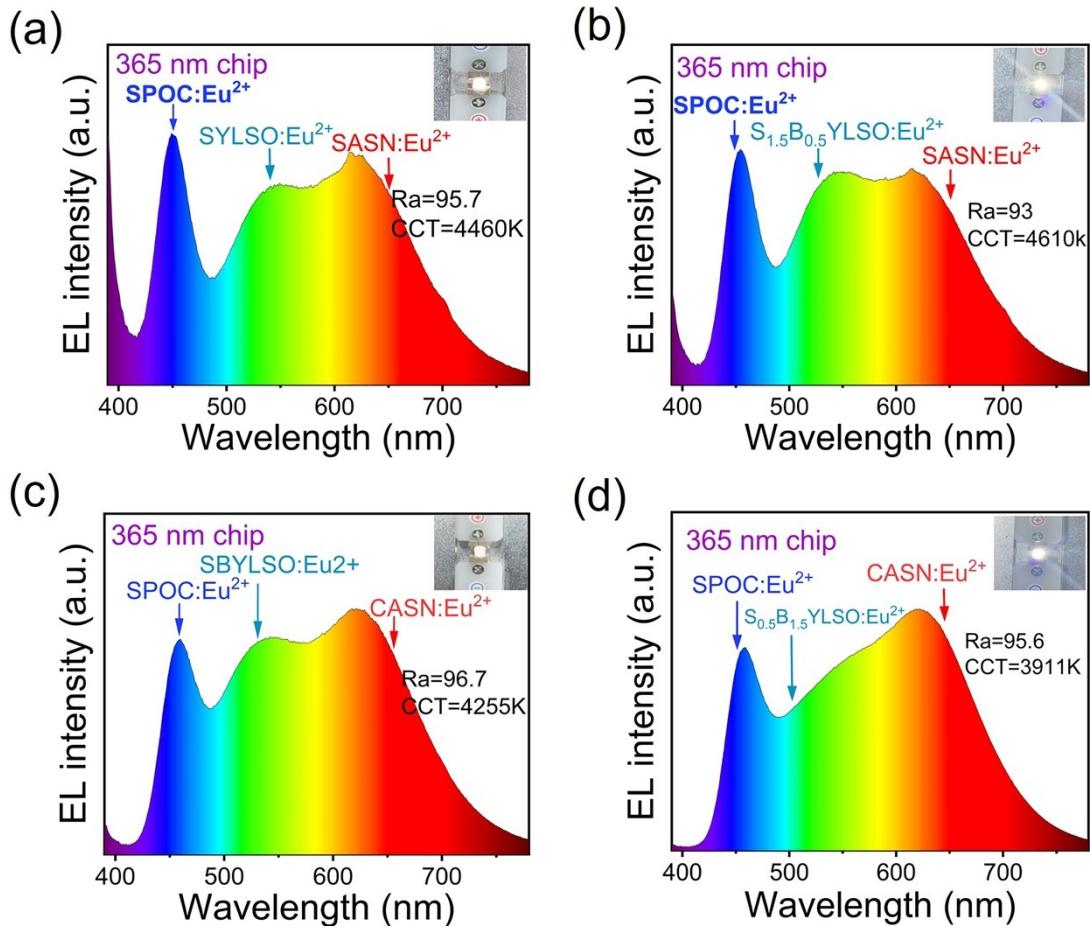
| Sample   | $\text{Sr}_2$ | $\text{Sr}_{1.5}\text{Ba}_{0.5}$ | $\text{SrBa}$ | $\text{Sr}_{0.5}\text{Ba}_{1.5}$ | $\text{Ba}_2$ |
|----------|---------------|----------------------------------|---------------|----------------------------------|---------------|
| slope    | -1.321        | -2.030                           | -1.491        | -1.628                           | -1.320        |
| $\theta$ | 3.963         | 6.090                            | 4.473         | 4.884                            | 3.960         |

**Table S5.** Gaussian deconvolution of the  $\text{Sr}_{(2-y)}\text{Ba}_y\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:x\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.08$ ,  $0.5 \leq y \leq 2$ ) series of phosphors and their corresponding excitation and emission wavelengths.

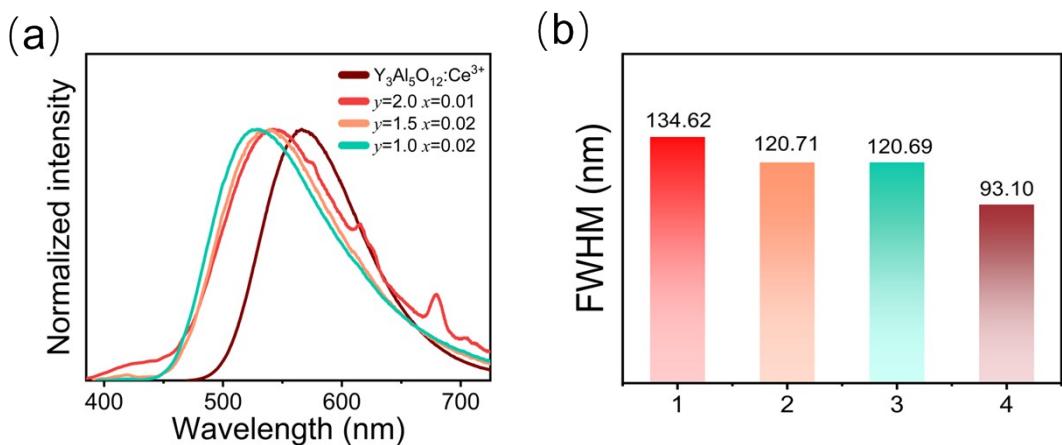
| $y$ | EuI(nm) | EuII(nm) | Ex1(nm) | Ex2(nm) | Em1(nm) | Em2(nm) |
|-----|---------|----------|---------|---------|---------|---------|
| 0.0 | 537     | 603      | 338     | 340     | 545     | 543     |
| 0.5 | 530     | 592      | 373     | 343     | 537     | 542     |
| 1.0 | 524     | 591      | 374     | 340     | 528     | 534     |
| 1.5 | 504     | 560      | 373     | 372     | 500     | 501     |
| 2.0 | 490     | 531      | 371     | 379     | 494     | 495     |



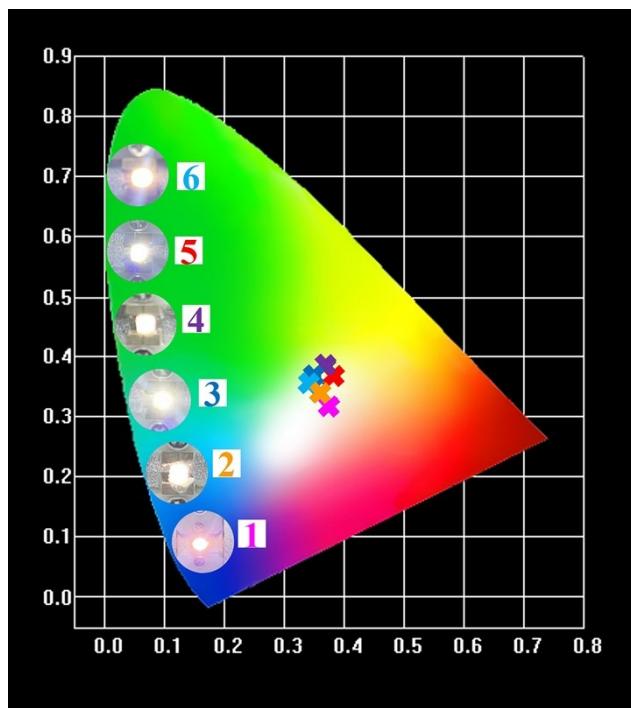
**Figure S6.** (a) Photoluminescence spectra of  $\text{Ba}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.02\text{Eu}^{2+}$  phosphors in the temperature range of 25-200°C. (b) Relationship between luminescence intensity and temperature for the  $\text{Sr}_{(2-y)}\text{Ba}_y\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.08$ ,  $0.5 \leq y \leq 2$ ) series of phosphors, the inset shows the relationship between  $\ln((I_0/I)-1)$  and  $1/kT$  for the  $\text{Sr}_{(2-y)}\text{Ba}_y\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:\text{Eu}^{2+}$  ( $0.005 \leq x \leq 0.08$ ,  $0.5 \leq y \leq 2$ ) series of phosphors.



**Figure S7.** EL spectra of devices fabricated by uniformly mixing the prepared yellow phosphors (a)  $\text{Sr}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.01\text{Eu}^{2+}$  / (b)  $\text{Sr}_{1.5}\text{Ba}_{0.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.02\text{Eu}^{2+}$  / (c)  $\text{SrBaY}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.01\text{Eu}^{2+}$ , and / (d) yellow-green phosphor  $\text{Sr}_{0.5}\text{Ba}_{1.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.01\text{Eu}^{2+}$  with commercial blue phosphors  $\text{Sr}_5(\text{PO}_4)_3\text{Cl}:\text{Eu}^{2+}$  (SPOC:Eu<sup>2+</sup>) and commercial red phosphors  $\text{CaAlSiN}_3:\text{Eu}^{2+}$  (CASN:Eu<sup>2+</sup>) and then coating them onto 365 nm chips, inset displays a photograph of the packaged WLED under illumination.



**Figure S8.** (a) Normalized spectral diagrams of  $\text{Sr}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.01\text{Eu}^{2+}$ ,  $\text{Sr}_{1.5}\text{Ba}_{0.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.02\text{Eu}^{2+}$ ,  $\text{SrBaY}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.02\text{Eu}^{2+}$  and  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ . (b) full-width at half-maximum (FWHM) of the spectra of  $\text{Sr}_2\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.01\text{Eu}^{2+}$ ,  $\text{Sr}_{1.5}\text{Ba}_{0.5}\text{Y}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.02\text{Eu}^{2+}$ ,  $\text{SrBaY}_4\text{La}_4(\text{SiO}_4)_6\text{O}_2:0.02\text{Eu}^{2+}$  and  $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$ .



**Figure S9.** Emission positions of devices (1-6) WLED(0), WLED( $\text{Sr}_2$ ), WLED( $\text{Sr}_{1.5}$ ), WLED( $\text{Sr}_1$ ), WLED( $\text{Sr}_{0.5}$ ), and WLED( $\text{Ba}_2$ ), in the CIE diagram.