Electronic Supplementary Material (ESI) for Dalton Transactions. This journal is © The Royal Society of Chemistry 2023

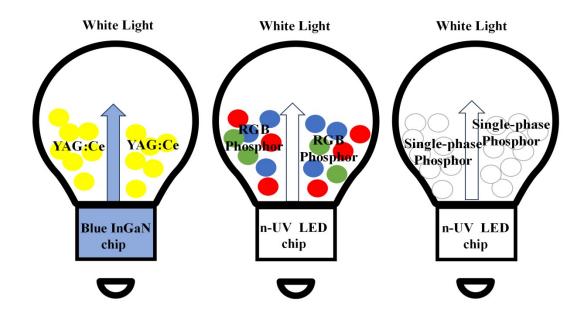
## **Supporting information**

Structure and Luminescence Properties of Single-Component Melilite Sr<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub>: Ce/Tb/Sm for *n*-UV *w*LEDs

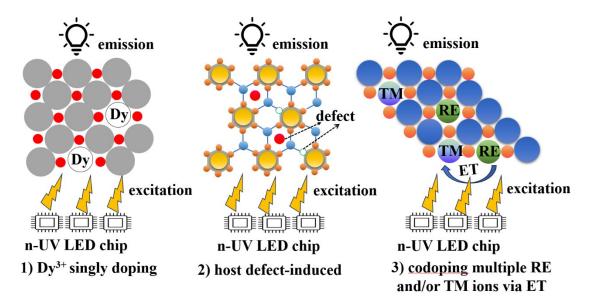
X. Z. Zheng, L. Yue, C. Wang, P. J. Xia, M. Xu and W. B. Dai\*

Hubei Key Laboratory of Plasma Chemistry and Advanced Materials & Key Laboratory of Green Chemical Engineering Process of Ministry of Education, Wuhan Institute of Technology, 430205, Wuhan, China

Corresponding author: wubin.dai@wit.edu.cn (W. Dai)



Scheme S1 Sketch map of the three mainstream modes for construction of pc-wLEDs (from left to right), blue InGaN LED chip + yellow phosphor YAG: Ce, ii) *n*-UV chip + red, green and blue phosphors, and iii) *n*-UV chip + single-phase RE/TM ions (co)doped phosphor.



Scheme S2 Sketch map of three ways to achieve white PL *via* the single-phase phosphor (from left to right), 1) singly doping, i.e., Dy<sup>3+</sup>, 2) host defect-induced, and 3) codoping multiple RE and/or TM ions.

| Items                           | Parameters |
|---------------------------------|------------|
| Primary and second radius       | 217.5 nm   |
| Receiving slit length           | 13.65°     |
| Source and sample length        | 12 mm      |
| Primary slit aperture           | 2.5°       |
| Reception slit divergence angle | 0.2°       |
| Receiving slit width            | 0.1 mm     |
| Peak-shape function             | Lorentzian |

Table S1 Instrumental data used for Rietveld refinements of the Sr2MgSi2O7 host and its RE

and/or TM ions (co)doped derivatives

**(b) (a)** SMS:1.5%Sm SMS:1%Ce × 15-0016 15-0016 obs obs X calc calc Intensity(a.u.) Intensity(a.u.) diff diff Rwp=13.324% R<sub>wp</sub>=13.819% R<sub>p</sub>=9.716% R<sub>n</sub>=10.039% 40 50 2θ(°) . 30 4020(°)50 20 30 70 80 10 20 60 70 80 10 60 90 90 SMS:10%Tb,7%Sm (c) 15-0016 L obs × Intensity(a.u.) calc diff R<sub>wp</sub>=13.336% R<sub>0</sub>=9.641% i i 1III 50 2θ(°) 10 20 30 40 60 70 80 90

Fig.S1 Rietveld refinement of the powder XRD profiles for the representative samples SMS:1%Ce a), SMS: 1.5%Sm b) and SMS: 10%Tb, 7%Sm c). The experimental data, calculated data and the corresponding difference profiles were also shown.

Table S2 Lattice parameters and atom positions of the Sr<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub>: 1%Ce<sup>3+</sup>, 13%Tb<sup>3+</sup>,

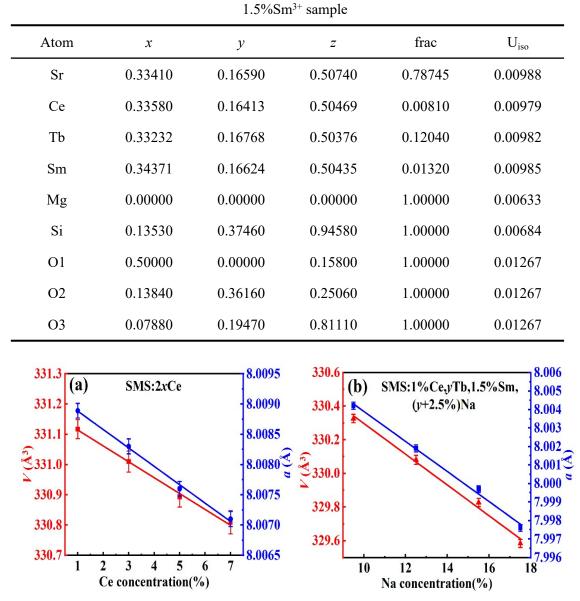


Fig.S2 The variations of the lattice parameters a and V as a function of Ce content a) and Tb/Na contents b), respectively.

Table S3 The weight and atomic percent of each atom in Sr<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub>: 1%Ce<sup>3+</sup>,

|      |            | •          |
|------|------------|------------|
| Atom | Weight (%) | Atomic (%) |
| Sr   | 42.72      | 15.375     |
| Mg   | 6.42       | 8.33       |
| Si   | 14.84      | 16.67      |
| 0    | 29.59      | 58.34      |
| Ce   | 0.37       | 0.08       |
| Tb   | 5.46       | 1.08       |
| Sm   | 0.6        | 0.125      |

13%Tb<sup>3+</sup>,1.5%Sm<sup>3+</sup> sample

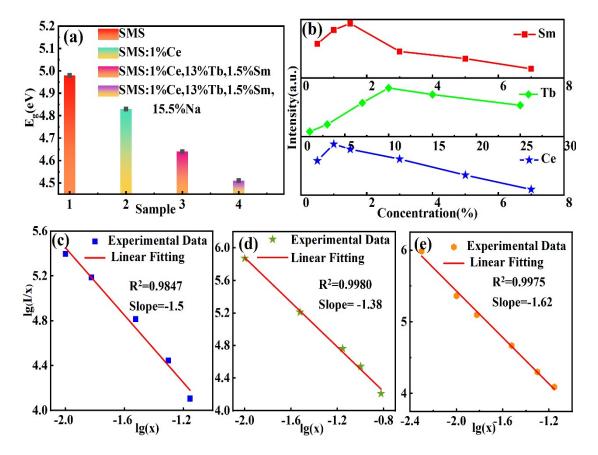


Fig.S3 a) The variation of the  $E_g$  values without and with different content of dopants for the SMS host. b) The variation of the PL intensity as a function of the doping concentration for the Ce, Tb and Sm singly doped samples, respectively. The fitting lines of the lg(I/x) vs. lg(x) in the phosphors of SMS:  $2xCe^{3+}$  c), SMS:  $2yTb^{3+}$  d) and SMS:  $2zSm^{3+}$  e), respectively.

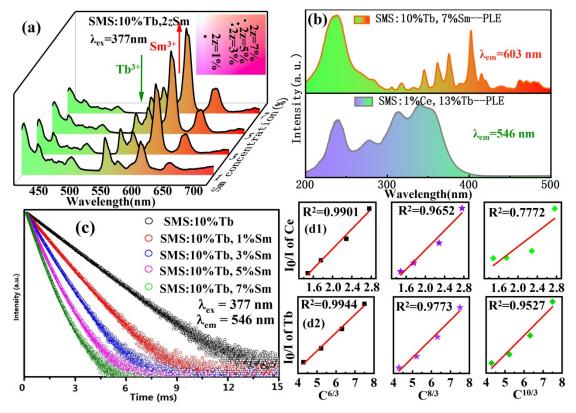


Fig.S4 a) The variations of the PL intensities for the Tb and Sm in the codoped phosphors SMS: 10%Tb, 2zSm under λ<sub>ex</sub> = 377 nm. The inset shows the CIE coordinates of the phosphors. b) The PLE spectra of the codoped phosphors SMS: 1%Ce, 13%Tb and SMS: 10%Tb, 7%Sm, respectively under monitored at 546 nm and 603 nm. c) The decay curves of Tb<sup>3+</sup> PL in SMS: 10%Tb, 2zSm phosphors under under λ<sub>ex</sub> = 377 nm and λ<sub>em</sub> = 546 nm. The fitting lines of the *I*<sub>0</sub>/*I vs*. C<sup>θ/3</sup> (θ = 6, 8, and 10) in the phosphors of SMS: 1%Ce, 2yTb d1) and SMS: 10%Tb, 2zSm d2), respectively.

Table S4 The average lifetimes of Ce/Tb ions and ET efficiencies in phosphors SMS: 1%Ce,

| 2yTb and | SMS: | 10%Tb, | 2zSm |
|----------|------|--------|------|
|          |      |        |      |

| Phosphor            | 0        | 5        | 7        | 10       | 13       |
|---------------------|----------|----------|----------|----------|----------|
| 1%Ce, 2 <i>y</i> Tb | 49.32 ns | 33.02 ns | 27.91 ns | 21.49 ns | 17.76 ns |
| η                   | -        | 33.05%   | 43.41%   | 56.43%   | 64%      |
| Phosphor            | 0        | 1        | 3        | 5        | 7        |
| 10%Tb, 2zSm         | 1.81 ms  | 0.35 ms  | 0.27 ms  | 0.23 ms  | 0.19 ms  |
| η                   | -        | 80.73%   | 85.07%   | 87.53%   | 89.5%    |

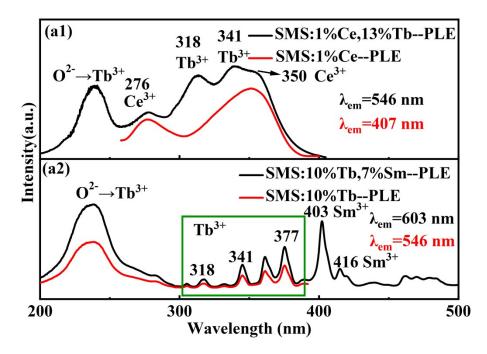


Fig.S5 The comparisons of the PLE spectra for the phosphors SMS: Ce (SMS: Tb) and SMS: Ce/Tb (SMS: Tb/Sm) monitored at the values of PL of Ce (Tb) and PL of Tb (Sm),

respectively, in the a1 and a2.

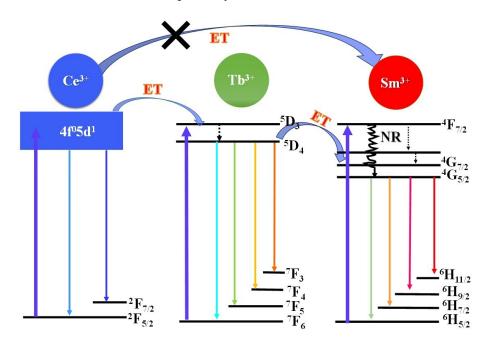


Fig.S6 ET processes, partial energy levels and visible emission transitions among the Ce/Tb/Sm ions in SMS host.

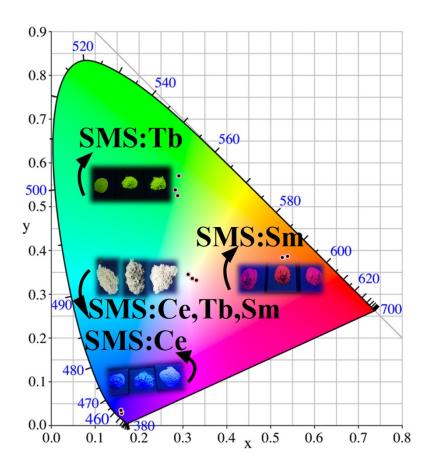


Fig.S7 CIE chromaticity diagram and digital photographs of the singly- and tri-doped SMS: Ce/Tb/Sm phosphors.

| Samples <sup>a</sup>                                       | CIE(x,y)         | Peak   | CCT/K    |
|--|------------------|--|----------|
| <i>x</i> = 0.5%  | (0.1604, 0.0315) | 406 nm (Ce <sup>3+</sup> )   | 1736.725 |
| x = 1%   | (0.1598, 0.0285) | 407 nm (Ce <sup>3+</sup> )   | 1745.604 |
| x = 3%   | (0.1584, 0.0346) | 408 nm (Ce <sup>3+</sup> )   | 1722.446 |
| <i>y</i> = 7%  | (0.2886, 0.5252) | 546 nm (Tb <sup>3+</sup> )   | 6451.44  |
| <i>y</i> = 10%   | (0.2829, 0.539)  | 546 nm (Tb <sup>3+</sup> )   | 6538.854 |
| <i>y</i> = 13%   | (0.2906, 0.5703) | 546 nm (Tb <sup>3+</sup> )   | 6295.343 |
| <i>z</i> =0.5%   | (0.5381, 0.3683) | 603 nm (Sm <sup>3+</sup> )   | 1729.224 |
| z=1%   | (0.5293, 0.3842) | 603 nm (Sm <sup>3+</sup> )   | 1822.758 |
| <i>z</i> =1.5%   | (0.5391, 0.3881) | 603 nm (Sm <sup>3+</sup> )   | 1795.578 |
| <i>x</i> = 1%, <i>y</i> = 13%, <i>z</i> = 1.5%             | (0.3312, 0.3226) | 407 nm (Ce <sup>3+</sup> ),546 nm (Tb <sup>3+</sup> ),603 nm (Sm <sup>3+</sup> ) | 5554.556 |
| x = 1%, y = 13%, z = 1.5%<br>(x + y + z) = 15.5%           | (0.3124, 0.3458) | 407 nm (Ce <sup>3+</sup> ),546 nm (Tb <sup>3+</sup> ),603 nm (Sm <sup>3+</sup> ) | 6409.623 |
| x = 1%, y = 13%, z = 1.5%<br>(x + y + z) = 15.5%, 2a = 10% | (0.3217, 0.3361) | 407 nm (Ce <sup>3+</sup> ),546 nm (Tb <sup>3+</sup> ),603 nm (Sm <sup>3+</sup> ) | 6001.544 |

Table S5 CIE coordinates and CCT of  $Sr_2MgSi_2O_7$ :  $xCe^{3+}$ ,  $yTb^{3+}$ ,  $zSm^{3+}$ ,  $(x + y + z) Na^+$ ,  $2aBa^{2+}$  phosphors

<sup>a</sup>x, y, z, a and (x + y + z) represent the doping concentrations of Ce, Tb, Sm, Ba and Na in Sr<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub> phosphor, respectively.

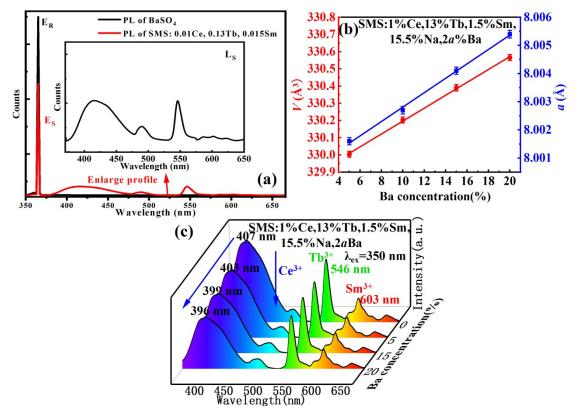


Fig.S8 a) Excitation line of BaSO<sub>4</sub> and the PL of the SMS: 1%Ce, 13%Tb, 1.5%Sm phosphor collected *via* the integrating sphere. The inset shows a magnification of the PL spectrum. b)
The variation of the lattice parameters *a* and *V* as a function of Ba content in the phosphor
SMS: 1%Ce, 13%Tb, 1.5%Sm, 15.5%Na.c) The variation of PL spectra from Ce as a function of Ba content in the same phosphor.