

Supplementary Information

Elevated colour rendering of white-LEDs by microwave synthesized red-emitting (Li, Mg)₃RbGe₈O₁₈: Mn⁴⁺ nanophosphors

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Experimental techniques:

GATAN software: The distance between two diametrically opposite spots is measured by drawing a line passing through the central bright spot using GATAN software [ref. S1]. From this value, the distance between the central bright spot and the diffracted spot is calculated, the inverse of which gives the d-value. This d-value is compared with that obtained from the XRD refinement, from which (h, k, l) values can be obtained. Each circle connects all the diffracted spots having the same distance from the central bright spot.

Colour purity calculations: The values of red color purity were calculated via the equation given below [ref. S2]:

$$\text{Colour purity} = [\{(x_s - x_i)^2 + (y_s - y_i)^2\}^{1/2} / \{(x_d - x_i)^2 + (y_d - y_i)^2\}^{1/2}] \times 100\% \quad (\text{equ. S1})$$

where (x_s, y_s) are designated as the CIE coordinates of the experimental sample, (x_d, y_d) are the CIE coordinates of the dominant wavelength, and (x_i, y_i) are the CIE coordinates of the illuminant point. Herein, $(x_i, y_i) = (0.3101, 0.3162)$. The overall process was carried out according to the ref. S3.

Low-temperature photoluminescence: Low-temperature PL measurements were performed using a continuous-flow liquid He cryostat mounted on a three-axis motorized translation stage to control the sample position. The excitation of the sample was provided by a continuous-wave Oxxius laser with a wavelength of 266 nm operating at a constant power of 4 mW, focused into a spot of about 5 μm . The PL spectra were recorded for temperatures from 7 K to 300 K using a Horiba HR460 spectrometer with 600 grooves/mm grating equipped with a Si charge coupled device (CCD) detectors.

Results and discussions:

Figures S1 and S2 show the survey spectra and the core level spectra for the sample prepared through SSR and MWD methods respectively. The binding energy curve of Li 1s atom for the sample synthesized through the SSR method could be deconvoluted into 55.34 eV and 51.84 eV while for the sample synthesized through MWD, the peak could be fitted with a single curve centered at 54.45 eV. The core-level spectrum of Ge 3d for the sample synthesized through SSR method could be deconvoluted into two peaks centered at 32.56 eV and at 32.19 eV and they correspond to Ge1 and Ge2 respectively. The de-convoluted peaks Ge1 and Ge2 of the sample prepared through MWD method are centered at 32.59 eV and 32.30 eV respectively. The binding

energy peaks of Rb $3d_{5/2}$ and Rb $3d_{3/2}$ of the Rb 3d atom prepared through the SSR can be observed at 110.03 eV and 111.5 eV respectively. For the sample prepared through MWD method, the binding energy peaks corresponding to Rb $3d_{5/2}$ and Rb $3d_{3/2}$ are located at 110.16 eV and 111.65 eV respectively. Considering the O 1s region of the spectra, for the sample prepared through the SSR method, the peak is observed at 531.88 eV. This corresponds to the O^{2-} oxidation state of O ion. For the sample prepared through MWD method, the spectra consist of two peaks, at 531.59 eV and at 533.18 eV, the lower binding energy peak corresponding to the O^{2-} oxidation state and the higher one corresponding to O^{chem} , the chemically adsorbed oxygen on the surface.

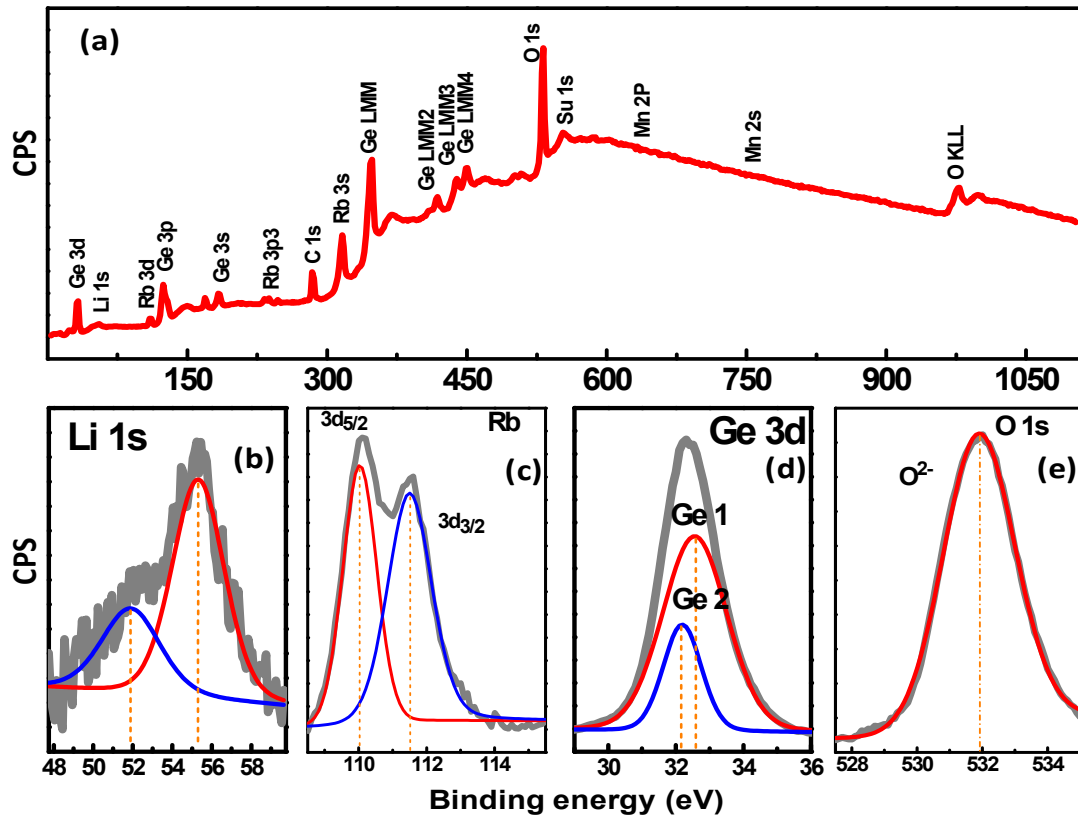


Figure S1: (a) The XPS survey spectrum of the SSR derived $Li_3RbGe_{7.995}O_{18}: 0.005Mn^{4+}$, (b), (c), (d) and (e) show the XPS core level spectra of Li 1s, Rb 3d, Ge 3d and O 1s, respectively, of the sample $Li_3RbGe_{7.995}O_{18}: 0.005Mn^{4+}$ prepared through SSR method.

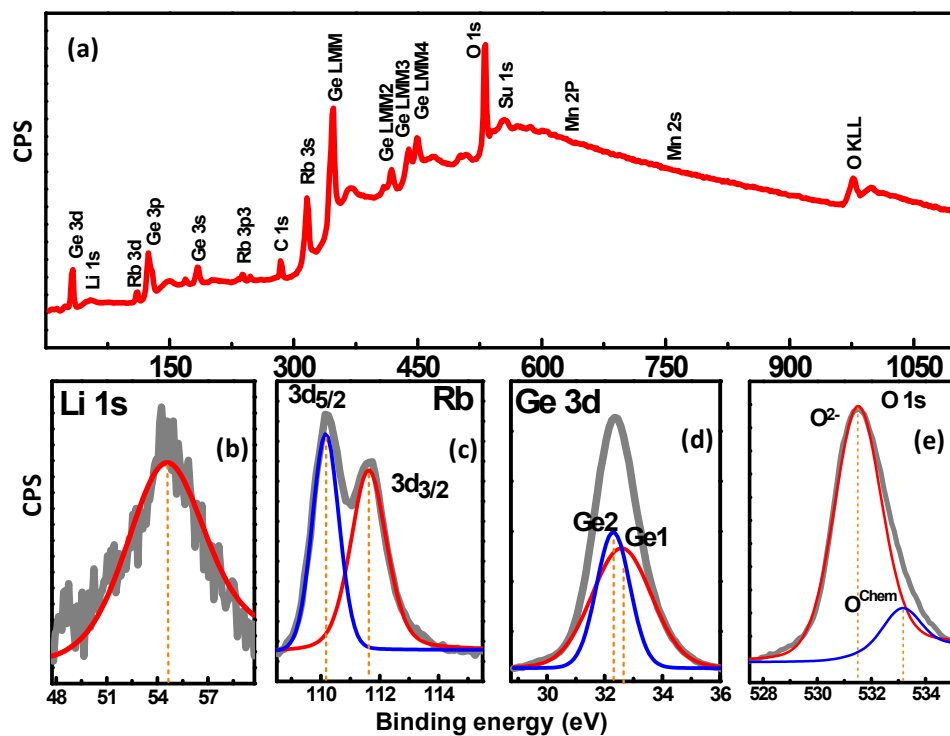


Figure S2: The XPS survey spectrum of MWD derived $\text{Li}_3\text{RbGe}_{7.995}\text{O}_{18}: 0.005\text{Mn}^{4+}$. (b), (c), (d) and (e) show the XPS core level spectra of Li 1s, Rb 3d, Ge 3d and O 1s, respectively, of $\text{Li}_3\text{RbGe}_{7.995}\text{O}_{18}: 0.005\text{Mn}^{4+}$ prepared through MWD method.

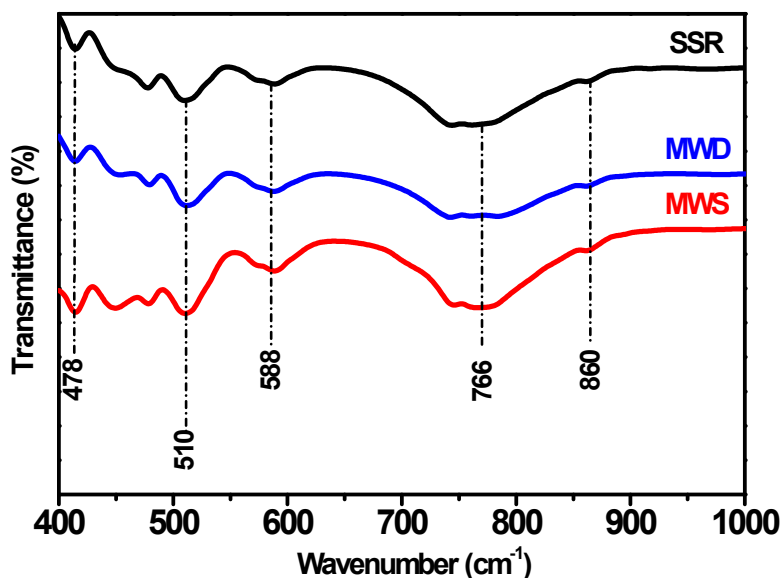


Figure S3: The FTIR spectra of the $\text{Li}_3\text{RbGe}_{7.995}\text{O}_{18}: 0.005\text{Mn}^{4+}$ sample synthesized through (i) the solid state reaction (SSR) method, (ii) the microwave assisted diffusion (MWD) method, and (iii) the microwave assisted sol-gel (MWS) method.

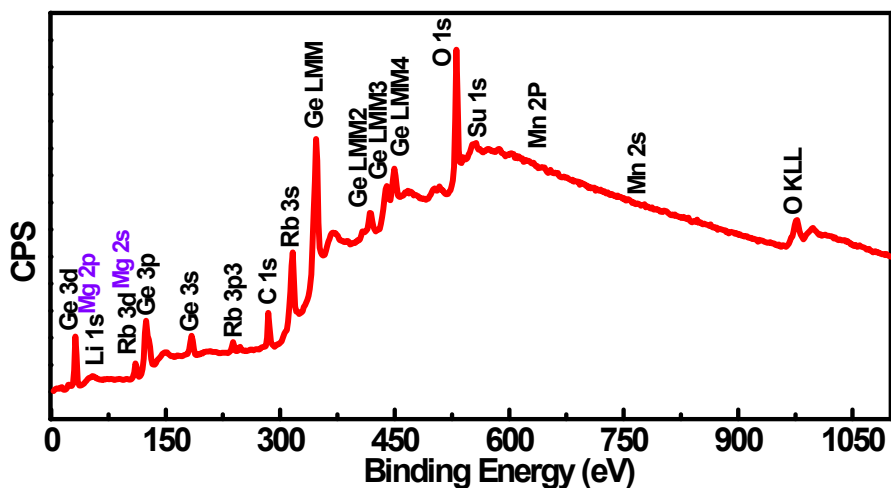


Figure S4: The XPS survey spectrum of the sample $\text{Li}_{1.97}\text{RbGe}_{7.995}\text{O}_{18} \cdot 0.005\text{Mn}^{4+}, 0.015\text{Mg}^{2+}$.

Table S1: The table shows the atomic coordinates, fractional occupancies and isotropic displacement parameters.

Atoms	Name	x	y	z	Fraction of occupancy	Uiso
Rb	Rb	0	0	0	1	0.03039
Ge	Ge1	0.39664	0.19835	0.5	1	0.00625
Ge	Ge2	0.66667	0.33333	0	0.995	0.01569
O	O1	0.50358	0.33241	0.22749	1	0.01623
O	O2	0.27197	0	0.36233	1	0.04331
Li	Li	0.5	0	0	1	0.09153
Mn	Mn	0.66667	0.33333	0	0.005	0.32179

Table S2: The table shows the bonds and corresponding bond lengths that exist between the elements in the sample.

Bond	Bond length (Å)
Rb-O2	3.13257 (×6)
Ge1-O1	1.75251
Ge1-O1	1.75277
Ge1-O2	1.79467
Ge1-O2	1.79424
Ge2-O1	1.90198 (×2)
Ge2-O1	1.9019 (×2)
Ge2-O1	1.90194 (×2)
Ge2-Li	2.77828 (×2)
Ge2-Li	2.77836
Li-O1	1.9559 (×4)
Mn-O1	1.90196
Mn-O1	1.90192 (×2)
Mn-O1	1.90195 (×2)
Mn-O1	1.90191
Mn-Li	2.7783
Mn-Li	2.77828
Mn-Li	2.77833
O1-Ge1	1.75251
O1-Ge2	1.90198
O1-Mn	1.90196
O1-Li	1.9559
O2-Rb	3.13257
O2-Ge1	1.79467 (×2)
Li-Ge2	2.77828 (×2)
Li-Mn	2.7783
Li-Mn	2.77828

Table S3: The value of CCT, CRI and CIE of the EL spectra recorded for the W-LEDs fabricated with the various mixing ratios of YAG: Ce³⁺ and LRGO: 0.005Mn⁴⁺, 0.015Mg²⁺ ((i) 1:0, (ii) 1:1, (iii) 2:3, and (iv) 3:7, respectively), and a 460 nm blue InGaN chip.

Sample	CCT	CRI	CIE-x	CIE-y
YAG	6952	71	0.31	0.34
YAG: LRGO (50: 50)	6149	79	0.32	0.35
YAG: LRGO (40:60)	5674	82	0.33	0.35
YAG: LRGO (30:70)	5025	92	0.34	0.35

References:

- S1.** H. C. A. Murthy, T. D. Zeleke, C. R. Ravikumar, M. R. A. Kumar, and H. P. Nagaswarupa, Mater. Res. Express 7 (2020) 055016.
- S2.** Z. Liang, Z. Yang, X. Xie, H. Pu. D. Shi, Q. Zhou, Z. Wang, J. Guo and M. Wu, Dalton Trans., 2019, 48, 12459.
- S3.** T. M. Goodman, 7 - International standards for colour, Colour Design, Woodhead Publishing, 2012, Pages 177-218