Lanthanide doped lead-free double perovskite La₂MgTiO₆ as ultra-bright multicolour LEDs and novel selfcalibration partition optical thermometer Keming Zhu^a, Hanyu Xu^a, Zhiying Wang^a, Zuoling Fu^a*

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Fig. S1 XRD patterns of (a-b) Tm-Yb; (c-d) Er-Yb; (e-f) Ho-Yb; (g) Ho/Er/Tm-Yb

doped LMTO phosphors.



Fig. S2 EDS spectrum of LMTO: 0.2%Tm³⁺, 0.05%Er³⁺, 7%Yb³⁺ phosphor.



Fig. S3 (a) FE-SEM image and (b-g) elemental mapping images of the LMTO: 0.2%Tm³⁺, 7%Yb³⁺ phosphor. (h) EDS spectrum of LMTO: 0.2%Tm³⁺, 7%Yb³⁺ phosphor.



Fig. S4 (a) FE-SEM image and (b-g) elemental mapping images of the LMTO: 4%Er³⁺, 5%Yb³⁺ phosphor. (h) EDS spectrum of LMTO: 4%Er³⁺, 5%Yb³⁺ phosphor.



Fig. S5 (a) FE-SEM image and (b-g) elemental mapping images of the LMTO: 1%Ho³⁺, 5%Yb³⁺ phosphor. (h) EDS spectrum of LMTO: 1%Ho³⁺, 5%Yb³⁺ phosphor.



Fig. S6 UC emission spectra of LMTO: x%Tm³⁺, 5Yb³⁺ phosphors. Inset: Emission intensity as a function of the Yb³⁺ contents. λ_{ex} = 980 nm.



Fig. S7 UC emission spectra of LMTO: $x\%Er^{3+}$, $5Yb^{3+}$ phosphors. Inset: Emission intensity as a function of the Yb³⁺ contents. λ_{ex} = 980 nm.



Fig. S8 XRD pattern of LMTO:4%Er³⁺, 6%Yb³⁺ phosphor.



Fig. S9 UC emission spectra of LMTO: x%Ho³⁺, 5%Yb³⁺ phosphors. Inset: Emission intensity as a function of the Yb³⁺ contents. λ_{ex} = 980 nm.



Fig. S10 Raman spectrum of the LMTO host.



Fig. S11 FT-IR spectrum of the LMTO host.



Fig. S12 CIE chromatic coordinates of LMTO:0.2%Tm³⁺, 7%Yb³⁺, LMTO:4%Er³⁺, 5%Yb³⁺ and LMTO:0.2%Tm³⁺, 0.05Er³⁺, 7%Yb³⁺ phosphors.

Table S1. The color purity of LMTO: $1\%Ho^{3+}$, $5\%Yb^{3+}$, LMTO: $4\%Er^{3+}$, $5\%Yb^{3+}$ andLMTO: $0.2\%Tm^{3+}$, $7\%Yb^{3+}$ phosphors.

| Compound | (x, y) | $(\mathbf{x}_i, \mathbf{y}_i)$ | $(\mathbf{x}_d, \mathbf{y}_d)$ | Color purity (%) | |
|---|---------------|--------------------------------|--------------------------------|---------------------|--|
| LMTO: 1%Ho ³⁺ ,5%Yb ³⁺ | (0.276,0.713) | | (0.264,0.725) | 91.8% | |
| LMTO: 4%Er ³⁺ ,5%Yb ³⁺ | (0.314,0.675) | (0.3101,0.3162) | (0.288,0.700) | 93.4% | |
| LMTO:0.2%Tm ³⁺ ,7%Yb ³⁺ | (0.118,0.139) | | (0.095,0.131) | 92.2% | |



Fig. S13 The curve of red-green ratio varying with temperature.



Fig. S14 The PL emission intensity of La_2MgTiO_6 : 0.2%Tm³⁺, 0.05%Er³⁺, 7%Yb³⁺ phosphor versus various temperatures.

| Compounds | Transitions | Range (K) | S _a (% K ⁻¹) | S _r (% K ⁻¹) | Refs |
|---|---|--------------|--|--|------|
| $\begin{tabular}{ c c c c c } \hline Na_2 YMg_2 (VO_4)_3: \\ Er^{3+}/Yb^{3+} \end{tabular}$ | ${}^{2}\text{H}_{11/2}, {}^{4}\text{S}_{3/2} \rightarrow {}^{4}\text{I}_{15/2}$ | 303-573 | 0.77 | 1.104 | 1 |
| $Ba_2SrLu_4O_9{:}Er^{3+}\!/Yb^{3+}$ | ${}^{2}\text{H}_{11/2}, {}^{4}\text{S}_{3/2} \rightarrow {}^{4}\text{I}_{15/2}$ | 303-573 | 0.46 | 0.99 | 2 |
| Na ₃ Gd (VO ₄) ₂ : Er ³⁺ /Yb ³⁺ | ${}^{2}H_{11/2}, {}^{4}S_{3/2} \rightarrow {}^{4}I_{15/2}$ | 291-578 | 0.48 | 0.83 | 3 |
| $La_{2}Ti_{2}O_{7}:$ Ho ³⁺ /Yb ³⁺ | ${}^5F_5/{}^5F_4, {}^5S_2 {\longrightarrow} {}^5I_8$ | 293-473 | 0.32 | 1.41 | 4 |
| TeO ₂ -ZnO-BaO: Ho ³⁺ /Yb ³⁺ | ${}^5\mathrm{F}_{5}\!/{}^5\mathrm{F}_{4,}{}^5\mathrm{S}_2\!\!\rightarrow\!\!{}^5\mathrm{I}_8$ | 303-503 | 0.49 | 0.41 | 5 |
| NaLuF ₄ : Ho ³⁺ /Yb ³⁺ | ${}^{5}F_{1}, {}^{5}G_{6}/{}^{5}F_{2,3}, {}^{3}K_{8} \rightarrow {}^{5}I_{8}$ | 390-780 | 0.14 | 0.83 | 6 |
| YOF: Tm ³⁺ /Yb ³⁺ | ${}^{3}\mathrm{H}_{4(2)}, {}^{3}\mathrm{H}_{4(2)} \rightarrow {}^{3}\mathrm{H}_{6}$ | 190-300 | 0.27 | 0.1207 | 7 |
| Sr_2GdF_7 : Tm^{3+}/Yb^{3+} | ${}^3F_3 \rightarrow {}^3H_6/{}^1G_4 \rightarrow {}^3F_4$ | 293-563 | 3.9 | 1.97 | 8 |
| Bi ₂ SiO ₅ :Tm ³⁺ , Yb ³⁺ @SiO ₂ | $^1\mathrm{G}_4 \rightarrow {}^3\mathrm{F}_4 / {}^3\mathrm{F}_{2,3} \rightarrow {}^3\mathrm{H}_6$ | 280-400 | 1.68 | 1.95 | 9 |
| LMTO:0.2%Tm ³ | LIR 1 _{Tm} | | 4.94 | 1.92 | |
| +, 7%Yb ³⁺ | LIR 2 _{Tm} | | 3.32 | 1.63 | |
| LMTO:4%Er ³⁺ , 5%Yb ³⁺ | LIR 3 _{Er} | 313-573 | 0.68 | 1.13 | T1 · |
| LMTO:1%Ho ³⁺ , 5%Yb ³⁺ | LIR 4 _{Ho} | | 0.18 | 0.58 | work |
| LMTO:0.2%Tm ³⁺ , 0.05%Er ³⁺ , 7%Yb ³⁺ | LIR 2 _{Tm} | | 0.81 | 1.36 | |
| | LIR 3 _{Er} | | 1.47 | 1.09 | |
| | LIR 5 _{Er+Tm} | | 1.06 | 1.21 | |

Table S2. Sensing sensitivities of Ln^{3+} -activated luminescent thermometers.



Fig. S15 Temperature uncertainty δT of Mode I-IV.

| Compounds | LIR | δT_{min} | Repeatability (R) |
|---|------------------------|------------------|-------------------|
| $I MTO(0.20/Tm^{3+}.70/Vh^{3+})$ | LIR 1 _{Tm} | 0.53 | 96.1% |
| LIVITO.0.27011115, 770105 | LIR 2 _{Tm} | 1.02 | 91.4% |
| LMTO:4%Er ³⁺ , 5%Yb ³⁺ | LIR 3 _{Er} | 3.73 | 98.5% |
| LMTO:1%Ho ³⁺ , 5%Yb ³⁺ | LIR 4 _{Ho} | 2.12 | 92.1% |
| | LIR 2 _{Tm} | 1.3 | 95.6% |
| $LM10:0.2\%1 \text{m}^{-3}$, 0.059/Er3+.79/Vb3+ | LIR 3 _{Er} | 2.7 | 96.3% |
| 0.0370E^{13} , $770 \text{Y} \text{D}^{37}$ | LIR 5 _{Er+Tm} | 0.73 | 97% |

Table S3. The δT_{min} and Repeatability (R) values of four temperature measurement modes.

References

- 1. Y. Tong, W. Zhang, R. Wei, L. Chen, H. Guo, Na₂YMg₂(VO₄)₃:Er³⁺,Yb³⁺ phosphors: Up-conversion and optical thermometry, *Ceram. Int.*, 2021, **47**, 2600-2606.
- J. Hu, X. Zhang, H. Zheng, F. Lu, X. Peng, R. Wei, F. Hu, H. Guo, Improved photoluminescence and multimode optical thermometry of Er³⁺/Yb³⁺ co-doped (Ba,Sr)₃Lu₄O₉ phosphors, *Ceram. Int.*, 2022, **48**, 3051-3058.
- K. Saidi, M. Dammak, K. Soler-Carracedo, I.R. Martín, Optical thermometry based on upconversion emissions in Na₃Gd (VO₄)₂: Yb³⁺-Er³⁺/Ho³⁺ micro crystals, *J. Alloys Compd.*, 2022, **891**, 161993.
- Y. Zhao, Z. Su, Y. Huang, Y. Liu, S. Xu, G. Bai, Dual-functional lanthanide ions doped lanthanum titanate microcrystals for simultaneous temperature detection and photothermal conversion, *J. Lumin.*, 2021, 239, 118335.
- A. Doğan, M. Erdem, K. Esmer, G. Eryürek, Upconversion luminescence and temperature sensing characteristics of Ho³⁺/Yb³⁺ co-doped tellurite glasses, *J. Non-Cryst. Solids*, 2021, 571, 121055.
- S. Zhou, S. Jiang, X. Wei, Y. Chen, C. Duan, M. Yin, Optical thermometry based on upconversion luminescence in Yb³⁺/Ho³⁺ co-doped NaLuF₄, *J. Alloys Compd.*, 2014, **588**, 654-657.
- H. Lu, J. Yang, D. Huang, Q. Zou, M. Yang, X. Zhang, Y. Wang, H. Zhu, Ultranarrow NIR bandwidth and temperature sensing of YOF:Yb³⁺/Tm³⁺ phosphor in low temperature range, *J. Lumin.*, 2019, **206**, 613-617.
- W. Chen, J. Cao, F. Hu, R. Wei, L. Chen, H. Guo, Sr₂GdF₇:Tm³⁺/Yb³⁺ glass ceramic: A highly sensitive optical thermometer based on FIR technique, *J. Alloys Compd.*, 2018, **735**, 2544-2550.
- E. Casagrande, M. Back, D. Cristofori, J. Ueda, S. Tanabe, S. Palazzolo, F. Rizzolio, V. Canzonieri, E. Trave,
 P. Riello, Upconversion-mediated Boltzmann thermometry in double-layered Bi₂SiO₅:Yb³⁺, Tm³⁺@SiO₂ hollow nanoparticles, *J. Mater. Chem. C*, 2020, **8**, 7828-7836.