

## Supporting Information

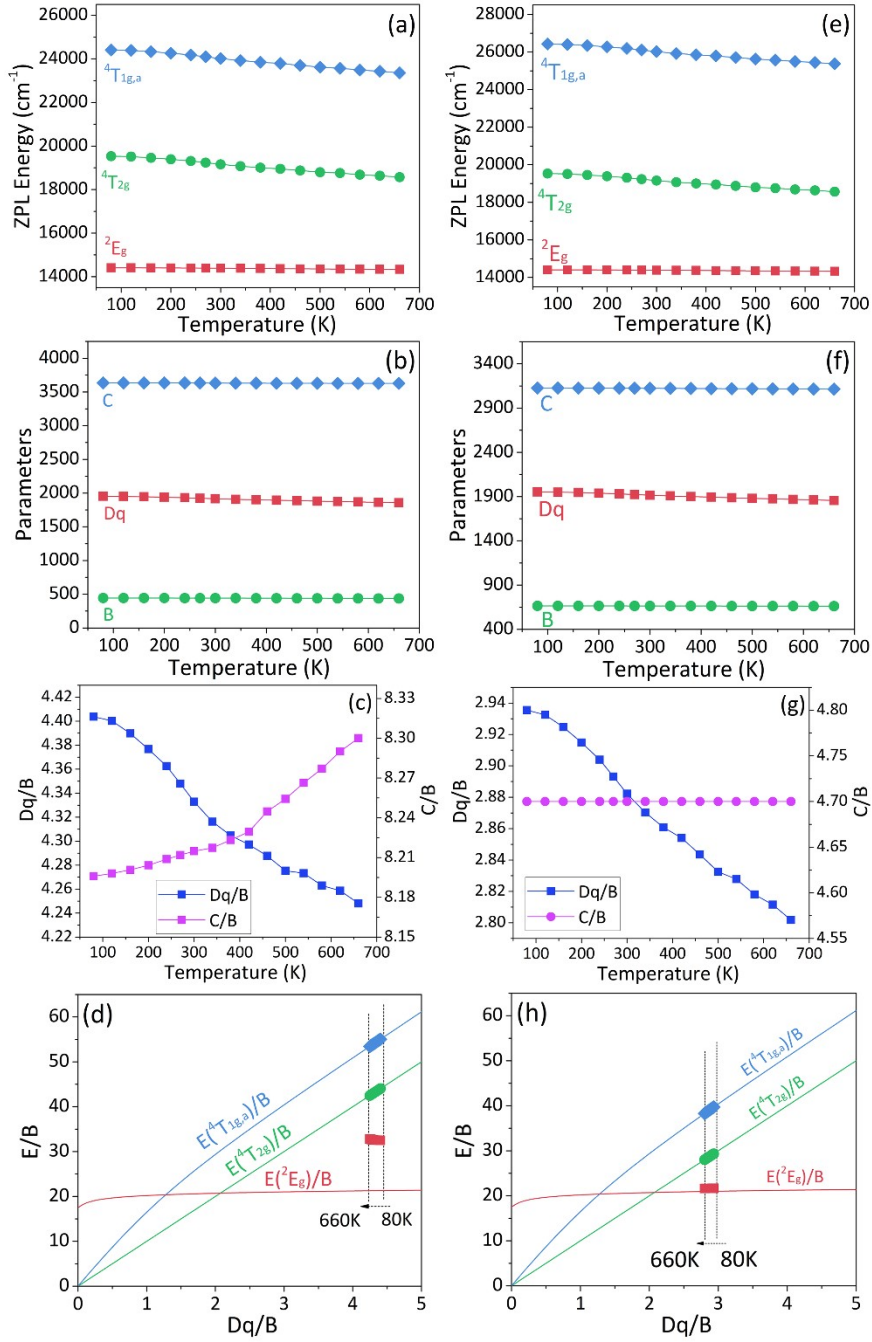
### High-concentration $\text{Mn}^{4+}$ doping in boron-modified $\text{Ca}_{14}\text{Zn}_6\text{Al}_{10}\text{O}_{35}$ -based phosphors: Decoding superior luminescence performances

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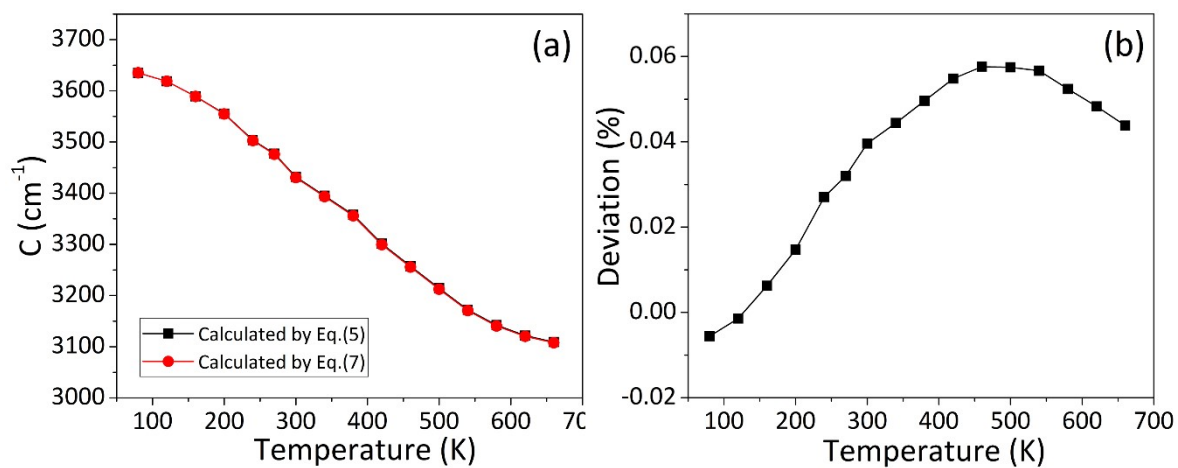
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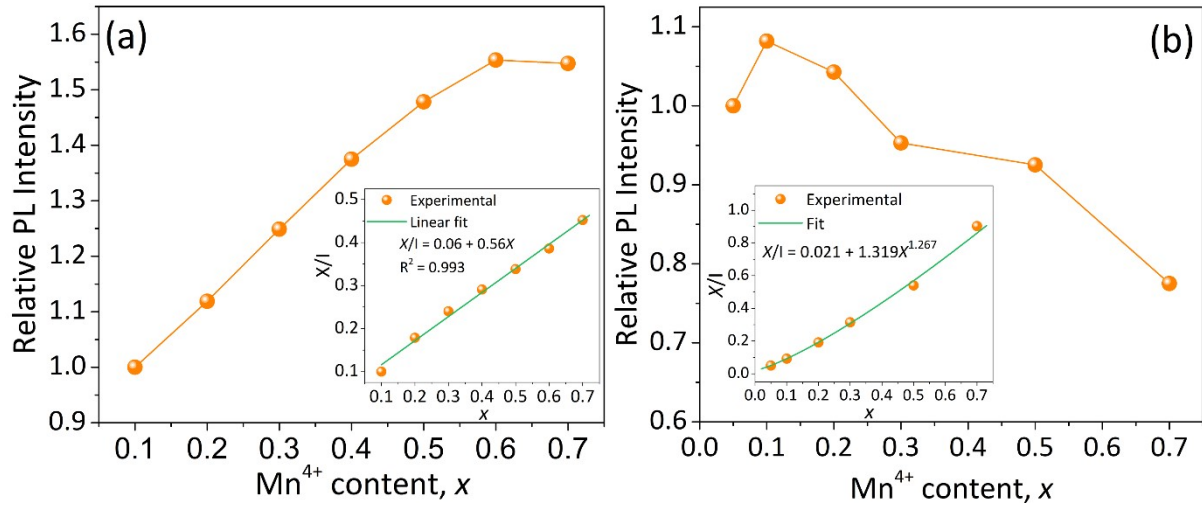
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**Fig. S1** Temperature-dependence of the corrected (a, e)  $E(^2E_g)_{\text{ZPL}}$ ,  $E(^4T_{2g})_{\text{ZPL}}$ , and  $E(^4T_{1g,a})_{\text{ZPL}}$ , (b, f) parameters  $Dq$ ,  $B$ , and  $C$ , (c, g)  $Dq/B$  and  $C/B$ , and (d, h) Excitation/emission energies (divided by  $B$ ) vs.  $Dq/B$  on the Tanabe-Sugano energy diagram. For (a)-(d), the values of  $E(^4T_{2g})_{\text{ZPL}}$  and  $E(^4T_{1g,a})_{\text{ZPL}}$  were corrected based on the redshift of the peak position of the corresponding transition bands, and then the values of  $Dq$ ,  $B$ , and  $C$  were calculated by Eqs.(3)-(6). For (e)-(h), the values of  $E(^4T_{2g})_{\text{ZPL}}$  were corrected based on the redshift of the peak position of the  $^4T_{2g}$  transition band, and then the values of  $E(^4T_{1g,a})_{\text{ZPL}}$ ,  $Dq$ ,  $B$ , and  $C$  were calculated from  $E(^2E_g)_{\text{ZPL}}$  and  $E(^4T_{2g})_{\text{ZPL}}$  by assuming the Racah parameter ratio of  $C/B = 4.7$ , proposed by Adachi (S. Adachi, *ECS J. Solid State Sci. Technol.*, 2020, **9**, 046004).



**Fig. S2** (a) Temperature-dependent Racah parameter  $C$  calculated by formulas (5) and (7), and (b) their divergence  $[(C5 - C7)/C5 \times 100\%]$ , where  $C5$  and  $C7$  are the  $C$  values calculated by formulas (5) and (7), respectively].



**Fig. S3** Dependence of the integrated PL intensity on the Mn-doping concentration for (a) the  $Ca_{14}Zn_6Al_{9.85-2x}B_{0.15}Mn_xMgO_{35}-0.42\% B_2O_3$  phosphors and (b)  $Ca_{14}Zn_6Al_{10-2x}Mn_xMgO_{35}$  phosphors.

For  $Ca_{14}Zn_6Al_{10-2x}Mn_xMgO_{35}$  phosphors, the optimal doping concentration of  $Mn^{4+}$  is 1%. Beyond this concentration, the PL intensity decreases as the doping concentration increases. However, by introducing B doping and using  $B_2O_3$  as a flux, the PL intensity of the  $Ca_{14}Zn_6Al_{9.85-2x}B_{0.15}Mn_xMgO_{35}-0.42\% B_2O_3$  phosphors increases with  $Mn^{4+}$  concentration ( $x$ ), up to  $x = 0.6$ . This observation confirms that B-modification can effectively reduce the crystal defects, thereby suppressing the concentration quenching.