Supporting Information

Phase-Transition-Induced Giant Enhancement of Red Emission in Mn⁴⁺ Doped Fluoride Elpasolite Phosphors

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Figure S1. XRD patterns of trigonal $\text{K}_2\text{LiAlF}_6$ matrixes before and after HF solution treatment.

Figure S2. (a) Structure of $\text{K}_2\text{MnF}_6$, bond lengths and angles of (b) $[\text{MnF}_6]$ octahedrons, (c) $[\text{AlF}_6]$ octahedrons in cubic phase, (d) $[\text{Al}_1\text{F}_6]$ and (e) $[\text{Al}_2\text{F}_6]$ octahedrons in trigonal phase.
Figure S3. The calculated energy levels of Mn$^{4+}$ (vertical bars) and experimental photoluminescence spectra of cubic K$_2$LiAlF$_6$: Mn$^{4+}$.

Figure S4. The calculated energy levels of Mn$^{4+}$ (vertical bars) and experimental photoluminescence spectra of trigonal K$_2$LiAlF$_6$: Mn$^{4+}$. 
Figure S5. PL spectra of K$_2$LiAlF$_6$ : 3 %Mn$^{2+}$ (T-C) and (T-T) phosphors.

Figure S6. Mn concentrations of A-E samples.
Figure S7. (a) XRD patterns, (b) PLE spectra and (c) PL spectra of $\text{K}_2\text{LiAlF}_6: x\%\text{Mn}^{4+}$ ($x = 1, 2, 3, 4$ and $5$) (the insert is dependence of the integrated emission intensity).

Figure S8. (a) XRD patterns, (b) PLE spectra and (c) PL spectra of $\text{K}_2\text{LiAlF}_6: 3\%\text{Mn}^{4+}$ synthesized for different reaction times (the insert is dependence of the integrated emission intensity).
Figure S9. (a) XRD patterns, (b) PL spectra, (c) PL spectra (the insert is dependence of the integrated emission intensity) and (d) decay of K₂LiAlF₆: 3 %Mn⁴⁺ synthesized for different reaction temperatures.

Figure S10. The electron energy loss spectroscopy (EELS) of K₂LiAlF₆: 3 %Mn⁴⁺.
Figure S11. The X-ray photoelectron spectroscopy (XPS) of K$_2$LiAlF$_6$: 3%Mn$^{4+}$.

Table S1. Typical LED photoelectric parameters under different current.

<table>
<thead>
<tr>
<th>Current (mA)</th>
<th>Ra</th>
<th>R9</th>
<th>CCT (K)</th>
<th>Luminescent efficiency (lm/W)</th>
<th>(x, y)</th>
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<tbody>
<tr>
<td>20</td>
<td>86</td>
<td>81</td>
<td>3498</td>
<td>63.3</td>
<td>(0.396,0.367)</td>
</tr>
<tr>
<td>50</td>
<td>85</td>
<td>77</td>
<td>3624</td>
<td>59.9</td>
<td>(0.390,0.362)</td>
</tr>
<tr>
<td>100</td>
<td>85</td>
<td>76</td>
<td>3731</td>
<td>55.7</td>
<td>(0.384,0.355)</td>
</tr>
<tr>
<td>150</td>
<td>85</td>
<td>75</td>
<td>3841</td>
<td>52.1</td>
<td>(0.378,0.349)</td>
</tr>
<tr>
<td>200</td>
<td>84</td>
<td>73</td>
<td>3913</td>
<td>49.7</td>
<td>(0.375,0.347)</td>
</tr>
<tr>
<td>250</td>
<td>84</td>
<td>74</td>
<td>3917</td>
<td>49.6</td>
<td>(0.374,0.345)</td>
</tr>
</tbody>
</table>