Supporting Information

Enhanced Chromaticity and Dual Emission in Double-Site Mn^{4+}-Activated $KLiTiF_6$ Red Phosphor for Wide Color Gamut Displays

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1.Figures

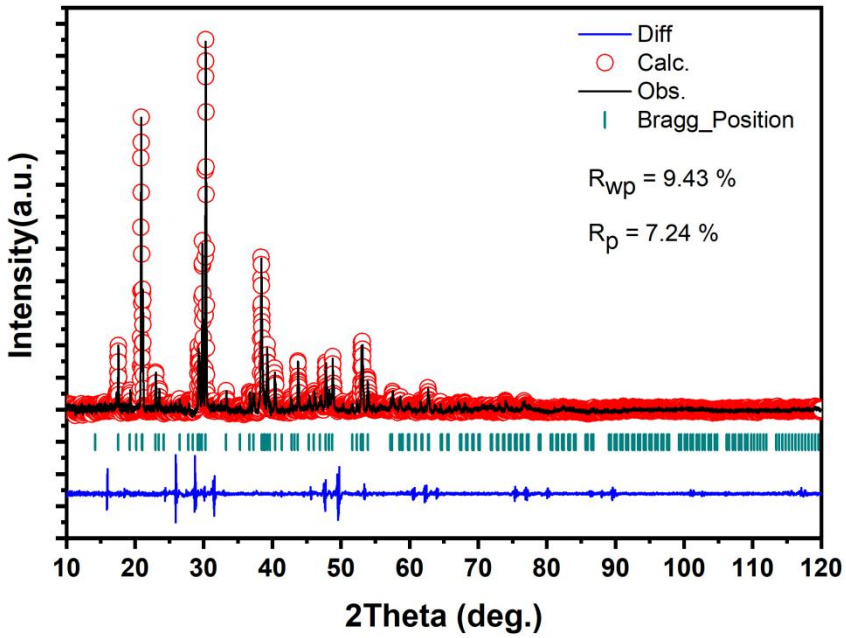

Fig. S1 XRD Rietveld refinement patterns of $KLiTiF_6$:4%Mn^{4+} phosphor.

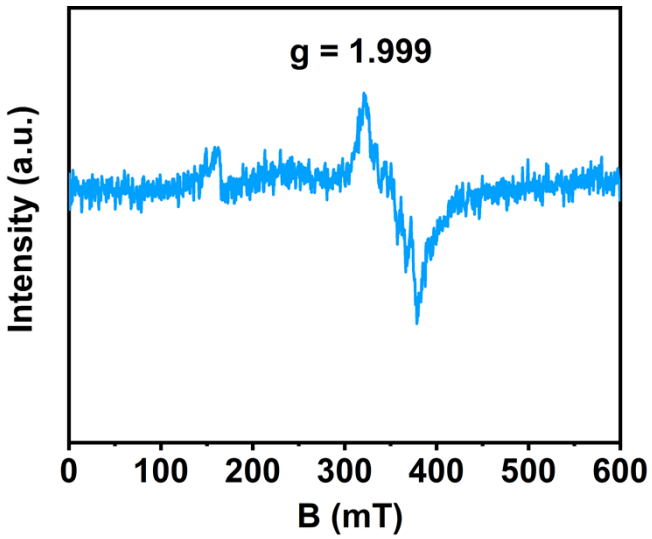

Fig. S2 EPR spectrum of $KLiTiF_6$:4%Mn^{4+}.

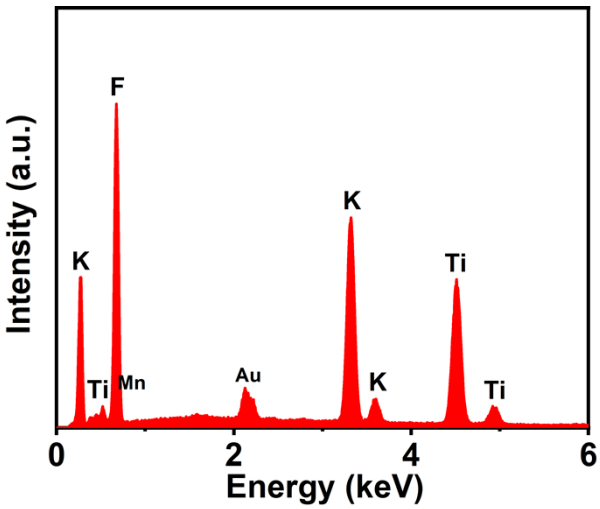

Fig. S3 EDS spectrum of $KLiTiF_6$:4%Mn^{4+}.

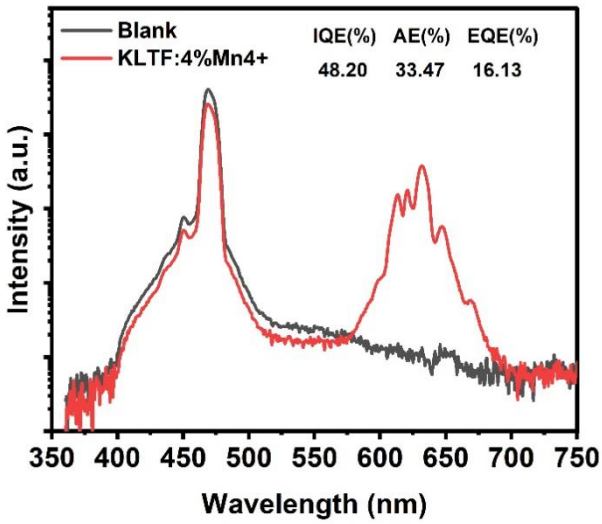

Fig. S4 Internal and external quantum efficiency, absorption efficency of KLTF:4%Mn^{4+} phosphor.

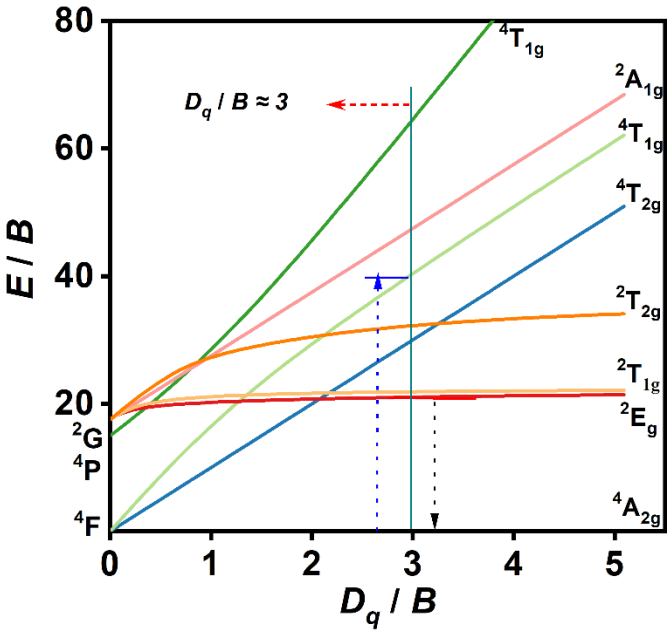

Fig. S5 Tanabe−Sugano energy-level diagram of Mn^{4+} in an octahedral crystal field.

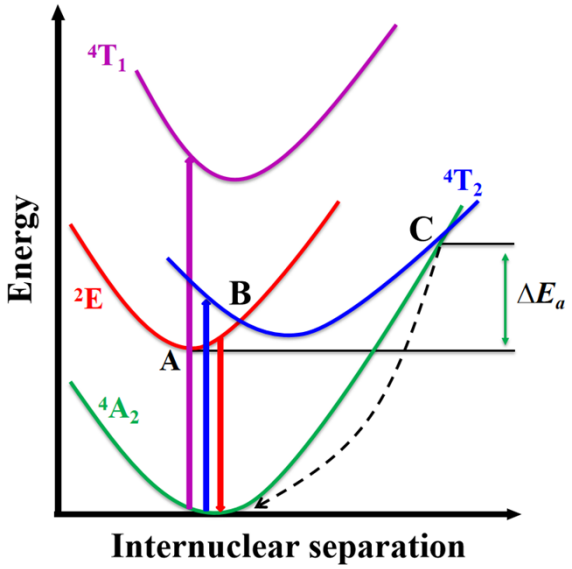

Fig. S6 Thermal quenching mechanism diagram for Mn^{4+} ions.

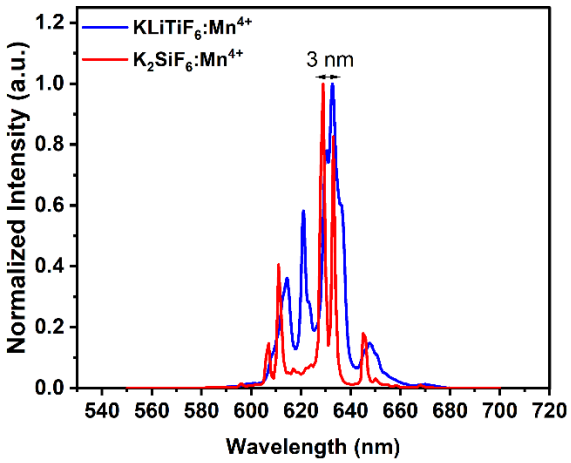

Fig. S7 Spectral comparison with the commercial $K_2SiF_6:Mn^{4+}$ phosphor

2. Tables

Table S1. Crystallographic parameters of KLTF:4%Mn^{4+} phosphor from XRD Rietveld refinement.

Items	Parameters
Space group	*Pca2₁*
Crystal structure	orthorhombic
a (Å)	12.51057
b (Å)	7.60580
c (Å)	10.01616
α = β = γ (°)	90
Volume (Å^3)	955.424
R_{wp} (%)	9.43
R_p (%)	7.24
Refined atomic positions	Mn_{Ti}(0.25086, 0.63854, 0.07226)
	Mn_{Li}(0.24583, 0.64239, 0.43179)
Bond angles(°), the most distorted angle	F–Mn–F: 177.7675
	F–Mn–F: 173.7681

Table S2. Crystal structure and luminescent properties of Mn^{4+}-doped fluoride red phosphors

Series	Host	Crystal Space group	Excitation (nm)	Emission (nm)	ZPL(nm)	PLQY (%)	Refs.
A_2XF_6	$(NH_4)_2SiF_6$	Cubic, O_{h}^{5}-$Fm\bar{3}m$	380, 480	632	weak	64.6*	1
	Na_2SiF_6	Trigonal, D_{3}^{2}-$P321$	355, 460	627	Strong, 618	-	2
	K_2SiF_6	Cubic, O_{h}^{5}-$Fm\bar{3}m$	349, 450	630	weak	80*, 54#	3, 4
	$KNaSiF_6$	Orthorhombic, D_{2h}^{16}-$pnma$	355, 460	629	Strong, 620	90*, 41#	5, 6
	Rb_2SiF_6	Cubic, O_{h}^{5}-$Fm\bar{3}m$	350, 450	630	weak	-	7
	Cs_2SiF_6	Cubic, O_{h}^{5}-$Fm\bar{3}m$	353, 454	633	weak	89*, 71#	8, 9
	$(NH_4)_2GeF_6$	Trigonal, D_{3d}^{3}-$P\bar{3}m1$	365, 468	633	weak	-	1
	Na_2GeF_6	Trigonal, D_{3}^{2}-$P321$	355, 460	627	Strong, 617	-	2, 10
	K_2GeF_6	Trigonal, D_{3d}^{3}-$P\bar{3}m1$	357, 461	631	weak	54#	11
	K_2GeF_6	Hexagonal, C_{6v}^{4}-$P6_3mc$		631	Strong, 621	-	12
	Rb_2GeF_6	Hexagonal, C_{6v}^{4}-$P6_3mc$	360, 460	630	Strong, 620	73*, 57#	13
	Cs_2GeF_6	Cubic, O_{h}^{5}-$Fm\bar{3}m$	352, 456	633	weak	-	8
	$(NH_4)_2SnF_6$	Trigonal, D_{3d}^{3}-$P\bar{3}m1$	365, 468	633	weak	-	1
	Na_2SnF_6	Tetragonal, D_{4h}^{14}-$P4_2/mnm$	360, 470	626	weak	-	14
	$K_2SnF_6{\cdot}H_2O$	Orthorhombic, D_{2h}^{24}-$Fddd$	365, 465	632	Strong, 623	-	15
	$NaKSnF_6$	Orthorhombic, $Pna21(33)$	378, 467	627	Strong, 618	84*	16
	Rb_2SnF_6	Trigonal, D_{3d}^{3}-$P\bar{3}m1$	360, 460	630	weak	70.3*	17

	Cs_2SnF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	370, 470	633	weak	-	[14]
	$(NH_4)_2TiF_6$	Trigonal, D_{3d}^3-$P\bar{3}m1$	380, 480	632	weak	16.4*	[1, 18]
	Na_2TiF_6	Trigonal, D_3^2-$P321$	355, 460	627	Strong, 617	-	[10]
	K_2TiF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	355, 456	631	weak	98*	[19]
	Rb_2TiF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	468	630	weak	91*	[20-22]
	Cs_2TiF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	360, 471	632	weak	-	[23]
	Li_2ZrF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	376, 463	631	weak	-	[24]
	Rb_2ZrF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	360, 474	629	weak	75*	[25]
	Cs_2ZrF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	364, 475	631	weak	56.9*	[26]
	Rb_2HfF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	360, 465	630	weak	55.6*	[27]
	Cs_2HfF_6	Trigonal, D_{3d}^3-$P\bar{3}m1$	379, 477	632	weak	90*	[27, 28]
BXF_6	$BaSiF_6$	Trigonal, D_{3d}^5-$R\bar{3}m$	365, 466	632	weak	-	[29-32]
	$ZnSiF_6{\cdot}6H_2O$	Trigonal, C_{3i}^2-$R\bar{3}$	360, 460	630	weak	-	[33-35]
	$BaGeF_6$	Trigonal, D_{3d}^5-$R\bar{3}m$	358, 460	634	weak	-	[36, 37]
	$ZnGeF_6{\cdot}6H_2O$	Trigonal, C_{3i}^2-$R\bar{3}$	370, 470	630	weak	-	[33, 38]
	$BaSnF_6$	Trigonal, C_{3i}^2-$R\bar{3}$	368, 467	631	weak	-	[39, 40]
	$ZnSnF_6{\cdot}6H_2O$	Trigonal, C_{3i}^2-$R\bar{3}$	370, 480	629	weak	-	[41]
	$BaTiF_6$	Trigonal, D_{3d}^5-$R\bar{3}m$	356, 466	634	weak	73.2*	[42, 43]
	$ZnTiF_6{\cdot}6H_2O$	Trigonal, C_{3i}^2-$R\bar{3}$	360, 465	631	weak	26*, 6#	[44, 45]
$A_2A'MF_6$	Na_3AlF_6	Monoclinic, C_{2h}^5-$P2_1/c$	356, 465	627	Strong, 620	-	[46]
	K_2LiAlF_6	Cubic, O_h^5-$Fm\bar{3}m$	360, 468	635	Strong, 625	-	[47]

	K_3AlF_6	Tetragonal, C_{4h}^6-$I4_1/a$	360, 460	626	Strong, 618	88*, 51#	48
	K_2NaAlF_6	Cubic, O_h^5-$Fm\bar{3}m$	464	630	Strong, 624	58.4*	49, 50
	Cs_2KAlF_6	Cubic, O_h^5-$Fm\bar{3}m$	469	632	weak	50.6*	51
	Cs_3AlF_6	Cubic, O_h^5-$Fm\bar{3}m$	361, 467	634	weak	48.2*	52
	Na_3GaF_6	Monoclinic, C_{2h}^5-$P2_1/c$	360, 467	627	Strong, 620	69*	53
	K_2LiGaF_6	Cubic, O_h^5-$Fm\bar{3}m$	363, 467	635	Strong, 626	20*	54
	K_3GaF_6	Tetragonal, C_{4h}^6-$I4_1/a$	467	626	Strong, 618	46*	55
	K_2NaGaF_6	Cubic, O_h^5-$Fm\bar{3}m$	365, 467	629	Strong, 621	61*	56
A_2NF_7	K_2TaF_7	Monoclinic, C_{2h}^5-$P2_1/c$	362, 461	627	Strong, 619	99.6*	57
	K_2NbF_7	Monoclinic, C_{2h}^5-$P2_1/c$	369, 467	628	Strong, 620	71.3*	57, 58
$A_2NO_xF_{6-x}$	Cs_2NbOF_5	Trigonal, *P3*(No.143)	371, 474	633	weak	63.4*	59, 60
	Rb_2NbOF_5	Trigonal, D_{3d}^3-$P\bar{3}m1$	630, 465	631	weak	68*, 19#	61
	$Na_2WO_2F_4$	Orthorhombic, *Pbcn*(60)	367, 469	619	超强, 619	76.6*	62-64
	$Cs_2WO_2F_4$	Trigonal, D_{3d}^3-$P\bar{3}m1$	374, 470	632	weak	-	65
$A_2A'XF_7$	K_3SiF_7	P4/mbm	360, 460	631	weak	-	66
	Rb_3SiF_7	P4/mbm	360, 460	631	weak	-	67
	$RbCs_2SiF_7$	P4/mbm	360, 460	631	weak	-	67
Organic-Inorginic Compounds	$[C(NH_2)_3]AlF_6$	$P\bar{a3}$	358, 462	630	Strong	27.8*, 6.4#	68
	$[N(CH_3)_4]_2GeF_6$	$R\bar{3}$	367, 470	630	weak	92.2*,64.6#	69
	$[N(CH_3)_4]_2TiF_6$	$R\bar{3}$	367, 470	630	weak	83.3*,54.5#	69
	$[N(CH_3)_4]_2ZrF_6$	$R\bar{3}$	367, 470	630	weak	79.2*,50.7#	69
	$KLiTiF_6$	**orthorhombic *Pca*2$_1$**	**371, 469**	**632**	**Strong,612.0, 613.7**	**48.2*,16.1#**	**This work**

* indicates internal quantum efficiency, # indicates external quantum efficiency, and blank indicates no reference data.

Table. S3. The formation energy of Mn^{4+} doping in the $KLiTiF_6$ matrix under different charge compensation mechanisms.

Models	Charge compensation for Mn^{4+} doping	Formation energy (eV)
M1	$Mn_{Ti}^{\times} + V_{Ti}^{\times}$	5.12
M2	$Mn_{Li}^{\cdots} + Li_{Ti}^{\cdot} + V_{Ti}^{\times}$	10.13
M3	$Mn_{Li}^{\cdots} + 2V_{K}^{'} + F_{i}^{\cdot}$	12.68
M4	$Mn_{Li}^{\cdots} + 2V_{Li}^{'} + F_{i}^{\cdot}$	14.46
M5	$Mn_{Li}^{\cdots} + 3V_{Li}^{'}$	16.58
M6	$Mn_{Li}^{\cdots} + 3V_{K}^{'}$	17.43

Table. S4. The structure parameters of M1 and M2 charge compensation mechanisms.

Structure parameters	M1	M2
F1-Mn-F2	176.5767°	/
F3-Mn-F6	179.4912°	/
F4-Mn-F5	178.7213°	/
F7-Mn-F8	/	178.1263°
F9-Mn-F12	/	172.8671°
F10-Mn-F11	/	179.4115(0)
Average bond length	1.8359 Å	1.8312 Å
Polyhedral volume	8.2426 $Å^3$	8.1632 $Å^3$
Distortion index (bond length)	0.00457	0.01271
Quadratic elongation	1.0007	1.0022
Bond angle variance	2.2677 $deg.^2$	6.6138 $deg.^2$
Effective coordination number	5.9947	5.9492

Table. S5. Color coordinate offset for KLTF:4%Mn^{4+} under different temperatures

Temperature (K)	CIE coordinates (x, y)	*ΔE*
300	0.695,0.305	0
320	0.694,0.306	0.00285
340	0.693,0.307	0.00568
360	0.692,0.308	0.00849
380	0.690,0.310	0.01408
400	0.687,0.313	0.02236
420	0.681,0.319	0.03852
440	0.673,0.327	0.05932

Table. S6.

Phosphors	Chromaticity coordinate of phosphor	Chromaticity coordinate of white LED	CCT (K)	LE (lm/W)	color gamut of NTSC (%)	Refs.
$Cs_2KInF_6:Mn^{4+}$	(0.696, 0.304)	(0.3059, 0.3191)	7001	-	101.7	[70]
$Cs_2KScF_6:Mn^{4+}$	(0.6933, 0.3064)	(0.3009, 0.3144)	6088	68.21	107.9	[71]
$Cs_2KGaF_6:Mn^{4+}$	-	(0.2811, 0.3107)	8951	106.3	114.8	[72]
$Rb_2MoO_2F_4:Mn^{4+}$	-	(0.2801, 0.2864)	10172	82.75	101.6	[73]
$Cs_2NaAlF_6:Mn^{4+}$	(0.7032, 0.2967)	(0.7032, 0.2967)	7466	87.7	107.8	[74]
$(Me4N)_2BF_6:Mn^{4+}$	-	(0.2886, 0.3140)	8262	143.09	112.0	[69]
$Cs_2NaGaF_6:Mn^{4+}$	(0.7027, 0.2968)	(0.2966, 0.2995)	7274	97.29	110.64	[75]
$Cs_2NaGaF_6:Mn^{4+}$, Li+	(0.7029, 0.2970)	(0.2742, 0.3460)	7312	110.57	110.68	[75]
K_2NaScF_6: Mn^{4+}	(0.6941, 0.3057)	(0.2883, 0.2996)	8841	73.7	105.6	[76]
K_2NaScF_6: Mn^{4+}	-	(0.2883, 0.2996)	8841	73.7	105.6	[77]
Rb_2NaScF_6: Mn^{4+}	-	(0.2891, 0.3056)	8458	80.7	108.4	[77]
Cs_2NaScF_6: Mn^{4+}	-	(0.2852, 0.3159)	8470	77.0	105.6	[77]
$Cs_2MoO_2F_4:Mn^{4+}$	(0.6958, 0.3041)	(0.2948, 0.3104)	7896	114.7	109.1	[78]
$BaTaF_7:Mn^{4+}$	-	(0.31, 0.33)	6693	22.06	113.4	[79]
$[C(NH_2)_3]AlF_6:Mn^{4+}$	-	(0.2929, 0.3050)	8172	45.96	108.3	[68]
$(Me_4N)_2SnF_6:Mn^{4+}$	-	(0.3015, 0.3063)	7489	122.42	124	[80]
$Rb_2NaGaF_6:Mn^{4+}$	-	(0.2928, 0.3264)	7703	59.43	108.6	[81]
$KLiTiF_6:Mn^{4+}$		(0.272, 0.311)	9961	91.6	116.1	This Work

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