Supporting Information

Dual-Emissive Luminescence in OIHMH Single Crystals: Tunable Red-Green Emissions via Mn^{2+} Doping and Theoretical Insights

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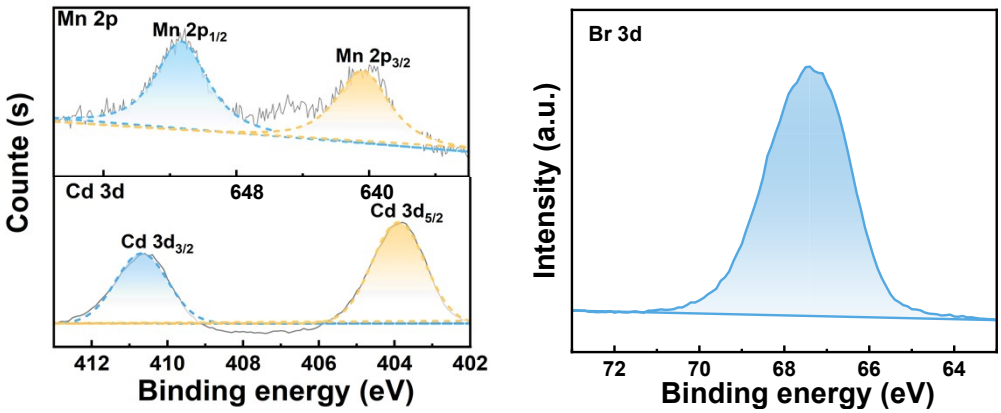

Fig. S1 High-resolution XPS full spectra of Mn 2p, Cd 3d, Br 3d, for $C_6H_{14}N_2CdBr_4$:Mn^{2+}.

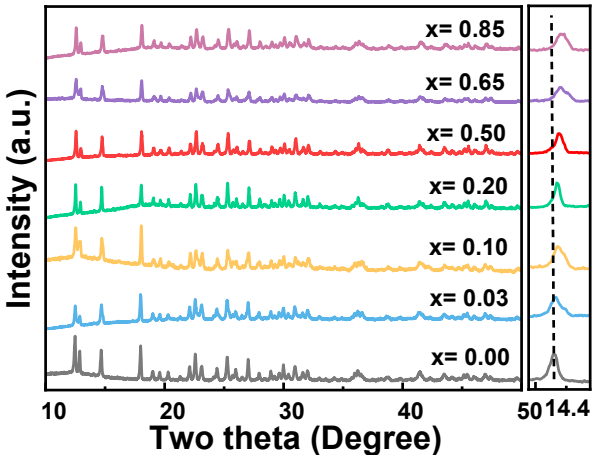

Fig. S2 The XRD patterns of $C_6H_{14}N_2Cd_{1-x}Mn_xBr_4$ with various Mn^{2+} ions concentrations.

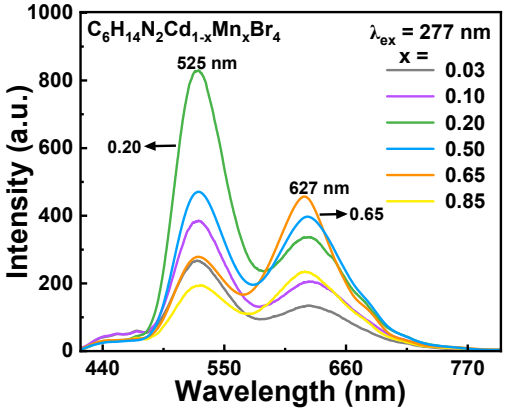

Fig. S3 PL spectra of $C_6H_{14}N_2Cd_{1-x}Mn_xBr_4$ with various x values under 277 nm excitation.

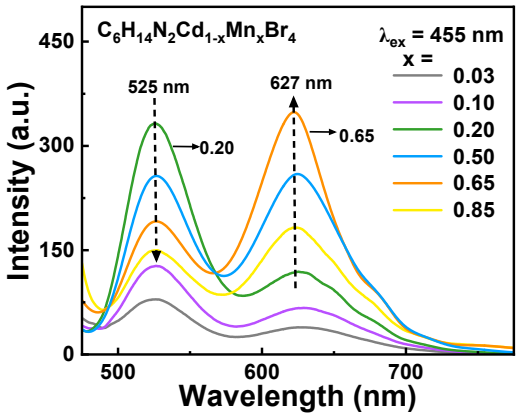

Fig. S4 PL spectra of $C_6H_{14}N_2Cd_{1-x}Mn_xBr_4$ with various x values under 455 nm excitation.

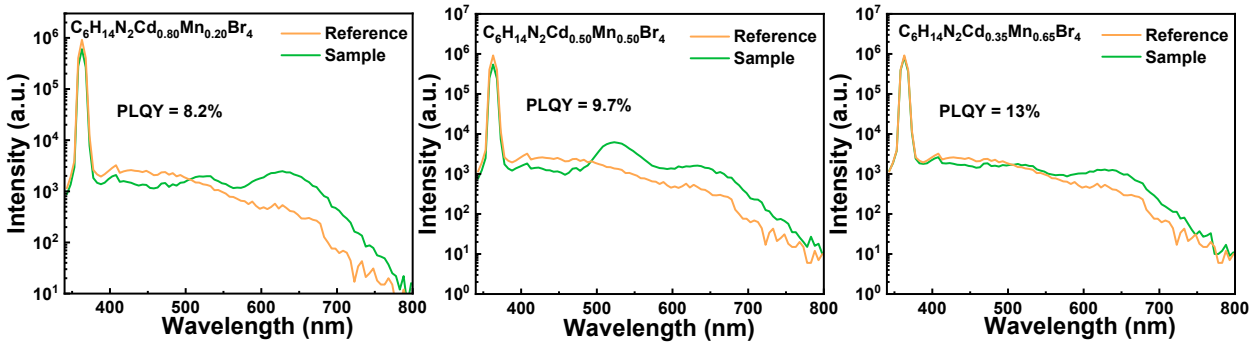

Fig. S5 PLQY measurement of $C_6H_{14}N_2Cd_{1-x}Mn_xBr_4$ crystals in an integrated sphere with 363 nm excitation source.

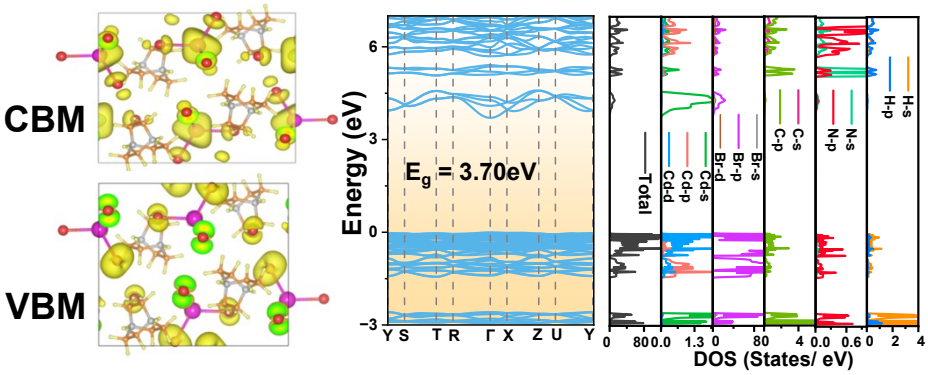

Fig. S6 Charge density distribution of the conduction band minimum (CBM) and the valence band maximum (VBM), and calculated electronic band structure and partial density of states of $C_6H_{14}N_2CdMnBr_4$.

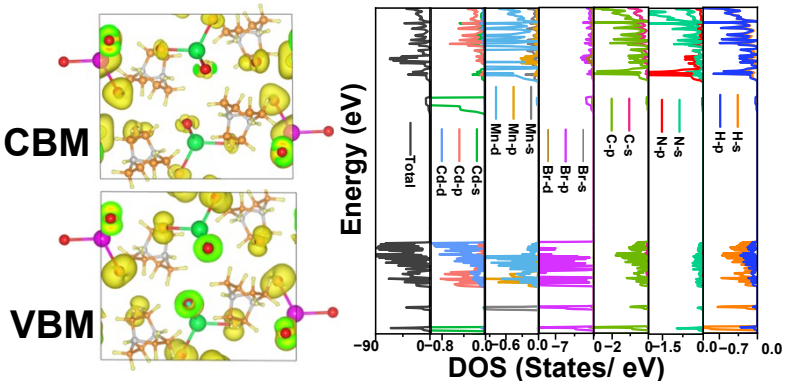

Fig. S7 Charge density distribution of the CBM and VBM and partial density of states

(spin-down) of $C_6H_{14}N_2Cd_{0.50}Mn_{0.50}Br_4$ with Mn^{2+}-Mn^{2+} distance at 6.90 Å.

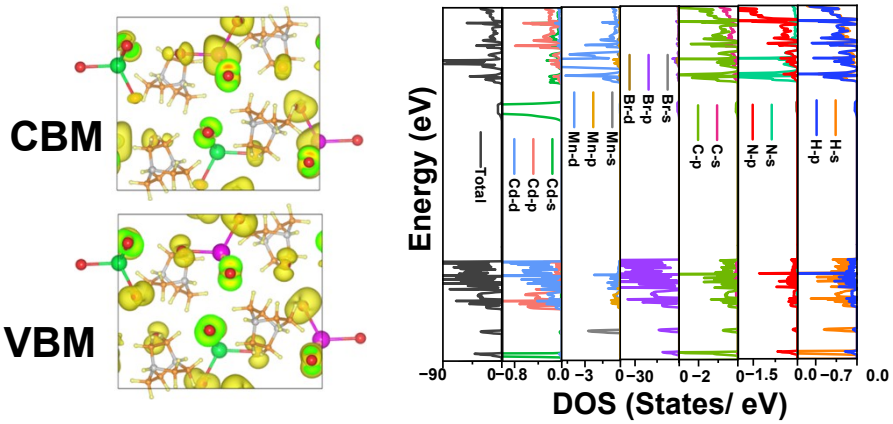

Fig. S8 Charge density distribution of the CBM and VBM and partial density of states (spin-down) of $C_6H_{14}N_2Cd_{0.50}Mn_{0.50}Br_4$ with Mn^{2+}-Mn^{2+} distance at 9.87 Å.

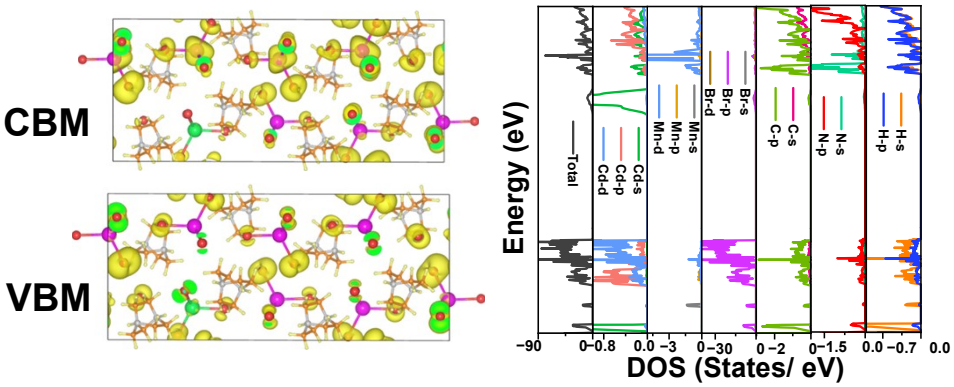

Fig. S9 Charge density distribution of the CBM and VBM and partial density of states (spin-down) of $C_6H_{14}N_2Cd_{0.875}Mn_{0.125}Br_4$.

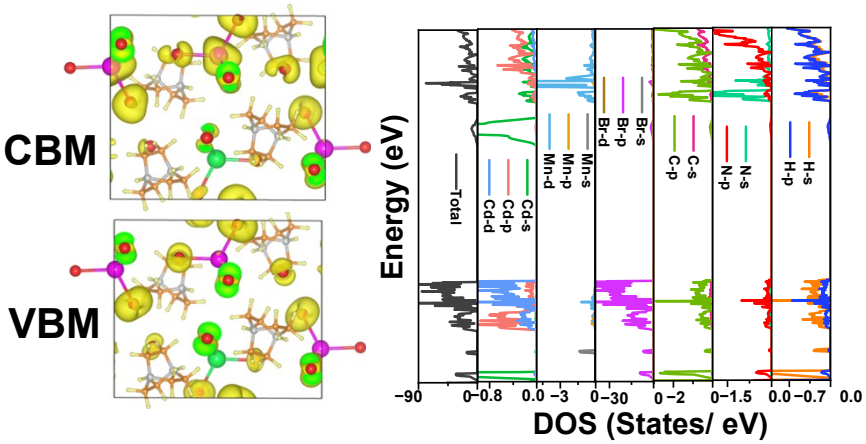

Fig. S10 Charge density distribution of the CBM and VBM and partial density of states (spin-down) of $C_6H_{14}N_2Cd_{0.75}Mn_{0.25}Br_4$.

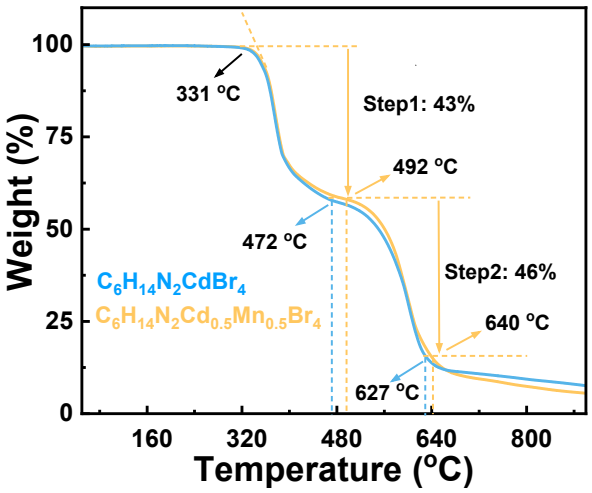

Fig. S11 Thermal gravimetric analysis (TGA) curves measured from room temperature to 900 °C at the heating rate of 10 °C min^{-1} on a synchronous thermal analyzer (TGA/DSC, Mettler, Switzerland).

Table S1 Summary of the fitting parameters of time-resolved PL decay curves, the calculated average lifetimes (τ_{ave}) at 525 nm.

$C_6H_{14}N_2Cd_{1-x}Mn_xBr_4$	A_1	τ_1(ms)	A_2	τ_2(ms)	τ_{ave}(ms)
X = 0.03	11109.9	0.00104	5987.14	0.13354	9.24
X = 0.10	6341.4	0.00114	4020.77	0.12639	8.89
X = 0.20	10387.2	0.00102	4986.43	0.12527	7.39
X = 0.50	15869.4	0.00118	5625.36	0.13760	5.69
X = 0.65	18649.9	0.00107	5360.65	0.13511	4.90

Table S2 Summary of the fitting parameters of time-resolved PL decay curves, the calculated average lifetimes (τ_{ave}) at 627 nm.

$C_6H_{14}N_2Cd_{1-x}Mn_xBr_4$	A_1	τ_1(ms)	A_2	τ_2(ms)	τ_{ave}(ms)
X = 0.03	28382	0.00146	14209	0.11171	4.28
X = 0.10	23051.9	0.00122	9580.7	0.10738	3.92
X = 0.20	48518	0.00165	18562	0.11550	3.09

X = 0.50	38429	0.00156	13250	0.11450	2.89
X = 0.65	37925	0.00138	6559.8	0.11323	1.61