Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information for Mechanistic Insights on Regulating the Site Occupancy, Valence States and Optical Transitions of Mn Ions in Yttrium-Aluminum Garnet via Codoping

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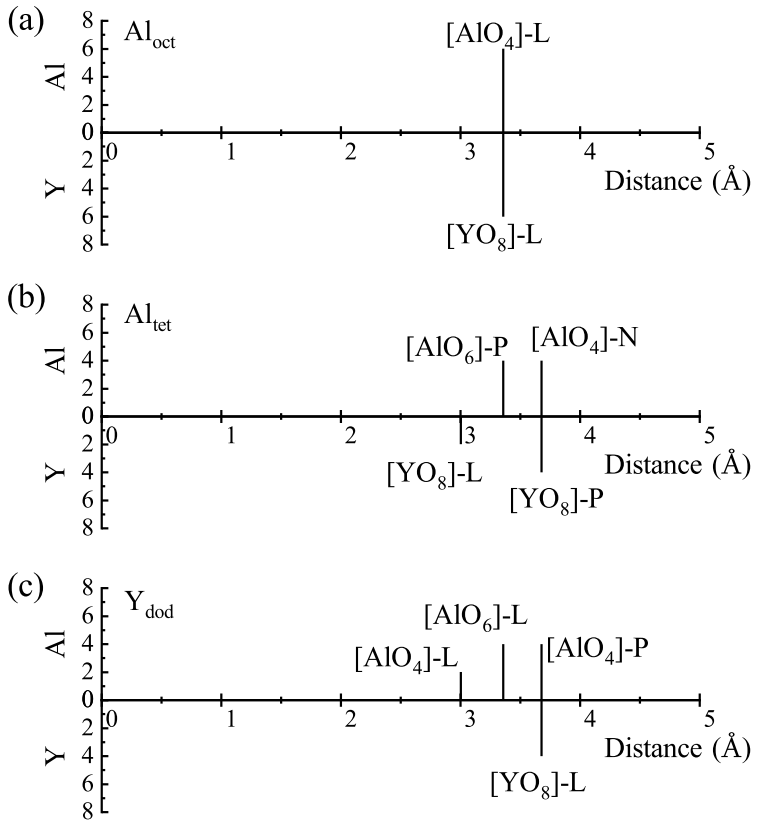

Figure S1: The numbers of near Al/Y ions of Al_{oct} (a), Al_{tet} (b) and Y_{dod} (c) ions as the function of distance. The near Al/Y–O uints and Al_{oct}–O (a), Al_{tet}–O (b) and Y_{dod}–O (c) units are connected by the line (L), point (P) and no (N) sharing mode with two, one and no sharing oxygen ions, respectively.

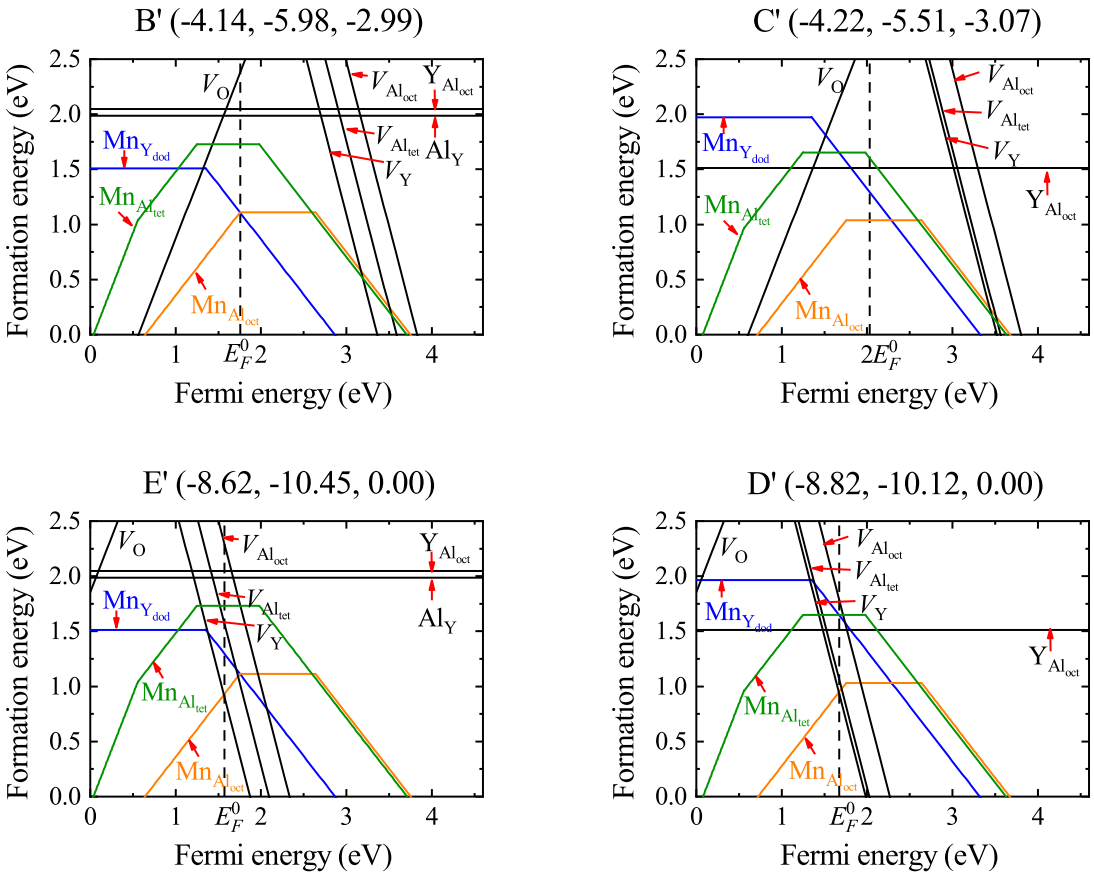

Figure S2: The formation energies of Mn dopant and intrinsic defects as a function of Fermi energy in the critical chemical potentials ($\Delta\mu_{\mathrm{Al}}$, $\Delta\mu_{\mathrm{Y}}$, $\Delta\mu_{\mathrm{O}}$) conditions of the B′, C′, D′ and E′ points in Figure 1 of main text. The vertical dashed lines are the equilibrium Fermi energies E_F^0.

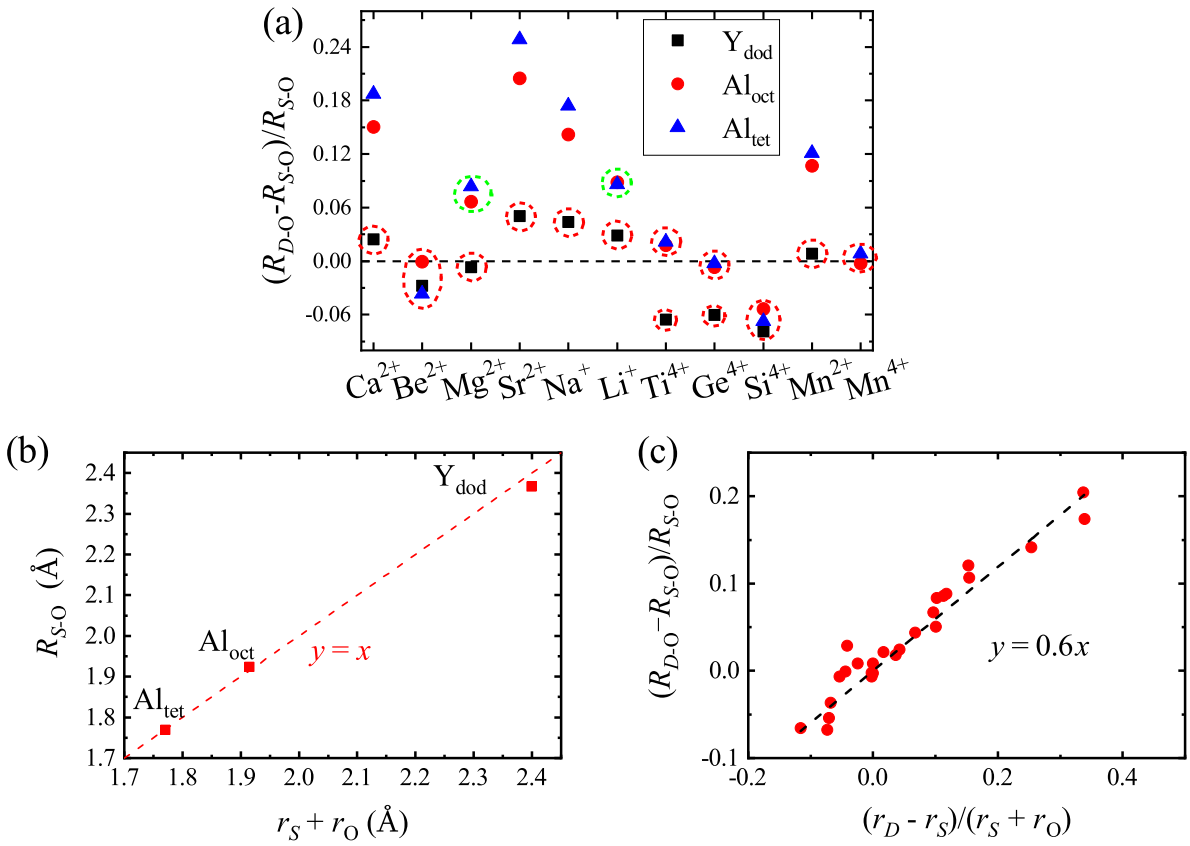

Figure S3: (a) The relative changes of calculated bond lengths with various cation substitutions, where R_{D-O} and R_{S-O} are the average bond length of the dopant and substituted ions with coordinated oxygen ions, respectively. Black square, red circle and blue triangle represent that substituted ions are Y_{dod}, Al_{oct} and Al_{tet} ions, respectively. Red dotted circles are the cation substitutions of low defect formation energies. (b) The calculated bond length R_{S-O} *vs* the crystal ionic radii determined bond length $r_S + r_O$ of the substituted ions (Al_{tet}, Al_{oct} and Y_{dod}) in YAG host. (c) The comparison of the relative changes of calculated bond lengths $(R_{D-O} - R_{S-O})/R_{S-O}$ *vs* the relative mismatch of the crystal ionic radii $(r_D - r_S)/(r_S + r_O)$ with various cation substitutions.