

## **Supporting Information**

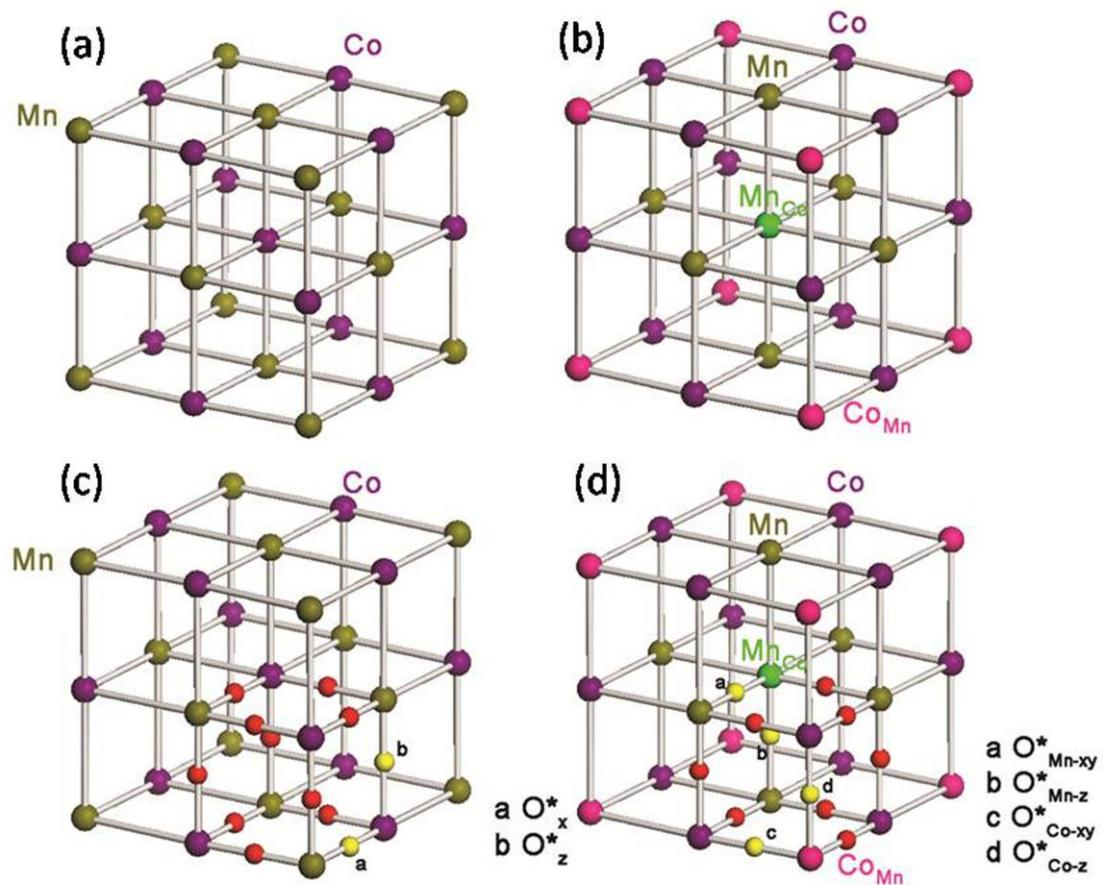
### **Synergistic Effects of Intrinsic Cation Disorder and Electron-Deficient Substitution on Ion and Electron Conductivity in $\text{La}_{1-x}\text{Sr}_x\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$ ( $x = 0, 0.5$ , and $0.75$ )**

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Defeng Zhou,<sup>§</sup> Fanzhi Meng,<sup>\*,†</sup> Jian Meng<sup>†</sup>

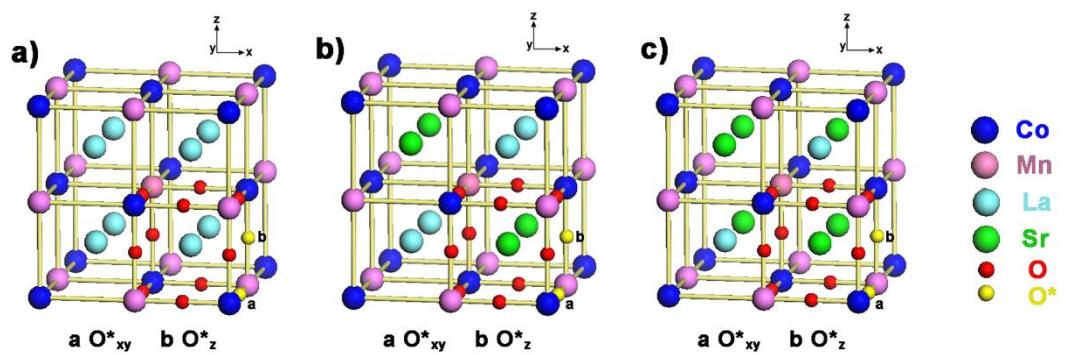
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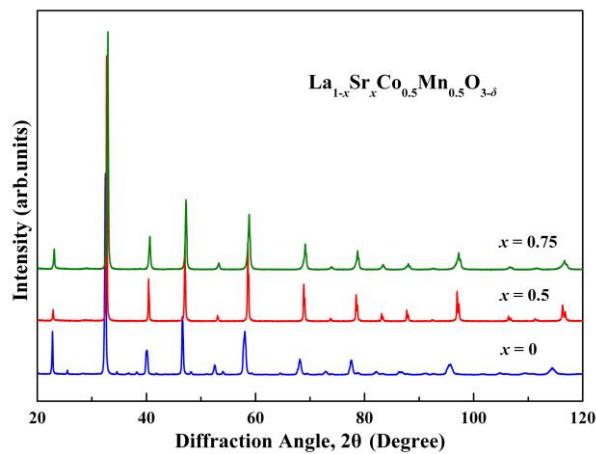
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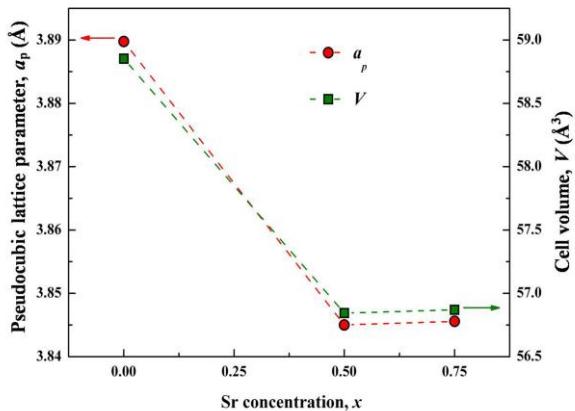
**Figure S1.** (a) Perfect LCMO supercell; (b) LCMO with antisite defects (AD: LCMO); (c) Oxygen vacancies considered in perfect LCMO supercell; (d) Oxygen vacancies consider in AD: LCMO. Different atoms are represented in different color as labeled in the figure.



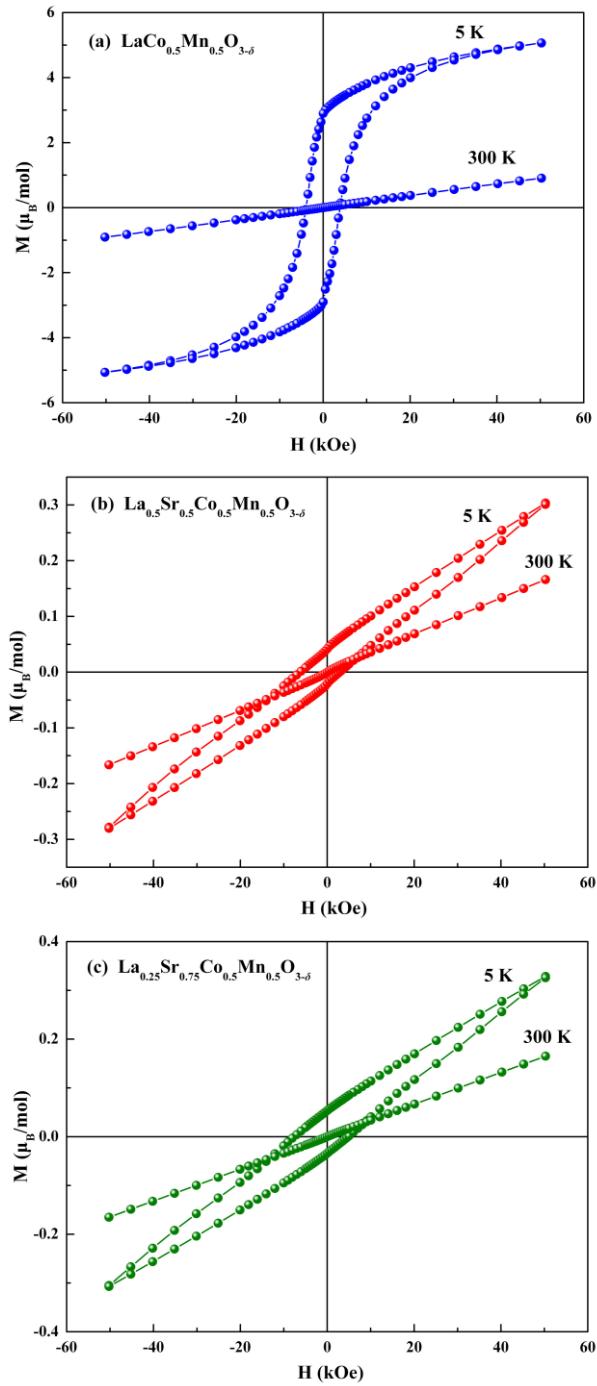
**Figure S2.** (a) Oxygen vacancies in LCMO ( $O^*$ : LCMO); (b) oxygen vacancies in  $L_{0.5}S_{0.5}\text{CMO}$  ( $O^*$ :  $L_{0.5}S_{0.5}\text{CMO}$ ); (c) oxygen vacancies in  $L_{0.25}S_{0.75}\text{CMO}$  ( $O^*$ :  $L_{0.25}S_{0.75}\text{CMO}$ ).



**Figure S3.** Powder X-ray diffraction patterns of the  $\text{La}_{1-x}\text{Sr}_x\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$  ( $x = 0, 0.5$ , and 0.75) series at room temperature.



**Figure S4.** The pseudocubic lattice parameter and cell volume for  $\text{La}_{1-x}\text{Sr}_x\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$  ( $x = 0$ ,  $0.5$ , and  $0.75$ ) obtained from Rietveld Refinement of powder X-ray diffraction data.



**Figure S5.** The M–H hysteresis loops of  $\text{La}_{1-x}\text{Sr}_x\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$  ( $x = 0, 0.5$ , and  $0.75$ )

at  $T = 5$  K and  $300$  K over the magnetic field range from  $-5$  T to  $5$  T.

**Table S1.** Bond lengths and angles for  $\text{La}_{1-x}\text{Sr}_x\text{Co}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$  ( $x = 0$ , 0.5, and 0.75)

obtained from Rietveld refinement of powder X-ray diffraction data.

Doping content	$x = 0$	$x = 0.5$	$x = 0.75$
<b>Space group</b>	<i>Pnma</i> (No. 62)	<i>P4/mmm</i> (No. 123)	<i>Pnma</i> (No. 62)
<b>Co(Mn)-O1 ×2 (Å)</b>	2.024(4)	1.928(6)	1.937(13)
<b>Co(Mn)-O2 ×2(Å)</b>	1.911(12)	1.922(3)	1.951(29)
<b>Co(Mn)-O2 ×2 (Å)</b>	2.027(11)	—	1.964(28)
<b>Co(Mn)-O3 ×2(Å)</b>	—	1.917(6)	—
<b>Co(Mn)-O1-Co(Mn)(deg)</b>	147.5(7)	180	164.5(6)
<b>Co(Mn)-O2-Co(Mn) (deg)</b>	162.74(2)	179.674(2)	158.85(31)
<b>Co(Mn)-O3-Co(Mn) (deg)</b>	—	179.960(0)	—