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TEM Study of Two-Dimensional Incommensurate Modulation in $\text{La}(2-2x)\text{Ca}(1+2x)\text{Mn}_2\text{O}_7$ (0.6;x;0.8) LEONID A. BENDERSKY, Metallurgy Division, NIST 100 Bureau Drive, Stop 8554 Gaithersburg, MD 20899-8554, IAN D. FAWCETT, MARTHA GREENBLATT, Department of Chemistry and Chemical Biology, Rutgers, The State University of New Jersey, Piscataway, NJ 08854 — Ruddlesden-Popper $n = 2$ compounds $\text{La}_{2-2x}\text{Ca}_{1+2x}\text{Mn}_2\text{O}_7$ with $0.6 < x < 0.8$ were synthesized by a citrate gel technique. Electron diffraction identified the presence of two-dimensional incommensurate modulations. The modulation is sensitive to electron-beam irradiation and rapidly degrades under a focused beam. In-situ heating experiments indicated the existence of possible tetragonal-to-incommensurate phase transition around 350°C . High-resolution imaging proves that the modulation in the real space is truly two-dimensional and incommensurate. Such modulation has never been observed before for La-Ca-Mn-O or any other perovskite system. The best approximation of the incommensurate superstructure is by a commensurate lattice $\mathbf{a}_x' = \mathbf{a}_{xt} + 2\mathbf{a}_{yt}$; $\mathbf{a}_y' = -2\mathbf{a}_{xt} + \mathbf{a}_{yt}$; $\mathbf{c}' = \mathbf{c}_t$ (t-tetragonal I4/mmm). Such superstructure could be satisfied with a 4:1 two-dimensional ordering in a (001) plane. For the studied compositions, such 4:1 ordering is plausibly described by charge ordering between Mn^{4+} and Mn^{3+} ions. Structural models of the charge ordering between Mn^{4+} and Mn^{3+} ions are suggested.

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