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**Exotic Bilayer Manganite Phase Diagram near x=0.6 Hole Doping.**

KENNETH GRAY, HONG ZHENG, QING’AN LI, JOHN F. MITCHELL, STINE NYBORG ANCONA, STEPHAN ROSENKRANZ, RAY OSBORN, Argonne National Laboratory — Important modifications to the phase diagram of the bilayered manganites have been discovered through combined measurements of conductivity, magnetization and neutron and x-ray diffraction. The previously reported phase diagram for the CE-type orbital and charge order in LaSr$_2$Mn$_2$O$_7$ crystals (near hole doping of x=0.5) is qualitatively reproduced and graphically demonstrated for the bi-stripe (BIS) orbital and charge order in La$_{0.8}$Sr$_{2.2}$Mn$_2$O$_7$ crystals near x=0.6. Individual crystals, taken along the compositional gradient of a single boule made by the floating-zone technique with nominal composition x=0.61, display a large fraction of the phase diagram. As in the case of x=0.5, we find (1) crystals that enter the BIS phase at 240-300 K and reenter into an in-plane metal, A-type antiferromagnetic phase, and (2) crystals that retain BIS order down to at least 5 K. Surprisingly, the BIS state seems somewhat more stable than the CE state.

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