Coupling of Orbital and Magnetic Orders to Colossal Negative Thermal Expansion in Novel Mott Insulators\textsuperscript{1} T.F. Qi, O.B. KORNETA, S. PARKIN, L.E. DE LONG, G. CAO, Center for Advanced Materials, University of Kentucky, P. SCHLOTTMANN, Department of Physics, Florida State University — Ca\textsubscript{2}RuO\textsubscript{4} is intimately associated with both negative volume thermal expansion (NVTE) and negative linear thermal expansion (NLTV) when doped by a 3d transition metal ion M for Ru. The NVTE and NLTE observed in this system constitutes a compelling and extraordinary example in that (1) the coefficient of NVTE and NLTE reaches -213 \times 10^{-6} K\textsuperscript{-1} and -148 \times 10^{-6} K\textsuperscript{-1}, respectively, constituting colossal negative thermal expansion (NTE); (2) the NTE anomalies closely track the onset temperatures of orbital and magnetic orders, in sharp contrast to classic NTE that shows no relevance to physical properties; (3) the NTE and physical properties can be effectively tuned via varying M and x in Ca\textsubscript{2}Ru\textsubscript{1-x}M\textsubscript{x}O\textsubscript{4}; (4) the NTE occurs near room temperature and extends over a wide temperature interval ranging from 100 K to 350 K. Moreover, NTE and Invar effect commonly exist in these 4d-based ruthenates and 5d-based iridates, e.g. Sr\textsubscript{n+1}Ir\textsubscript{n}O\textsubscript{3n+1} and BaIrO\textsubscript{3}. These novel NTE materials provide a much-needed paradigm for functional materials with anomalous thermal expansion and electronic characteristics.

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