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Coupling of Orbital and Magnetic Orders to Colossal Negative Thermal Expansion in Novel Mott Insulators[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_2RuO_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^{-1} and -148 $\times$ 10^{-6} K^{-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 $Ca_2Ru_{1-x}M_xO_4$; (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. $Sr_{n+1}Ir_nO_{3n+1}$ and $BaIrO_3$. These novel NTE materials provide a much-needed paradigm for functional materials with anomalous thermal expansion and electronic characteristics.

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