Spin-orbital superstructure in strained ferrimagnetic perovskite cobalt oxide

J. Fujioka\textsuperscript{a}, Y. Yamasaki\textsuperscript{b}, H. Nakao\textsuperscript{b}, R. Kuma\textsuperscript{b}, Y. Murakami\textsuperscript{b}, M. Nakamura\textsuperscript{c}, M. Kawasaki\textsuperscript{a,c}, and Y. Tokura\textsuperscript{a,c}

\textsuperscript{a}Department of Applied Physics and Quantum-Phase Electronics Center (QPEC), University of Tokyo, Hongo, Tokyo 113-8656, Japan
\textsuperscript{b}Condensed Matter Research Center (CMRC) and Photon Factory, Institute of Materials Structure Science (IMSS), KEK, Tsukuba, 305-0801, Japan
\textsuperscript{c}Center for Emergent Matter Science, RIKEN, Wako 351-0198, Japan, e-mail fujioka@ap.t.u-tokyo.ac.jp

Electronic phases with the nanometer-scale self-organization of electrons are ubiquitously observed in correlated electron systems. In $d$-electron transition metal oxides, versatile quantum states with charge-spin-orbital ordering have been realized by tuning the material control parameter such as the effective one-electron band-filling or band-width. In this study, we show that the control of the spin-state degree of freedom [low/intermediate/high spin states, see Fig (a)] gives rise to a new complex spin-orbital superstructure with spontaneous magnetization in a thin film of perovskite LaCoO$_3$ by means of x-ray diffraction, optical spectroscopy and magnetization measurements. A tiny crystal-lattice strain can promote the spin-state transition of Co ions and the ordering of Co $3d$ orbitals modulates the spin exchange interactions to produce the unique ferrimagnetic structure [Fig (b)]. The spin-state variability as a tuning parameter of the spin-orbital entanglement offers a unique opportunity for designing novel magnetic phenomena and spintronic functions.

Fig (a) Schematic view of the spin states of trivalent Co-ion (b) Electronic phase diagrams of the tensile strained LaCoO$_3$ and the bulk $R$CoO$_3$. The circle, square and triangle indicate the typical spin state crossover temperature, the transition temperature of orbital ordering (OO) and that of the ferrimagnetic (FerriM) ordering, respectively.