BL-3A/2012S2-005

Spin-orbital superstructure in strained ferrimagnetic perovskite cobalt oxide

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1 Introduction

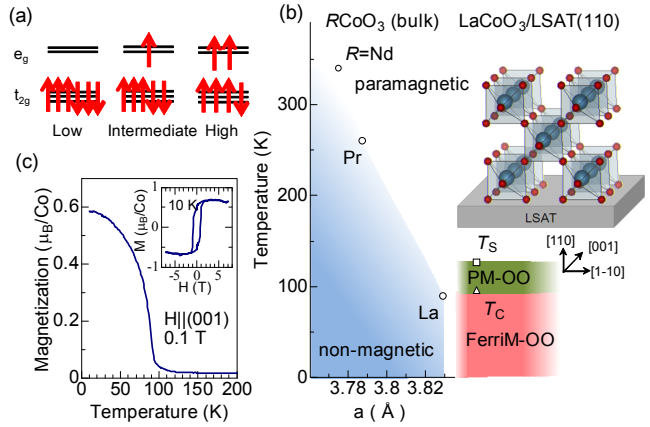
The interplay among charge, spin and orbital degrees of freedom in strongly correlated electron systems provides a unique arena where to produce emergent quantum states such as the multiferroelectricity and unconventional superconductivity. The perovskite $LaCoO_3$ is known as a prototype of the spin-state crossover material and has received much attention for these decades [1,2]. In this system, the nominally trivalent Co-ion (Co^{3+}) may take three different spin states as shown in Fig. 1(a); the low-spin (LS) state with filled 3d t_{2g} manifold (S=0), intermediate-spin (IS) state with active e_g and t_{2g} orbital degrees of freedom (S=1), and high-spin (HS) state with active t_{2g} orbital degree of freedom (S=2). Although the ground state is nonmagnetic phase with LS-state in the bulk form as shown in Fig 1(b), it has been reported that the spontaneous magnetization is observed in the epitaxial thin film of $LaCoO_3$ while keeping its insulating nature [3,4]. Several models have been proposed to explain the strain induced magnetization; the chemically inhomogeneous state including ferromagnetic metallic patches originating from unintentionally doped carriers or the ferromagnetism mediated by the orbital ordering. However, to the best of our knowledge, there is no experimental report exploring the microscopic spin-orbital structure for this enigmatic magnetic phase. Here we have investigated the spin-orbital state for the epitaxial thin film of $LaCoO_3$ by magnetization measurements and resonant x-ray diffraction [5].

2 Experiment

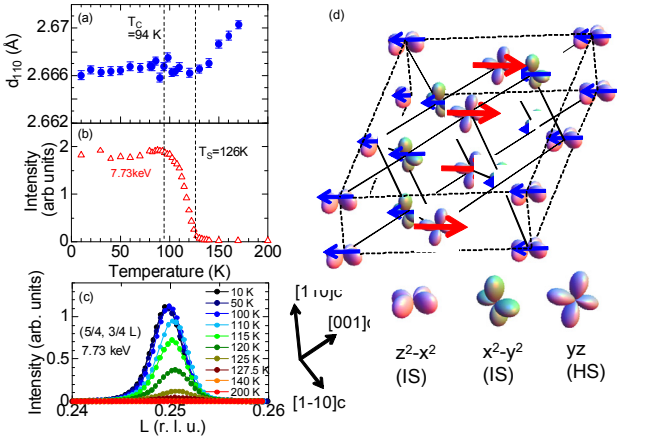
The single crystalline film (60 nm thick) of $LaCoO_3$ is fabricated on the $(LaAlO_3)_{0.3}(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}$ (LSAT) substrate with (110) orientation by pulsed laser deposition. The synchrotron-radiation x-ray diffraction was performed to detect the fundamental and superlattice reflection at BL-3A and BL-4C, Photon Factory of KEK, Japan.

3 Results and Discussion

Figure 1(c) shows the temperature dependence of magnetization. Magnetization increases at T_C=94 K and the spontaneous magnetization is about 0.7μ_B/Co at 10 K. Figure 2(a) shows the temperature dependence of the out-of-plane lattice constant (d_{110}). With lowering temperature, d_{110} steeply decreases down to 126 K and undergoes



(a) Schematic view of the spin states of Co^{3+} (b) Electronic phase diagrams of the tensile strained $LaCoO_3$ and the bulk $RCoO_3$ (R is rare earth element). The circle indicates the typical spin state crossover temperature from nonmagnetic to paramagnetic state. The square and triangle denote the transition temperature of orbital ordering (OO) and that of the ferrimagnetic (FerriM) ordering, respectively. (c) Temperature dependence of in-plane magnetization. Inset shows the in-plane magnetization (M) curve at 10 K.



Temperature dependence of (a) d_{110} and (b) scattering intensity of (5/4, 3/4, 1/4) reflection (c) Profile of (5/4, 3/4, 1/4) reflection (d) The plausible model of spin-orbital ordering.

minimal temperature dependence below 126 K. Moreover, we have identified a superlattice reflection characterized by the propagation vector q=(1/4, -1/4, 1/4) below 126 K. Figure 2(c) exemplifies the profile of superlattice reflection at (5/4, 3/4, 1/4). The intensity gradually increases below 126 K and nearly saturates below T_C [Fig. 2(b)].

These results point to the existence of the structural phase transition at 126 K (T_S), which quadruples the unit cell of pseudo-cubic setting along the [100], [010] and [001] axes. On the basis of the resonant x-ray scattering at Co K-edge, we attributed this structural phase transition to the ordering of Co-3d orbital with IS- or HS-state. Figure 2(d) shows the one plausible model of spin-orbital ordering; the spin and orbital alignment is viewed as the stacking of the two dimensional sheets of IS- and HS-state sites in order of IS ↑ -IS ↑ - HS ↓ -IS ↑ ⋯ spin configuration with dz^2-x^2-dx^2-y^2- d_{yz}-dx^2-y^2 ⋯ orbital configuration along [1 -1 1] direction. This model is also consistent with the observed net magnetization and result of resonant soft x-ray diffraction [6].

References

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