Photoinduecd spin-state dynamics in thin film of perovskite cobalt oxide investigated by time-resolved x-ray diffraction

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Research on macroscopic quantum systems, which involve interplays among charge, spin, and orbital degrees of freedom, is important in condensed matter physics because it can lead to the emergence of unique optical, magnetic, elastic, and thermoelectric phenomena. The phase transition caused by optical stimulation is called photoinduced phase transition (PIPT) and has been of current interest during the past decades. With time-resolved techniques by using femto- and picosecond pulsed lights, it is possible not only to observe the dynamics of the electronic interactions among the degrees of freedom but to control the electronic, magnetic, and orbital order on an ultrafast timescale.

Perovskite-type cobalt oxides showing spin-crossover phenomenon have been regarded as promising materials exhibiting novel PIPT phenomena [1-3]. The $LaCoO_3$ containing the nominally trivalent Co ion (Co^{3+}) is a typical spin-crossover system and may take three different spin state: the low-spin (LS) state (t_{2g}^6, S = 0), intermediate-spin (IS) state ($t_{2g}^5e_g^1$, S = 1), and high-spin (HS) state ($t_{2g}^4e_g^2$, S = 2). The spin-crossover transition from nonmagnetic LS ground state to paramagnetic HS and/or IS states occurs with increasing temperature around 90 K. The recent research has revealed that the magnetic properties and the Co-3*d* orbital ordering can be tuned by the epitaxial strains [4,5]. In this study, we investigated the photoinduced state in an epitaxial thin film of $LaCoO_3$. To identify the transient structural change correlated with the spin states directly, we measured a time-resolved x-ray diffraction at PF-AR NW14A.

The single crystalline film (60 nm thick) of $LaCoO_3$ was 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, which exhibits the strain induced spontaneous magnetization (T_c = 94 K) and the lattice distortion with Co-3*d* orbital ordering below 124 K [4]. Figure 1(a) shows the (620) fundamental reflection profiles in $LaCoO_3$. The peak, which shows a single-peak feature at 120 K, splits into two at 80 K. The peak splitting is ascribed to the lattice distortion with Co-3*d* orbital ordering [4]. Figure 1(b) shows the difference peak profile between before and after photoexcitation at 80 K, obtained by an optical-pump and x-ray-probe technique. The x-ray pulse duration is 100 ps and the photon energy of the optical pump pulse is 3.10 eV with the second harmonics of the fundamental Ti:sapphire regenerative amplified pulse (photon energy: 1.55 eV, pulse duration: 120 fs, and repetition rate: 1 kHz). The shape of the difference peak profile indicates that the two peaks are changed toward the single-peak feature. This result suggests the structural change due to the photoinduced melting of the orbital order. In this talk, we will also present the transient reflectivity change reflecting the change of the spin state, measured by femtosecond time-resolved reflection spectroscopy, and discuss the PIPT in terms of the spin-structural dynamics.

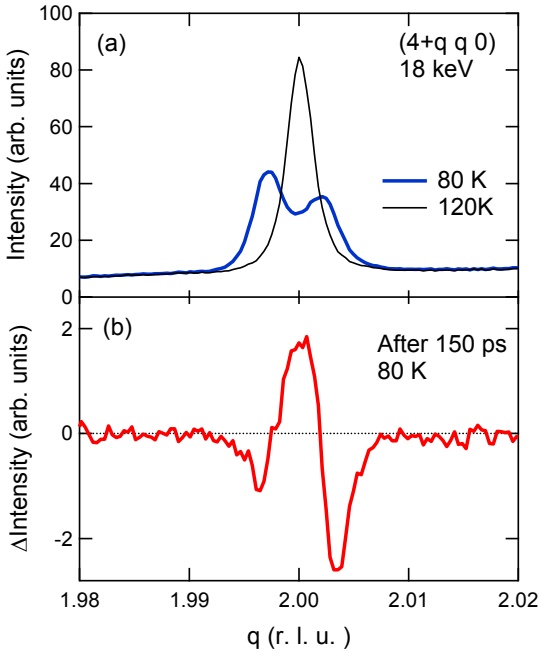

Fig. 1(a) X-ray diffraction profiles of (620) fundamental reflection along (110) direction at 80 K and 120 K. (b) Differential profile of fundamental reflection before and after photoexcitation (150 ps) at 80 K.

Reference

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