EPJ Web of Conferences **41**, 03013 (2013)
DOI: 10.1051/epjconf/20134103013

Photoinduced Coherent Spin Fluctuation in Primary Dynamics of Insulator to Metal Transition in Perovskite Cobalt Oxide

T. Ishikawa[1], K. Yamada[1], H. Itoh[1,2], S. Iwai*[1,2], T. Arima[3], S. Yamada[4], and T. Sasaki[2,5]

[1] Department of Physics, Tohoku University, Sendai 980-8578, Japan
*E-mail: s-iwai@m.tohoku.ac.jp
[2] JST, CREST, Sendai 980-8578, Japan
[3] Department of Advanced Materials Science, Univ. of Tokyo, Kashiwa, 277-8561, Japan
[4] International College of Arts and Sciences, Yokohama City University, Yokohama, 236-0027, Japan
[5] Institute for Materials Research, Tohoku University, Sendai, 980-8577, Japan

Abstract. Coherent spin fluctuation was detected in the photoinduced Mott insulator-metal transition in perovskite cobalt oxide by using 3 optical-cycle infrared pulse. Such coherent spin fluctuation is driven by the perovskite distortion changing orbital gap.

1 Introduction

Rhombohedrally distorted perovskite-type transition metal oxide $LaCoO_3$ (Fig. 1(a)) is of considerable interest, because it exhibits a unique two-step transitions (Fig. 1(b)): a spin state transition at T_s~120 K and an insulator-to-metal (I-M) crossover at T_{IM}~530 K [1-4]. At low temperatures of less than T_s, $LaCoO_3$ is a nonmagnetic band insulator. The electron configuration of Co^{3+} ion in $LaCoO_3$ is $3d^6$ with a low spin (LS, S=0; t_{2g}^6) state in which the t_{2g} level is fully occupied. Increased magnetic susceptibility with temperature up to T_s is attributed to the spin state transition

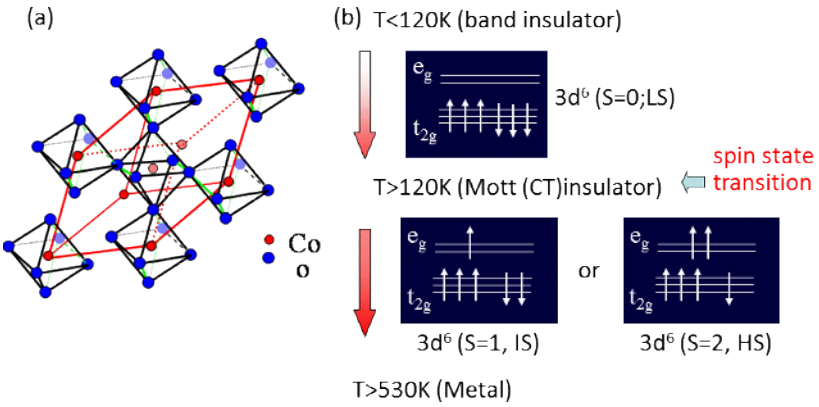

Fig. 1. (a) Crystal structure of $LaCoO_3$, (b)Schematic illustrations of spin state transition in $LaCoO_3$.

from LS to a high spin state (HS, S=2; $e_g^2\ t_{2g}^4$) or an intermediate spin (IS, S=1; $e_g^1\ t_{2g}^5$) state as shown in Fig. 1(b), which results from the competitive energy balance between the crystal field splitting (10*Dq*; t_{2g}- e_g gap) and the Hund's coupling energy. Because of the strong on-site Coulomb repulsion, the 3*d* band in the IS (or HS) state is split into an upper and lower Hubbard band. Because the O2*p* band is located in the Hubbard gap, the IS (or HS) state of $LaCoO_3$ is classified into the charge transfer (CT) insulator.

The multifarious ground states of $LaCoO_3$ enable us to expect a gigantic and/or ultrafast optical responses which are driven by the cooperative and critical characteristics in the strongly correlated system. Optical excitation of the CT band in the S=1 or S=2 state leads to the Photoinduced I-M trasnition (PIMT) and the photoinduced spin-state transition (PIST) in $LaCoO_3$ and in the related compounds [5-7]. In $LaCoO_3$, the characteristic IR active Co-O stretching mode [2, 3] and the Raman active breathing, Jahn-Teller (JT) modes [4] in the perovskite structure are know to be coupled with the spin-state transition. The frequency of such 500-700 cm^{-1} modes can be adressed in real time axis by the 10 fs pulse. In this study, we investigated the role of perovskite distortions interacting with spin transition in the primary dynamics of the photoinduced phase transitions (PIPTs).

2 Experiment

3-cycle 12 fs pulse in the 1.2-1.8 μm wavelength region which was generated in optical parametric amplifier using type I BBO with degenerate configuration and chirped mirror compressor. Pulse width evaluated from the FROG pattern is 12 fs which corresponds to 3-optical cycle. The time resolution of the reflection detected pump-probe measurement is 15 fs.

3 Results and discussions

Fig. 2 shows the time profile of reflectivity change ΔR/R reflecting the primary dynamics of the PIPT. Pumping energy was 0.68-0.93 eV corresponding to the CT excitation from O 2*p* to Co 3*d* upper Hubbard band. The build-up time of ΔR/R is faster than 50 fs and intense oscillation is observed. The oscillating period and the dumping time are evaluated as 30 fs (~1100 cm^{-1}) and 50 fs, respectively. In $LaCoO_3$, it is well known that characteristic infrared active [2, 3] and Raman active modes [4] (500~700 cm^{-1}) strongly couple with the spin-state transition. In particular, Raman active modes give the overtone at 1100-1300 cm^{-1}, respectively. Observation of such overtone is attributable to the nonlinearity of these Raman modes resulting from the interaction with the

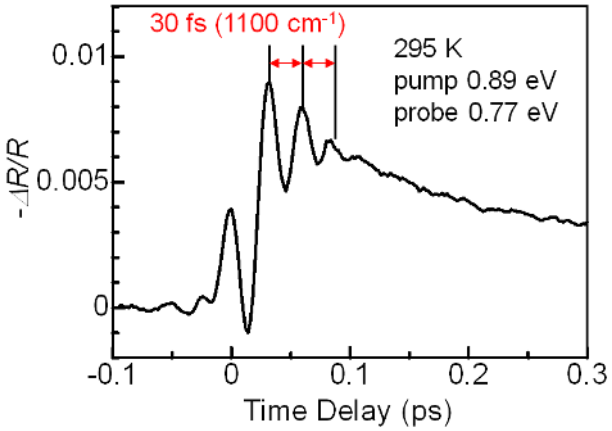

Fig 2. Time profile of reflectivity change ΔR/R induced by the photoexcitation of 12 fs, 0.89 eV pulse. Excitation intensity was 0.03mJ/cm^2.

electronic (and/or magnetic) states. Considering that, the observed 30 fs (1100 cm^{-1}) oscillation is attributable to the overtone of such perovskite distortions. The lifetime broadening of this oscillation (> 700 cm^{-1}) is much larger than the bandwidth of the steady state Raman and infrared spectra, suggesting that this 30 fs oscillation is not generated in the ground state but induced in the excited state, although the detailed mechanism remains unclear. Possible scenario leading to the PIST is that the coherent perovskite oscillation induces the instability of the spin state, considering the critical energy balance between $10Dq$ and the Hund's coupling energy.

4 Summary

Ultrafast coherent perovskite distortion was detected by using 12 fs, 3-optical cycle pulse in the early stage dynamics of the PIPT in $LaCoO_3$. The PIST is initially triggered by this oscillation which can modulate spin states.

Reference

1. S. Yamaguchi, Y. Okimoto, H. Taniguchi, and Y. Tokura, Phys. Rev. B, **53**, 2926R (1996).
2. S. Tajima, A. Masaki, S. Uchida, T. Matsuura, K. Fueki and S. Sugai, J. Phys. C: Solid State Phys., **20**, 3469 (1987).
3. S. Yamaguchi, Y. Okimoto, and, Y. Tokura, Phys. Rev. B, **55**, 8666R (1997).
4. A. Ishikawa, J. Nohara, and S. Sugai, Phys. Rev. Lett., **93**, 136401 (2004).
5. S. Iwai, S. Tomimoto, Okamoto, J. P. He, Y. Kaneko, H. Okamoto, Y. Tokura, Abstract of JPS meeting, **59**, 673(2004).
6. Y. Okimoto, T. Miyata, M. S. Endo, K. Onda, T. Ishikawa, S. Koshihara, M. Lorenc, E. Collet, H. Cailleau, and T. Arima, Phys. Rev. B, **84**, 121102R (2011).
7. Y. Kanamori, H. Matsueda, and S. Ishihara, Phys. Rev. Lett., **107**, 167403 (2011).
8. Y. Kawakami, T. Fukatsu, Y. Sakurai, H. Unno, H. Itoh, S. Iwai, T. Sasaki, K. Yamamoto, K. Yakushi, and K. Yonemitsu, Phys. Rev. Lett., **105**, 246402 (2010).