10 fs dynamics of photoinduced magnetic transition in double-layered charge ordering in $LuFe_2O_4$ under interlayer excitation

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Abstract: Photoinduced growth of the antiferromagnetic state in the ferrimagnetic phase were demonstrated in double layered Fe oxide $LuFe_2O_4$ by 12 fs infrared pulse under inter-layer excitation. Inter-layer charge imbalance successively induce the changes of charge/magnetic structures interacting with local Fe-O stretching and inter-layer sliding phonons through the exchange interaction.

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1. Introduction

Optical control of magnetic properties in strongly correlated system through the charge-spin interactions such as double-exchange interaction and Dzyaloshinsky-Moriya interactions attract much attention in perovskite manganese. On the other hand, double-layered charge ordering (CO) in $LaFe_2O_4$ enables us to expect another mechanism of photoinduced magnetic response. In $LuFe_2O_4$, as shown in Fig. 1(a), the LuO_2 and doubled FeO layers having a triangular lattice stack along the *c*-axis. While average valence of Fe ions is 2.5+, Fe^{2+}/Fe^{3+} superstructure appears in the FeO layers with an intra-layer three-fold periodicity. Below T_{CO3D} ~ 330 K, the CO develops along the *c*-axis and an inter-layer arrangement of the charge pattern is locked, in other words, three-dimensional (3-D) CO appears. Consequently, macroscopic polarization or ferroelectricity are expected to be appeared from periodical charge imbalance between neighboring FeO layers[1, 2]. It is noteworthy that such 3-D CO also arise the magnetic transitions; i.e., Ferrimagnetic (fE) ordering sets in below T_N = 240 K. Antiferromagnetic (AFM) ordering grows in fE phase below T_{LT} = 175 K through the charge -spin coupling in double layered structure (Fig. 1(b))[3]. In this compound, optical responses in visible [4] and THz [5] region have been investigated. However, ultrafast optical response in mid-IR has not been measued, although fm-AFM change markedly affect the mid-IR reflectivity as shown below.

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 [6].

3. Results and discussions

Fig. 1(c) shows the steady state reflectivity (*R*) spectrum at 150 K for E//c polarization. As shown in Fig. 1(d), *R* shows the anomalous change between 150-200 K for E//c (arrows). Therefore, *R* in mid-IR region reflects the change of the magnetic property, i.e., the growth of the AFM state in the FE phase.

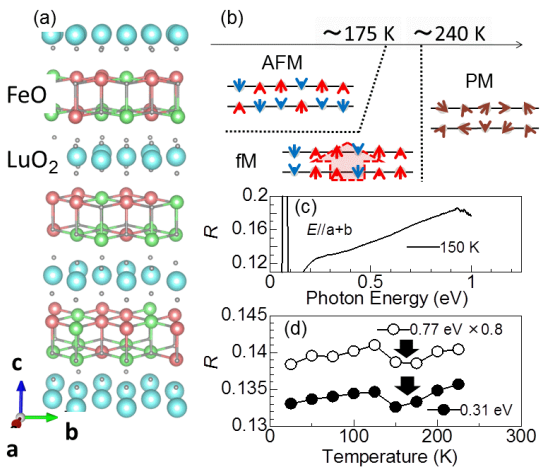

Fig. 1. **(a)** Crystal structure of $LuFe_2O_4$ consisting of FeO double -layers and LuO_2 blocks **(b)** Schematic illustration of magnetic transition. **(c)** Steady state reflectivity (R) at 150 K. **(d)** Temperature dependence of R at 0.77 eV and at 0.31 eV

Figs. 2(a)(b) show transient reflectivity $\Delta R/R$ at 150 K for E_{pump}//c (a) and for E_{pump}//a+b (b) measured at 0.1 ps after excitation of 100 fs pulse. Excitation energy 0.89 eV corresponds to the Fe^{2+}-Fe^{3+} CT excitation for both polarizations. For both excitation polarization, $\Delta R/R$ is positive at low energy (< 0.5 eV), whereas $\Delta R/R$ is negative at high energy region (>0.5 eV). As shown in Fig. 2(c), anomalous change in $\Delta R/R$ signal at 150-200 K indicates that the increase of *R* is closely related to the change of the magnetic property. Such anomaly in $\Delta R/R$ cannot be observed for E_{pump}//a+b, indicating that $\Delta R/R$ spectrum for E_{pump}//c does not reflect the magnetic property.

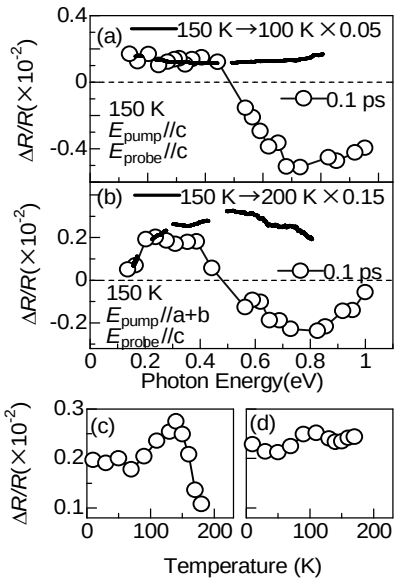

Fig 2. **(a)** Δ*R*/*R* spectrum for E_{pump}//c (0.3 mJ/cm^2), E_{probe} //c at 150 K **(b)** ΔR/R spectrum for E_{pump}//a+b (0.3 m/cm^2), E_{probe} //c at 150 K. Spectral differences [*R*(100K)-*R*(150 K)]/*R*(150 K) **(a)** and [*R*(200K)-*R*(150 K)}/*R*(150 K) **(b)** are also shown by solid curves. **(c)(d)** Temperature dependences of Δ*R*/*R* for E_{pump}//c , E_{probe} //c **(c)** and for E_{pump}//a+b , E_{probe} //c**(d)**

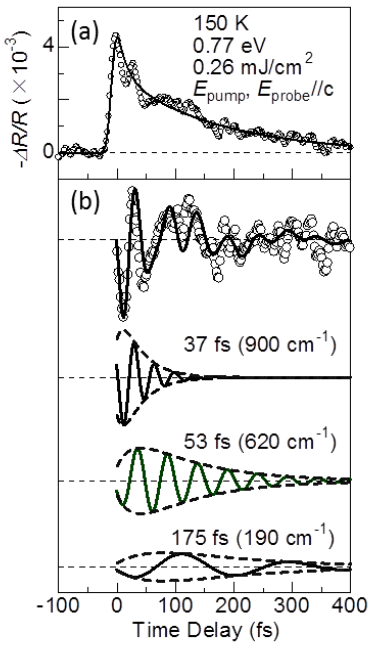

Fig.3. **(a)** Time evolution of ΔR/R at 150 K(0.26 mJ/cm^2) measured by using 12 fs pulse for E_{pump}//c , E_{probe}//c.
(b) Oscillating components .

The spectral shape of the reflectivity increases is analogous to that *R*(100K)-*R*(150 K)/*R*(150 K) (solid courve in Fig. 2(a)), reflecting the growth of the AFM in fE phase. On the other hand, Δ*R*/*R* at 300 fs is analogous to the [*R*(200K)-*R*(150 K)]/*R*(150 K)(solid curve in Fig. 2(b)), indicating the temperature rise.

Figs. 3 shows the time evolution of Δ*R*/*R* at 0.64 eV (Fig. 3(a)) measured by using 12 fs pulse and the oscillating component (Fig. 3(b)). The time profile exhibits instantaneous rise and 150 fs decay. Oscillating structures with the frequencies of 900 cm^{-1}, 620 cm^{-1} and 190 cm^{-1}. were also observed. According to the assignment for the IR and Raman spectra, these oscillations are attributable to the overtone of local Fe-O stretching mode (//c), Fe A_g (//c) mode, and E_g (//a+b) mode, respectively. In particular, A_g (//c) , and E_g (//a+b) modes has been known to be coupled to the the fM-AFM crossover. It is noteworthy that the decay time constant of the 900 cm^{-1} (35 fs) corresponds to the rise time of 620 cm^{-1}, and the decay of 620 cm^{-1} (100 fs) is approximately equal to the rise of 190 cm^{-1} mode. Such successive appearance of the the coherent phonons reflects the interactions between the change of the charge/magnetic properties triggered by the inter-layer CT excitation and the phonons; i.e., i) charge imbalance is locally induced , ii) such imbalance is extended and stabilized in the double layer , iii) magnetic ordering changes from fE to AFM. Here, it should be emphasized that the decay time of the 620 cm^{-1} mode 100 fs (=rise of 190 cm^{-1} mode) roughly corresponds to the energy scale of the exchange interaction and/or the spin-orbit interaction.

4. Summary

Photoinduced growth of the anti-ferromagnetic state in the ferrimagnetic phase was demonstrated by 100 fs mid-infrared transient reflectivity measurement. Furthermore, the fast dynamics were also investigated using 12 fs infrared pulse. Successive phonon dynamics interacting with the changes of the charge /magnetic properties have been clarified.

5. References

[1] N. Ikeda, H. Ohsumi, K. Ishii, T. Inami, K. Kakurai, Y. Murakami, K. Yoshii, S. Mori, Y. Horibe, and H. Kito, "Ferroelectricity from iron valence ordering in the charge-frustrated system $LuFe_2O_4$", Nature 436, 1136(2005).
[2] S. Ishihara, J. Phys. Soc. Jpn., 79, 011010(2010).
[3]J. Lee, S. A. Trugman, C. D. Batista, C. L. Zhang, D. Talbayev, X. S. Xu, S. -W. Cheong, D. A. Yarotski, A. J. Taylor, and R. P. Prasankumar, "Probing the interplay between quantum charge fluctuations and magnetic ordering in LuFe2O4", Sceintific Reports, 3, 2654
[4] J. Bourgeois, G. Andre, S. Petit, J. Robert, M. Poienar, J. Rouquette, E. Elkaim, M. Hervieu A. Maignan, C. Martin, and F. Damy, "Evidence of magnetic phase separation in $LuFe_2O_4$", Phys. Rev. B86, 024413(2012).
[5]H. Itoh, K. Itoh, K. Anjo, H. Nakaya, S. Iwai, S. Ishihara, H. Akahama, D. Ohishi, T. Kambe, and Ikeda, ""Ultrafast melting of charge ordering in LuFe2O4 probed by terahertz spectroscopy", J. Lumin. 56-58, 484(2011).
[6] Y. Kawakami, T. Fukatsu, Y. Sakurai, H. Unno, H. Itoh, S. Iwai, T. Sasaki, K. Yamamoto, K. Yakushi, and K. Yonemitsu, "Early-Stage Dynamics of Light-Matter Interaction Leading to the Insulator-to Metal Transition in a Charge Ordered Organic Crystal", Phys. Rev. Lett. 105, 246402(2010).