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Photoinduced Phase Transition in $Pr_{0.5}Ca_{0.5}CoO_3$ Studied by Sequential Pulse Excitations

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A perovskite-type cobalt oxide, $Pr_{0.5}Ca_{0.5}CoO_3$ (PCCO), shows photoinduced phase transition. In this study, we successively irradiated two laser pulses with different intensities to PCCO and probed the transient change of the reflection at 2.0 eV. Assuming propagation of the two different photoinduced metallic states, we could reproduce the time profiles as well as the magnitude in the reflectance change, indicating the fabrication of the photoinduced multilayered thin film in $Pr_{0.5}Ca_{0.5}CoO_3$.

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1. Introduction

In recent years, many studies have been done about photoinduced phase transition (PIPT) [1]. Among those materials showing the PIPT phenomena, a perovskite cobalt oxide, $Pr_{0.5}Ca_{0.5}CoO_3$ (PCCO) is one of the examples [2]. PCCO shows a metal–insulator transition with change of the spin configuration at around T_c = 89 K [3]. In this material, a nominal valence of cobalt ion is 3.5+ and hence Co^{3+}:Co^{4+} = 1:1. Upon the insulator–metal transition, the spin state in Co^{3+} changes from low spin (LS, $S = 0$) state to intermediate spin (IS, $S = 1$) state [2]. Very recently, we demonstrated PIPT from the LS insulating to the HS metallic state and a resultant real space propagation of the photoinduced metallic domain in the depth direction [3], which are characteristic in the PCCO material. In this study, we further investigated the PIPT phenomena in PCCO in the light of the spatial dynamics, aiming at photonically create a dynamical multilayer composed of two different metallic states on the sample of PCCO by irradiation for two successive laser pulses with different intensities as shown in Fig. 1.

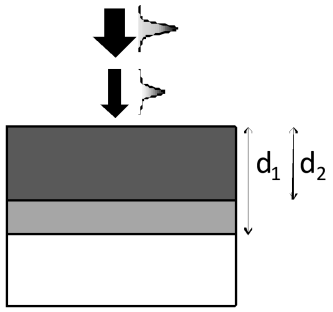

Fig. 1. A schematics of the dynamical bilayer composed of two different metallic states by the irradiations of two laser pulses. d_1 and d_2 denote the thickness of each photonic domain created by the successive laser pulses.

2. Experiment

The transient reflectivity change after irradiating femtosecond laser pulses at 30 K (the LS insulating state) was obtained by the pump–probe method. The light source is regenerated amplified Ti:sapphire laser (1.55 eV, 1 kHz, 120 fs). The laser light was divided into two beams: one is used for probe pulse to detect the reflection change, after converting the wavelength up to 2.0 eV by optical parametric amplifier (OPA). The other beam is used as a pump light. For the successive excitations with two pulses, the pump light further splits into two beams with different intensity and interval (Δt) between the two controlled by neutral density filters and a delay stage.

3. Results and discussion

Figure 2a shows the time profile of reflectivity change ($\Delta R/R$) after the photoirradiation. The vertical dashed

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lines denote the timing of each excitation and Δt was set at 10 ps. The pump fluences of the first and second pulse are about 2 mJ/cm^2 and 10 mJ/cm^2, respectively. After the first excitation, $\Delta R/R$ shows an instant jump and shows a broad peak at around 7 ps. This is due to the interference of the probe light within the photodomain as described in Ref. [2]. The enhanced $\Delta R/R$ further jumps by the second and stronger excitation at 10 ps, indicating the successive propagations of the photoinduced metallic domains.

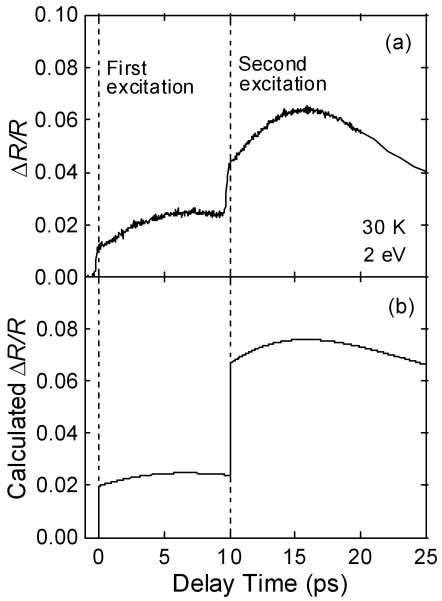

Fig. 2. (a) Time profile of $\Delta R/R$ after the sequential excitations at 2 eV. First and second pump laser power are about 2 mJ/cm^2 and 10 mJ/cm^2, respectively, and time interval between the two pulses is 10 ps. (b) Calculated time profile of $\Delta R/R$ using the parameters of γ_1 (= 0.25) and γ_2 (= 0.68).

To discuss the obtained time profile, we define the following function:

$$F\big(\varepsilon^{\mathrm{M}}, \varepsilon^{\mathrm{I}}, \gamma, z\big) = \gamma \exp(-z/d)\varepsilon^{\mathrm{M}} + [1 - \gamma \exp(-z/d)]\varepsilon^{\mathrm{I}}, \quad (1)$$

where γ is the efficiency of the photoinduced phase transition ($0 < \gamma < 1$), and ε^{M} and ε^{I} are the dielectric functions in the IS metallic phase and the initial insulating state, respectively. Besides, z is distance from the sample surface and d the penetration depth of the pump light (ca. 60 nm) [2]. Equation (1) is called a simple mixing model*, which could reproduce the time profile of $\Delta R/R$ after a single pulse excitation in PCCO, by regarding γ as the pump fluence [6]. If we assume that the second pulse excites the IS insulating region that was not still excited by the first pulse, the total efficiency γ is expressed as $\gamma = \gamma_1 + (1 - \gamma_1)\gamma_2$, γ_1 and γ_2 being that by first and second laser pulse excitation, respectively. Under these circumstances, the dielectric function after the sequential irradiations is represented as follows:

$$\varepsilon(z) = \begin{cases} F\big(\varepsilon^{\mathrm{M}}, \varepsilon^{\mathrm{I}}, \gamma, 0\big) & (0 < z < d_2) \\ F\big(\varepsilon^{\mathrm{M}}, \varepsilon_1, \gamma_2, z - d_2\big) & (d_2 < z < d_1) \\ \quad \big(\varepsilon_1 = F\big(\varepsilon^{\mathrm{M}}, \varepsilon^{\mathrm{I}}, \gamma_1, 0\big)\big). & \\ F\big(\varepsilon^{\mathrm{M}}, \varepsilon^{\mathrm{I}}, \gamma_1, z - d_1\big) & (d_1 < z) \end{cases} \quad (2)$$

In the above equations, the d_1 and d_2 are the thickness of each propagating domain as depicted in Fig. 1, and related with the delay time (t_{d}) by the relations that $d_1 = vt_{\mathrm{d}}$ and $d_2 = v(t_{\mathrm{d}} - \Delta t)$, where v is the sound velocity in the IS metallic state [6]. In terms of the dielectric functions expressed by Eq. (2), we can calculate $\Delta R/R$ after the sequential excitation as a function of t_{d}.

In Fig. 2b, we plotted the calculated time profile of $\Delta R/R$. In the calculation, the values of γ_1 and γ_2 are set at 0.25 and 0.68, according to the previous literature [6]. The calculated result semiquantitatively reproduces two experimental features, i.e., the shape of the time profile and the magnitude of $\Delta R/R$ as shown in Fig. 2a, supporting the model proposed in Fig. 1 after the sequential pulse excitations.

It is worth to noting that the refractive index in each layer can be controlled by changing the intensity of the excitation light, and that the spatial pattern of the photoexcited state can also be freely designed on the scale of the wavelength of pump light. This enables us to fabricate various dynamical and rewritable photonic crystals or optical waveguides on PCCO, which leads to applying ultrafast communication devices using photoinduced phenomena.

4. Summary

We performed pump–probe reflection spectroscopy by irradiation of two sequential femtosecond laser pulses. The time dependence of $\Delta R/R$ was semiquantitatively reproduced assuming the dynamical bilayer composed of two different photoinduced metallic states. This can be viewed as a photonic fabrication of the heterostructure on the perovskite-type cobalt oxide.

Acknowledgments

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[1] For a comprehensive review, see *Photoinduced Phase Transitions*, Ed. K. Nasu, World Sci., Singapore 2004.

* In addition to the simple mixing model, the Bruggeman effective medium theory (BEMT) [4] is also used to describe the photoinduced state [5]. In the case of PCCO, the difference between the ε^{M} and ε^{I} is so small that the EMT gives almost the same results as the simple mixing model.

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