

# Terahertz radiation characteristics of charge-ordered manganite $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ thin films excited by ultrafast optical pulses

Noriaki Kida <sup>a,b,1</sup>, Masayoshi Tonouchi <sup>a,b</sup>

<sup>a</sup>Research Center for Superconductor Photonics, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

<sup>b</sup>CREST, Japan Science & Technology Corporation (JST), 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

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## Abstract

Terahertz radiation characteristics of photoswitching devices made on charge-ordered manganites  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  have been investigated for thin films grown at different temperatures. We report that the temperature dependence of the THz radiation near the charge ordering temperature strongly depends on the growth condition of  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ , while that near the magnetic transition temperature does not.

**Key words:** terahertz radiation; manganites;  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ ; femtosecond optical pulses

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Terahertz (THz) frequency range covered  $0.1 \sim 10 \times 10^{12}$  Hz (corresponds to the photon energy between 0.4–40 meV), is at boundaries between electromagnetic and light waves, which are generated by an electrical circuit and a commercial light source, respectively. Due to the lack of the convenient generation technique, THz frequency range is referred to as “smoked THz gap” [1]. During last decade, the rapid progress in THz-pulse generation and detection system has been made by using the photoconducting semiconductor switch excited by femtosecond optical pulses [2] and by the subsequent discovery of THz radiation from new class of materials including semiconductors, quantum wells, and superconductors. Recently, we have reported the first finding of the THz radiation from the “magnetic” material; the charge-ordered manganite  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  thin films by exciting carriers with femtosecond optical pulses [3]. However, fundamental THz radiation characteristics are not examined so far. In the case of low-temperature (LT) GaAs, which is widely used as a THz emitters, its THz radiation characteristics strongly depends on the growth condition. In this work, we show THz radiation characteristics of  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  thin

films on MgO substrates grown at different temperatures ( $790^\circ\text{C}$  and  $950^\circ\text{C}$ ).

We used two samples, which are deposited at  $790^\circ\text{C}$  (sample A) and  $950^\circ\text{C}$  (sample B) by a pulsed laser deposition technique [3]. The femtosecond laser pulses (pulse width, 80 fs; wavelength, 800 nm; repetition rate, 82 MHz) are irradiated on the gap ( $\sim 5 \mu\text{m}$ ) of the bowtie antenna, which is fabricated on  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  thin film. The radiated THz pulse is detected by a bowtie-type LT-GaAs antenna. The detailed sample preparations, characterizations, and experimental setup can be found in Refs. [3,4].

Figures 1 (A) and (B) show the generated electromagnetic waveform of samples A and B measured at 23 K and 21 K, respectively. The bias voltage and the laser power for sample A (B) are 32 V (35 V) and 184 mW (138 mW), respectively. The main peaks were observed around 0 ps. The pulse widths (full width at half maximum) for samples A and B are estimated to be  $\sim 1$  ps and  $\sim 1.3$  ps, respectively. Such a picosecond electromagnetic pulse is usually regarded as THz radiation because its frequency component extends up to  $\sim 1$  THz. THz radiation from  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  can be phenomenologically explained by the creation of the transient photocurrent of the order of subpicosecond with ultrafast optical pulses (the radiated field is pro-

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<sup>1</sup> Corresponding author. Fax: +81-6-6879-7984. E-mail address: kida@rcsuper.osaka-u.ac.jp (N. Kida)

portional to the time derivative of the current) [3]. The oscillations after the main peak mainly arises from the absorption of a water vapor and the asymmetry of the propagated THz pulse. The pulse width of sample A is nearly same as that of sample B, which is in strong contrast to the THz properties of LT-GaAs. THz pulse width in LT-GaAs is mainly determined by the electron trapping time on the shallow donor state, therefore the precipitation of As by changing the growth condition plays crucial role to control the pulse width [5]. Besides, the strength of both the bias voltage and the laser power have no influence on the waveform in  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ , which have effectively affect in the case of LT-GaAs [6].

We show in Figs. 1 (a) and (b) the temperature  $T$  dependence of the main peak amplitude  $E_{\text{THz}}$  in the time-domain waveform around 0 ps of the samples A and B, respectively. With increasing  $T$ ,  $E_{\text{THz}}$  of both samples decrease and show a broad minimum around  $T_C \sim 130$  K, where the magnetization of both samples steeply increases with decreasing  $T$  [4]. For further increasing  $T$ , we observed different  $E_{\text{THz}}(T)$  behavior of two samples:  $E_{\text{THz}}$  of sample A exhibits the broad maximum around 230 K, which corresponds to the charge ordering temperature  $T_{\text{CO}}$  [7]. However,  $E_{\text{THz}}$  of sample B shows the less  $T$  dependence above  $T_C$  and no correlation with  $T_{\text{CO}}$ . According to x-ray diffraction profiles, the sample A is nearly  $a$ -axis oriented, while we detected the intense (110) peak instead of (100) peak in sample B, indicating that the sample B does not undergo the charge ordering transition.

In the cases of the THz radiation from semiconductor surfaces [8] and photoswitches [9], it was reported that  $E_{\text{THz}}(T)$  follows the  $T$  dependence of the static mobility. Therefore, in this classical picture, one can be anticipated that  $E_{\text{THz}}(T)$  from  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  resembles the  $T$  dependence of the conductivity  $\sigma$ . However,  $\sigma$  exponentially increases with increasing  $T$  and shows no clear correlation with  $T_C$  and  $T_{\text{CO}}$ . One possibility to understand  $E_{\text{THz}}(T)$  behavior is that the THz radiation is related to the photo-induced insulator-metal transition. We observed that the THz radiation in sample A suddenly decreases in intensity with increasing the bias voltage under irradiating laser pulses, which is a typical feature of the photo-induced phase transition. Fiebig *et al.* have already revealed that the metallic phase creates less than 1 ps as a result of the melting of the charge-ordered state [10]. This picture can explain that the pulse width does not strongly depend on growth temperatures, the negligible effect on the waveform by varying the strength of the bias voltage and the laser power, and different  $E_{\text{THz}}(T)$  behavior around  $T_{\text{CO}}$  between two samples. However,  $E_{\text{THz}}(T)$  below  $T_C$  of the sample B shows same tendency as that of the sample A whereas the sample B does not show the charge ordering. Therefore, we speculate that the

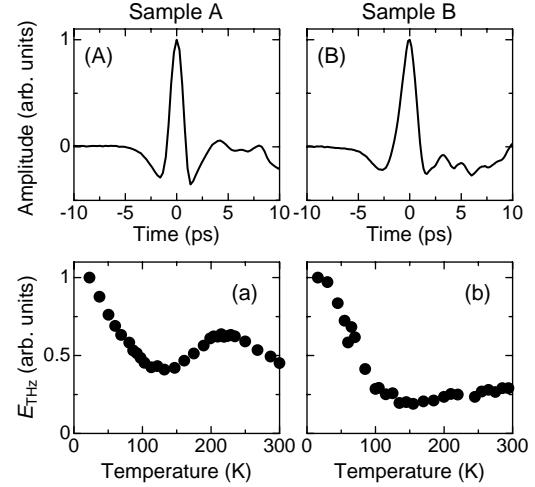


Fig. 1. Generated THz pulse in the time-domain of the samples A (A) and B (B) measured at 23 K and 21 K, respectively. The zero position of the time delay between pump and probe pulses is arbitrary units. Temperature dependence of the maximum amplitude  $E_{\text{THz}}$  of the samples A (a) and B (b) in the time-domain waveform around 0 ps. The respective data are normalized by the maximum value for comparison.

ultrafast modulation of spin ordering is responsible for the behavior of  $E_{\text{THz}}(T)$  below  $T_C$ . Further study is necessary and in progress to confirm the above scenarios.

In summary, we briefly report the THz radiation characteristics from  $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  thin films grown on different temperatures.  $E_{\text{THz}}$  exhibits the anomalous  $T$  dependence:  $E_{\text{THz}}(T)$  around  $T_{\text{CO}}$  strongly depends on the growth condition, while that around  $T_C$  does not, implying the photo-induced spin modulation as a source of THz radiation below  $T_C$ .

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