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The effect of optical pump on the absorption coefficient of $0.65CaTiO_3$-$0.35NdAlO_3$ ceramics in terahertz range

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A B S T R A C T

The absorption coefficient of $0.65CaTiO_3$-$0.35NdAlO_3$ ceramics under external optical fields was investigated by terahertz time-domain spectroscopy in a frequency range of 0.2 THz to 1 THz at room temperature. It could be found that the variation of the absorption coefficient is approximately from 8.95 cm^{-1} to 26.02 cm^{-1} in the frequency range of 0.2 THz to 1 THz, and the tuning range is about 3.32 cm^{-1} at 0.6 THz which almost reaches up to nearly 23.32%. The micromechanism of these results was attributed to the excited free carriers by the external optical pump intensity.

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1. Introduction

Over the past few decades, perovskite ceramics were widely used in various fields [1,2] because of their superior physical and chemical properties [3]. In particular, rapid development of wireless communication gave microwave dielectric ceramics with medium dielectric constant and high quality factor the opportunity to be commonly used in military and mobiles communication [4–7]. It was very easy to obtain the practical microwave dielectric ceramics by combining two or more perovskite structure compounds [8,9]. Widely used microwave dielectric device is based on a solid solution of $CaTiO_3$–$NdAlO_3$ (CTNA), which has medium dielectric constant ($\varepsilon_r \approx 45$), high quality factor ($Q \times f \approx 40000$GHz) and near zero temperature coefficient of resonant frequency τ_f [10], respectively.

According to the previous reports, different components of (1-x) $NdAlO_3$-x$CaTiO_3$ ceramics were synthesized successfully by various methods [11–13]. Their crystal structure changed with the composition, the dielectric constant and temperature coefficient of the resonant frequency enlarged with the increase of x [10,11]. Extended X-ray absorption fine structure (EXAFS) analysis was used to study the local structural environments for positive ions of $Ca_{0.7}Nd_{0.3}Ti_{0.7}Al_{0.3}O_3$ ceramics at 298 K [12]. Previous research group also investigated the influences of preparation process on the synthesis, sintering and microwave dielectric properties of $0.7CaTiO_3$-$0.3NdAlO_3$ ceramics [13].

However, the effect of optical pump on the absorption coefficient of $0.65CaTiO_3$-$0.35NdAlO_3$ ceramics in the terahertz (THz) range has rarely been reported so far. In this work, the transmittance spectra of $0.65CaTiO_3$-$0.35NdAlO_3$ ceramics in the THz range under an external optical pump field were measured. The growing light-induced carriers result in the increase of absorption coefficient. From the investigated results, it could be found that the $0.65CaTiO_3$-$0.35NdAlO_3$ was a potential candidate for applications in the THz range, such as THz absorber and THz capacitor.

2. Experiment

The sample of $0.65CaTiO_3$-$0.35NdAlO_3$ ceramics was grown using high energy ball milling technique. The starting materials were high purity Al_2O_3, Nd_2O_3, TiO_2, and $CaCO_3$. All of the materials were thoroughly mixed in nylon jar for 24 h with ZrO_2 balls, then dried calcined, milled and pressed into disk. Finally, the sample was sintered at 1420 °C for 6 h in air.

The crystalline phase was identified by X-ray diffraction (XRD) analysis using monochromatic Cu K_α radiation (X'Pert PRO of PANalytical B.V.). And the environmental scanning electron

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microscope (ESEM; Quanta 200, FEI Inc., Hillsboro, OR) was used to observed the grain size and morphology of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics.

The transmittance spectra of the 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics were measured with a THz time-domain spectrum (THz-TDS) system produced by Zomega Terahertz Co. (USA) at room temperature. The dynamic range was more than 70 dB, the measured frequency resolution was 10 GHz, and the THz spot size at the focus was 3 mm. The external optical pumping was employed with an all-solid-state green CW laser (center wavelength of 532 nm) which had a 5 mm diameter spot size. In the process of experiment the laser was obliquely incident on the surface of the 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics at an angle of 45°. In order to eliminate the absorption of terahertz waves by water vapor presented in the atmosphere, all the measurements were carried out at 22 °C (295 K) in a dry air environment.

3. Results and discussions

The structural investigation of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics has been performed by XRD scan, as shown in Fig. 1. It can be seen that the sample is well crystallized and exhibit the peaks characteristic to the simple perovskite phase.

Fig. 2 shows the surface ESEM micrographs of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics. The microstructure is dense with average grain size of approximately 1–2 μm.

Fig. 3 shows the time-domain waveforms of the 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics and the reference (air) which were obtained in the experiment. The time shift between the reference (air) and the 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics was 7.13 ps approximately. In addition, a time lag was also found in the time domain by applying external optical pumping. When the external optical light intensity was 300 mW, the transmittance waveform shifted approximately 0.06 ps compared with that without external optical pumping, as shown in Fig. 3(b).

The transmittance spectra of the 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics could be obtained from the ratio of the two waveforms via the equation: $T(\omega) = E_s(\omega, P)/E_r(\omega)$, where the $E_s(\omega, P)$ is the frequency waveform of the 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics and $E_r(\omega)$ is the frequency waveform of the air, P is the external optical light intensity. Thus the transmittance spectra $T(\omega)$ of the 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics under different external optical pump intensities have been calculated, as shown in Fig. 4.

Fig. 5 shows the real part $\varepsilon'(\omega)$ and imaginary part $\varepsilon''(\omega)$ of complex permittivity of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics under different external optical pump intensities, which were got through the transmittance spectra. From Fig. 5(b) it could be found that the magnitude of $\varepsilon''(\omega)$ was about 10.28–10.32 at 0.6 THz. In contrast, Fig. 5(a) demonstrates that the $\varepsilon'(\omega)$ does not show obvious difference with external optical pumping.

The absorption coefficient of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics can be expressed as $\alpha = -\ln(T)/d_0$, where T and d_0 are the transmittance and the thickness respectively, as shown in Fig. 6(a). It could be found that the variation of the absorption coefficient is approximately from 8.95 cm^{-1} to 26.02 cm^{-1} in the frequency range of 0.2 THz to 1 THz, and the tuning range under different external optical pump intensities is about 3.32 cm^{-1} at 0.6 THz which almost reaches up to nearly 23.32%, as shown in Fig. 6(b).

In addition, we have studied the relationship between the loss tangent and the optical pump intensity for the sake of clarifying the micromechanism of the variation of absorption coefficient of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics with external optical pump and

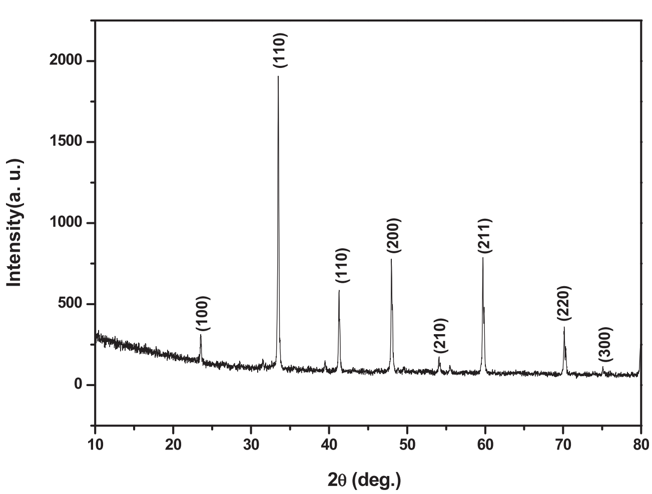

Fig. 1. X-ray diffraction pattern of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics.

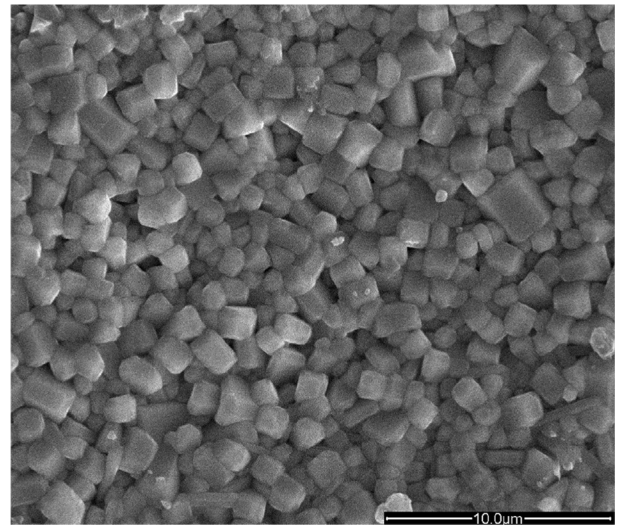

Fig. 2. ESEM micrographs of 0.65$CaTiO_3$-0.35$NdAlO_3$ ceramics.

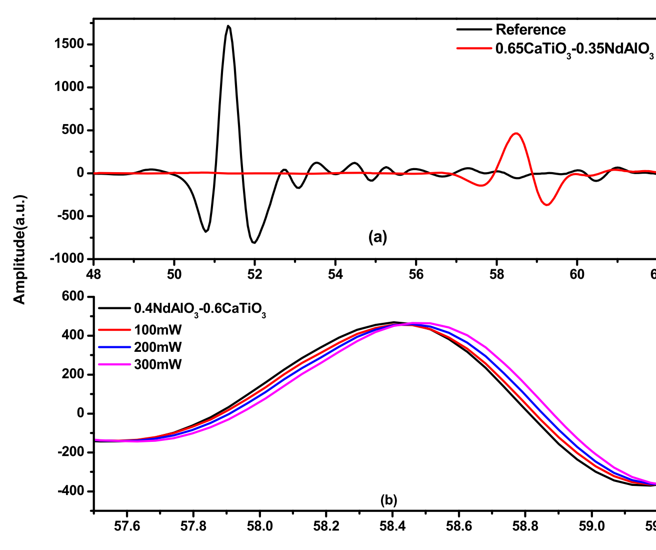

Fig. 3. (a) Time-domain transmittance waveform of 0.65$CaTiO_3$-0.35$NdAlO_3$ and (b) its time shift under different external optical fields at room temperature.

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