THz Spectrum of Cr^{3+}:$LiSrAlF_6$ Crystal

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***Abstract*—Absorption coefficient α(ν) and dielectric coefficient ε(ν) of crystal Cr^{3+}:$LiSrAlF_6$ were obtained by THz Time Domain Spectroscopy (THz-TDS) in the range of 0.2-1.2THz. The infrared transmission spectrum R(κ) was measured from 6.7cm^{-1}(0.2THz) to 4000cm^{-1}. There is a band of the phonon absorption in the crystal. The frequency ω_{LO} of the longitudinal optical phonon model LO is about 236 THz. And the ω_{TO} of the transverse optical phonon model TO is about 9.42THz.**

I. INTRODUCTION

The THz spectroscopic technology is playing a more and more important role in the study of the condensed matter physics at present. THz spectroscopic responses of the crystalline samples are primarily related to lattice vibration, optical phonon-photon, optical phonon-electron interaction, and other solid elementary excitations.

The Cr^{3+}:$LiSrAlF_6$ crystal is a promising material of laser and optics as an advanced all-solid-state tunable laser crystal[1]. In 1989, Payne S A et al. have grown Cr^{3+}:$LiSrAlF_6$ crystal firstly and studied its laser properties[2]. Near-infrared emissions observed from Cr^{3+}-doped $LiSrAlF_6$ (LSAF) are ascribed to a 3d-3d transition of Cr^{3+} replacing Al in the octahedral 2d sites.

The study on terahertz optical character of the Cr^{3+}: LSAF crystal is interesting in the THz techniques.

II. EXPERIMENT

The Cr^{3+}:$LiSrAlF_6$ single crystal is triangle structure, its point group is D_{3d} and space group is P31c[2]. The lattice parameters are a=0.502nm, and c=0.967nm. The sample was cut along the (100) plane and twin polished. The sample is cylinder, its thickness is 3.022mm and diameter is 5.718mm.

The THz spectrum was measured from 0.2THz to 1.2THz. Experimental measurement was conducted using the THz-TDS system[3]. Mai Tai laser is a pulse generator, with output frequency 82MHz, pulse width 100fs, center wavelength 800nm, and pulse power about 1080mW. The setup was placed in the chamber full of nitrogen in order to eliminate the influence of the vapour. The THz time domain spectrum was measured. The frequency-domain spectrum was also obtained through Fast Fourier Transformation. Absorption coefficient α(ν) of the sample was obtained through data computing. Dielectric coefficient ε(ν) was calculated from the relations between optical constants.

The Mid-infrared transmission spectra was measured on a Bruker Vertex 80v Fourier Transform Iinfrared(FTIR) spectrometer at the room temperature, and the wavenumber is from 50cm^{-1}(1.5THz) to 4000cm^{-1}.

III RESULTS AND ANALYSIS

The THz time domain waveforms of reference signal and transmission signal are shown in Fig.1. The measured pulses in the sample were delayed because of its slow velocity. The amplitude is suppressed because of the reflection and absorption of the THz wave. Fig. 2 shows Fourier transformation spectra of THz reference signal and the sample signal.

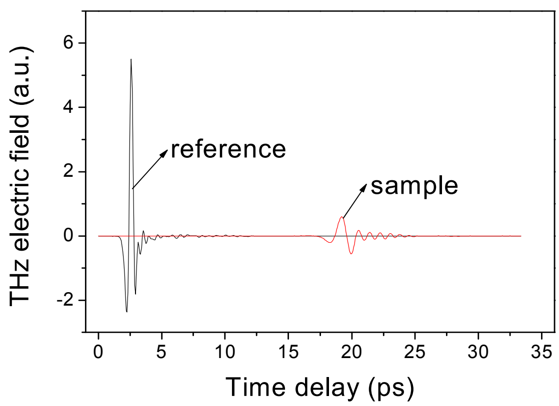

Fig. 1 THz time domain waveforms of reference signal and sample signal

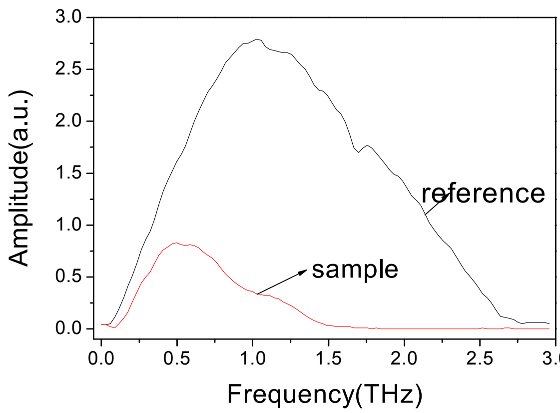

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Fig. 2 Fourier transform amplitude spectra of THz reference signal and transmission signal

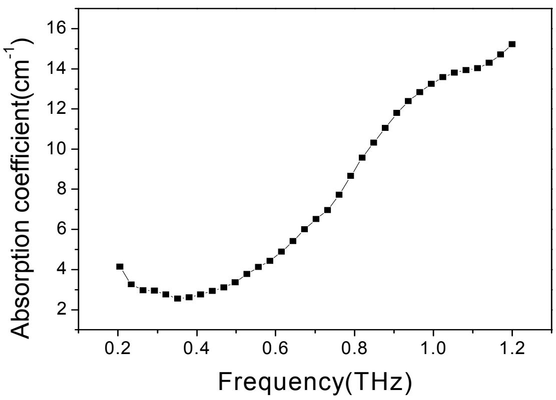

Fig. 3 The absorption coefficients $\alpha(\nu)$

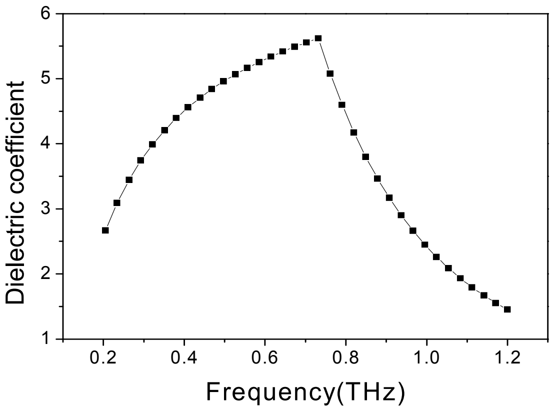

Fig. 4 The dielectric function $\varepsilon_1(\nu)$

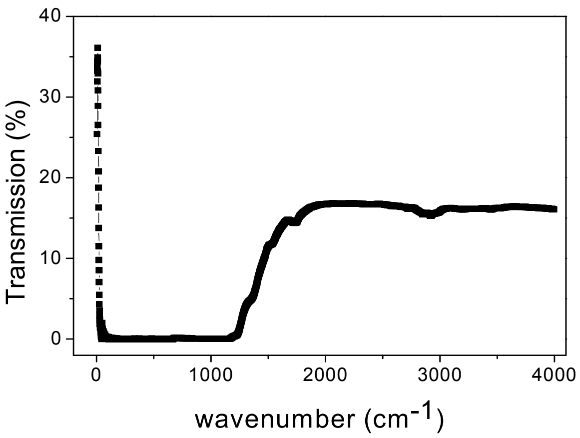

Fig. 5 The infrared transmission spectrum $R(\kappa)$

The value of absorption coefficient $\alpha(\nu)$ increases with increasing frequency as shown in Fig. 3. The $\alpha(\nu)$ is smaller than $10cm^{-1}$ under 0.8THz, and the $\alpha(\nu)$ is not larger than $34cm^{-1}$. A small protuberance is at 1.0 THz in the curve $\alpha(\nu)$. Maybe it is from the adulterated Cr^{3+} ions.

The infrared transmission spectrum $R(\kappa)$ measured is shown in Fig. 5. The equivalent frequency for smallest wavenumber $50cm^{-1}$ is 1.5THz. It is reasonable by comparison Fig. 3 and Fig. 5. The transmission spectrum $R(\kappa)$ is near zero in a region from $50cm^{-1}$ (1.5THz) to $1250cm^{-1}$ (37.5THz). It is a band of the phonon absorption in the crystal. The frequency ω_{LO} of the longitudinal optical phonon model LO is about $2\pi\times37.5THz=236$ THz. And the ω_{TO} of the transverse optical phonon model TO is about $2\pi\times1.5THz=9.42THz$.

The curve of the dielectric function $\varepsilon_1(\nu)$ decreases at 0.76THz as shown in Fig. 4 from 5.08(at 0.76THz) to 1.45 (at 1.2THz). The extinction coefficient $\kappa(\nu)$ is smaller than 0.03, we can consider $\varepsilon_1(\nu) = n^2(\nu)$. The dielectric function $\varepsilon_1(\nu)$ is a macroscopic measured quantity. Its change comes from the microcosmic polarizabilitiy $\alpha_k(\nu)$ as shown in the Clausius-Mossotti relation. The dipolar contribution to the polarizabilitiy is small and vanished at near microwave band. The ionic contribution is small and vanished at near infrared band because of the inertia of the molecular and ion. There is not dipolar contribution at the THz band, therefore the change of the dielectric function $\varepsilon_1(\nu)$ is from vanish of the contribution of the Sr^{2+}, Al^{3+}, F^- and Li^+ ions in turn in the range over 0.76THz .

CONCLUSIONS

The value of absorption coefficient $\alpha(\nu)$ of Cr^{3+}:$LiSrAlF_6$ crystal increases with increasing frequency in the range of 0.2-1.2THz. The transmission spectrum $R(\kappa)$ is near zero in a region from $50cm^{-1}$ (1.5THz) to $1250cm^{-1}$ (37.5THz). There is a band of the phonon absorption in the crystal. The frequency ω_{LO} of the longitudinal optical phonon model LO is about 236 THz. And the ω_{TO} of the transverse optical phonon model TO is 9.42THz..

The curve of the dielectric function $\varepsilon_1(\nu)$ decreases from 0.76THz. The change of the dielectric function $\varepsilon_1(\nu)$ originates from vanish of the ionic polarizabilitiy contribution.

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