

## Performance analysis of electro-optic sampling detection technique with thin GaSe crystal in mid-infrared band

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**Abstract:** Electro-optic sampling (EOS) detection technique has been widely used in the terahertz science and technology, and it also can measure the field time waveform of the few-cycle laser pulse. Its frequency response and band limitation are determined directly by the electro-optic crystal and duration of the probe laser pulse. Here, we investigate the performance of the EOS with thin GaSe crystal in the measurement of the mid-infrared few-cycle laser pulse. The shift of the central frequency and change of the bandwidth induced by the EOS detection are calculated, and then the pulse distortions induced in this detection process are discussed. It is found that this technique produces a red-shift of the central frequency and narrowing of the bandwidth. These changings decrease when the laser wavelength increase from 2  $\mu\text{m}$  to 10  $\mu\text{m}$ . This work can help to estimate the performance of the EOS detection technique in the mid-infrared band and offer a reference for the related experiment as well.

**Key words:** Electro-optic sampling, GaSe, mid-infrared, few-cycle laser pulse

**PACS:**

## 基于硒化镓晶体的电光取样探测技术在中红外波段的性能分析

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**摘要:** 电光采样探测技术已在太赫兹技术领域得到广泛应用, 该技术还可以测量少周期激光脉冲的电场时域波形。其频率响应和探测带宽直接由电光晶体和探测激光脉冲的脉宽决定。研究了基于薄硒化镓晶体的电光取样探测技术在中红外少周期激光脉冲测量中的频率响应特性和探测性能。计算了该技术在探测过程中引起的激光脉冲中心频率偏移和带宽变化规律, 进而讨论了引起的脉冲畸变规律。研究发现这种技术会导致探测的少周期激光脉冲中心频率发生红移、带宽变窄。当测量的激光波长从 2  $\mu\text{m}$  增加到 10  $\mu\text{m}$  时, 这种变化随之减小。本工作有助于评估电光取样探测技术在中红外波段的测量性能, 并为相关实验提供参考。

**关 键 词:** 电光取样; 硒化镓; 中红外; 少周期激光脉冲

**中图分类号:** O434.3

**文献标识码:** A

### 1 Introduction

Free-space electro-optic sampling (EOS) detection technique has been widely used in terahertz science and

technology since it has been demonstrated in the experiment in the last century<sup>[1-2]</sup>. This method can measure the time waveform of terahertz electric field by measuring the changing of the polarization of the probe laser when both copropagate through a thin electro-optic (EO) crystal. The crystal will appear birefringence in the biased of the terahertz field due to the electro-optic effect (Pockels effect)<sup>[3-4]</sup>. The probe laser has much higher frequency

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than the terahertz wave thus the latter can be treated as a DC field. Then, the time waveform of the terahertz pulse can be sampled by controlling the time delay between itself and the probe laser pulse<sup>[3-4]</sup>. Nowadays, this method, due to its simple optical alignment and high sensitivity, has been a very common and nearly standard coherent detection method in the terahertz science.

The performance of EOS is directly determined by the properties and thickness of the EO crystal, and the duration of the probe laser pulse. The EO crystal should have a low absorption in the terahertz range, high EO coefficient, and easier for the velocity match between the terahertz wave and the probe laser pulse, such as zinc telluride (ZnTe) and gallium phosphide (GaP). Different crystals make this detection technique different frequency response, and consequently different bandwidth limitation. The EOS technique with ZnTe at 1 mm thick has a detection bandwidth of 0.1-3 THz, while with GaP at 0.5 mm thick has a bandwidth up to 8 THz<sup>[5-7]</sup>.

The thin selenium gallium (GaSe) crystal also has been used in the EOS detection as a sensor, which can measure much broad terahertz radiation, up to several tens of terahertz<sup>[8-9]</sup>. The distortions of the broadband terahertz pulses induced by the EOS with thin GaSe are also systematically investigated<sup>[10]</sup>. As a good coherent detection, EOS with thin GaSe crystal even can work well in the mid-infrared band, especially obtaining the time waveform of the few-cycle laser pulse with high time resolution with a short enough probe laser pulse<sup>[11-14]</sup>. Liu *et al* and Eisele *et al* have separately demonstrated that well in experiment<sup>[11-12]</sup>, which open new applications of EOS in the measurement of the few-cycle laser pulse. However, the EOS detection has its band limitation and frequency response depending on the EO crystal<sup>[15]</sup>. These might produce some pulse distortions duration the measurement and affect the accuracy of the experimental measurement. Here, we calculate the frequency response of the EOS with thin GaSe crystal in the mid-infrared band (the wavelength from 2  $\mu\text{m}$ ~10  $\mu\text{m}$ , the frequency from 30 THz~150 THz), and investigate its performance in the measurement of mid-infrared few-cycle laser pulse. The pulse distortions of the few-cycle laser, including the shift of the central frequency and change of the bandwidth, are discussed based on the simulations as well.

## 2 Frequency response of EOS with thin GaSe crystal in the mid-infrared band

GaSe is a nonlinear crystal, which has been successfully employed to generate coherent radiation in the terahertz and mid-infrared range<sup>[16]</sup>. Its refractive indexes, including ordinary light and extraordinary light, in the optical band can be described by two fitted equations as<sup>[17]</sup>:

$$n_o^2(\lambda) = 7.443 + \frac{0.4050}{\lambda^2} + \frac{0.0186}{\lambda^4} + \frac{0.0061}{\lambda^6} + \frac{3.1485\lambda^2}{\lambda^2 - 2194}, \quad (1)$$

$$n_e^2(\lambda) = 5.760 + \frac{0.3879}{\lambda^2} - \frac{0.2288}{\lambda^4} + \frac{0.1223}{\lambda^6} + \frac{1.855\lambda^2}{\lambda^2 - 1780}, \quad (2)$$

where  $n_o$  is the refractive index of ordinary light,  $n_e$  is the that of extraordinary light, and  $\lambda$  is the wavelength of the light in micrometer. The absorption coefficient in the mid-infrared and terahertz range be obtained from the extinction coefficient by  $\alpha(f) = \frac{2\pi\kappa(f)}{c}$ . The extinction coefficient  $\kappa(f)$  and the refractive index  $n(f)$  are the imaginary and the real part of the complex dielectric index  $\varepsilon_c$ :  $\sqrt{\varepsilon_c(f)} = n(f) + i\kappa(f)$ . The complex dielectric index  $\varepsilon_c$  in this range is given by<sup>[18]</sup>:

$$\varepsilon_c(f) = S_1 + \frac{(f_L^2 - f_T^2)S_1}{f_T^2 - f^2 - i\Gamma_1 f}, \quad (3)$$

where  $f$  is the frequency of the wave in terahertz range,  $S_1 = 5.76$  THz is the amplitude of lattice vibration,  $f_T = 7.11$  THz is the transverse optical phonon vibration frequency,  $f_L = 7.36$  THz is the longitudinal optical phonon vibration frequency, and  $\Gamma_1 = 0.084$  THz is the damping constant. Figure 1(a) shows the ordinary and extraordinary light refractive indexes of GaSe in the mid-infrared band, and Fig. 1(b) shows its absorption coefficient. This calculated absorption coefficient is agreed well with the experimental result<sup>[19]</sup>. It can be seen there is no mutation in the curves higher than the phonon resonance 7.4 THz, which means it might work well in the mid-infrared band as a sensor.

When the probe laser pulse and the mid-infrared few-cycle pulse (to be detected) collinearly propagate along in the EO crystal, the mismatch between the phase velocity of few-cycle pulse and the group velocity of the probe laser determines the efficiency and bandwidth of the EOS detection technique. Here, the probe pulse is supposed to be short enough to be treated as a gate, such as 15 fs or even shorter<sup>[11-14]</sup>. (The probe laser pulses with duration of several femtoseconds are used in some experiments, as shown in the Table 1 below). A response function based on the propagation effect is described the efficiency on the frequency bandwidth of EOS detection as<sup>[20-21]</sup>

$$G(f, d) = \frac{2}{1 + n(f) + i\kappa(f)} \frac{1}{d} \int_0^d \int_{-\infty}^{+\infty} \exp\{[i(kz - 2\pi ft)] \cdot \delta(z - v_g t)\} \exp(-\alpha z) dz dt, \quad (4)$$

which is dependent on the thickness of the EO crystal  $d$  and the frequency  $f$ . Here, the term  $\frac{2}{1 + n(f) + i\kappa(f)}$  is the frequency-dependent transmission coefficient of the mid-infrared few-cycle pulse from the vacuum into the EO crystal,  $\delta$  function characterizes the transient feature of Pockels effect of the electric field on the probe laser pulse in the EO crystal,  $\alpha(f) = \frac{2\pi\kappa}{c}$  is the absorption coefficient of the crystal in the mid-infrared range. Equation (4) is simplified to

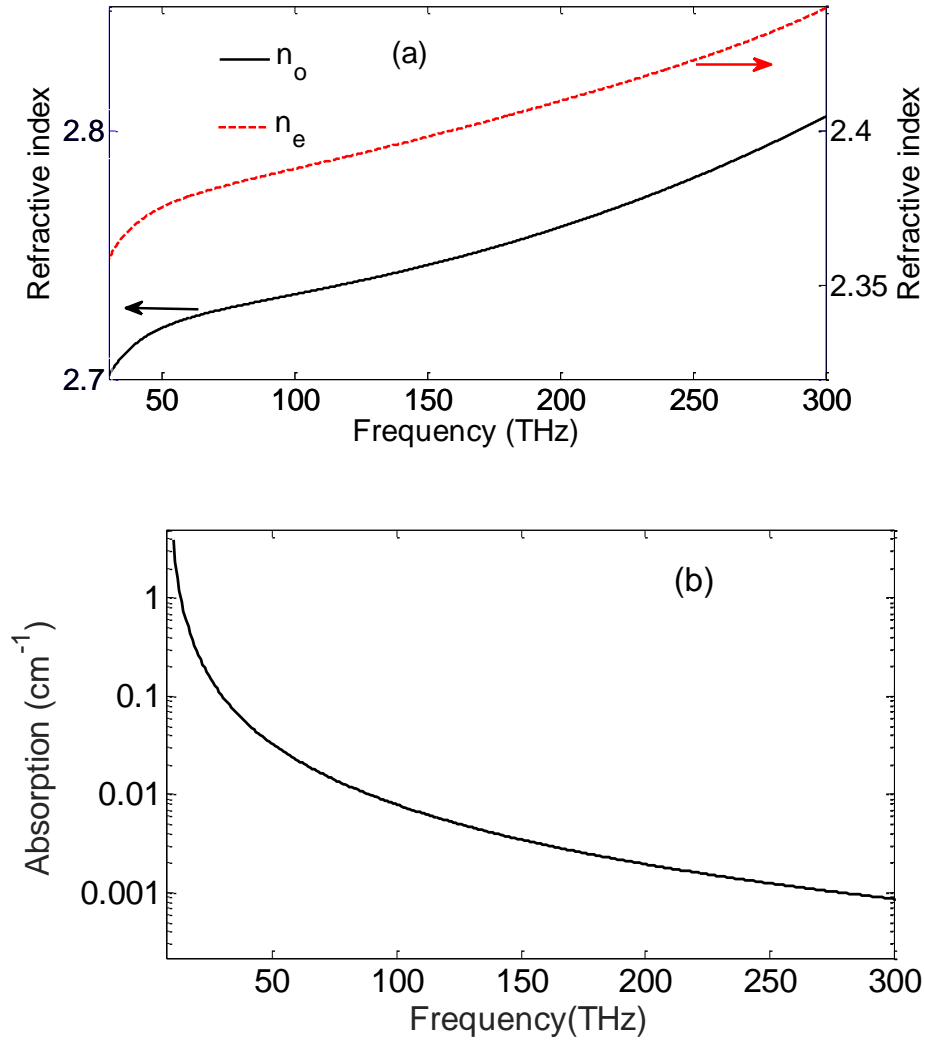


Fig. 1 (a) The ordinary and extraordinary light refractive indexes and (b) the absorption of GaSe crystal in the mid-infrared range.

图1 寻常光和非寻常光在硒化镓晶体中的(a)折射率和(b)吸收系数。

$$G(f, d) = \frac{2}{1 + n(f) + i\kappa(f)} \frac{1}{d} \int_0^d \exp \left\{ i2\pi f z \left( \frac{1}{v_{ph}(f)} - \frac{1}{v_g} \right) \right\} \exp(-\alpha z) dz, \quad (5)$$

where  $v_{ph}(f)$  is the phase velocity of mid-infrared few-cycle pulse at frequency  $f$  in the crystal,  $v_g$  is the group velocity of the probe laser in the crystal.  $v_{ph}(f) = c/n(f)$ ,

while  $v_g = \frac{c}{n_0} \left( 1 + \frac{\lambda}{n_0} \frac{dn_0}{d\lambda} \right)$ , both can be calculated through numerical method easily from Eqs. (1) - (2). The EO coefficient  $r_{41}$  of the crystal in the mid-infrared is treated with a constant, as there is no longer any transverse phonon. Thus, the whole response function including the EO coefficient is  $G_{EOS}(f, d) = G(f, d) \cdot r_{41}$ .

For two different probe laser pulse (the wavelengths 800 nm and 1030 nm), the response functions with GaSe at four different thicknesses (10  $\mu\text{m}$ , 20  $\mu\text{m}$ , 30  $\mu\text{m}$ ,

and 50  $\mu\text{m}$ ) are plotted in Figure 2. It shows that the thickness of GaSe decides the curves of the frequency response; the thinner the crystal is, the broader the frequency response is; the probe laser with a wavelength of 1030 nm is much better than that of 800 nm since the latter has more dips in the curve. The dips in the frequency response curve increase when the thickness of the crystal increases. In the frequency domain, these dips mean very low response for the corresponding frequency components of pulses. These will make the detection generate pulse distortions since the few-cycle laser pulse (including broadband terahertz pulse) is broadband. Many solid femtosecond laser systems (such as Ti: sapphire) offer laser pulses with a central wavelength of 800 nm, while Yb-based fiber laser systems offer the laser with a wavelength of 1030 nm. The response curves of the other wavelengths also can be obtained from the above equations.

Mid-infrared few-cycle laser pulse has huge applica-

tions in the gaseous spectroscopy and high field physics, and its generation and coherent detection are the basic of its applications. It is known that the EOS technique with thin GaSe crystal can measure the time waveform of terahertz pulse, while it has been used to measure the time waveform of the mid-infrared few-cycle laser pulse. Then the frequency distribution of few-cycle laser can be obtained by Fourier transform with phase together. This is an obvious advantage comparing with the spectrometer. Since the response function of the EOS detection with GaSe is not a flat curve, as shown in the Fig. 2, this detection method has different sensitivity for different fre-

quency components. The few-cycle laser pulse usually is several femtoseconds long, and with a broad bandwidth in the frequency domain. Therefore, this affects the accuracy of the detection, even might induce some pulse distortions in the detection process. The duration of the probe laser is also important in the measurement, but a probe pulse with 15 fs duration (or even shorter) has much broadband frequency bandwidth covering the frequency response limitation induced by the EO crystal.

### 3 Applications of EOS with GaSe crystal

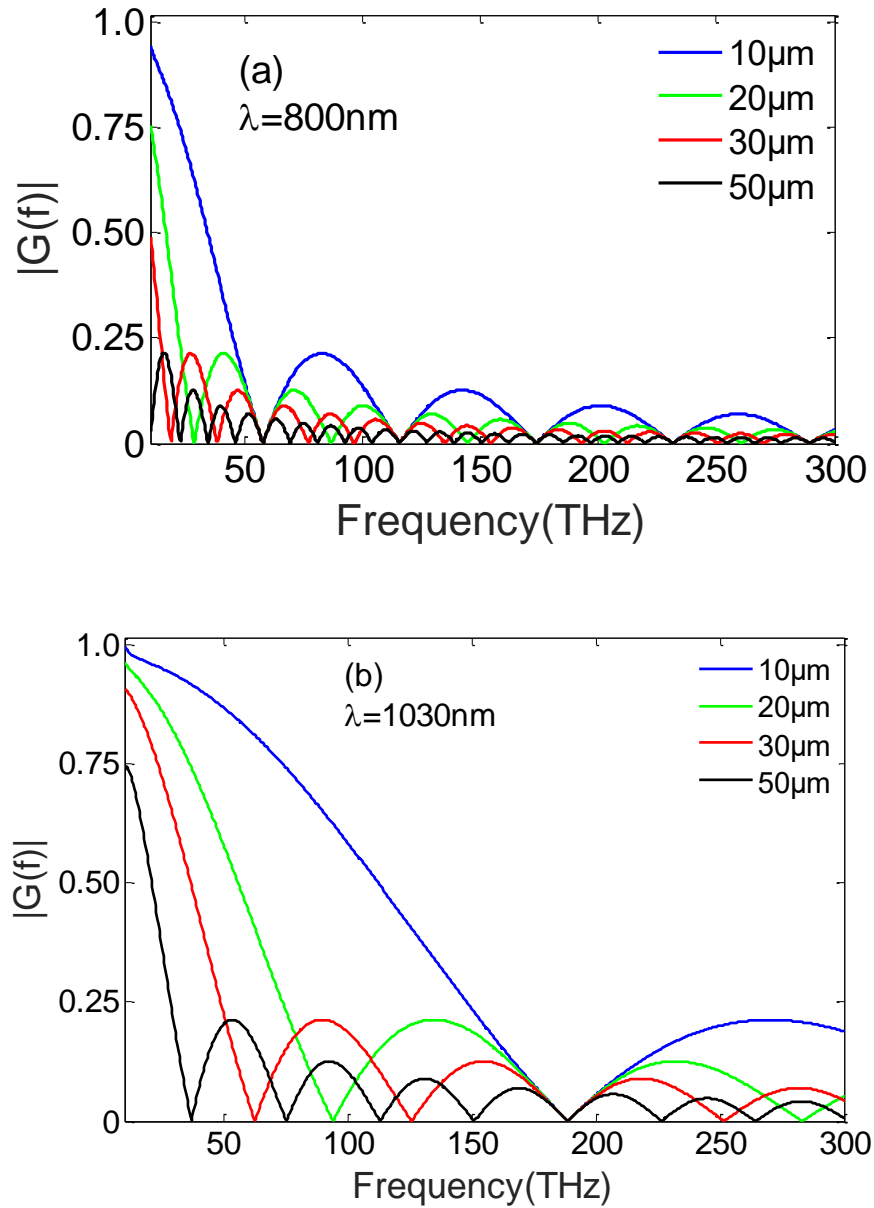


Fig. 2 The response function of the EOS with different GaSe thicknesses (10  $\mu\text{m}$ , 20  $\mu\text{m}$ , 30  $\mu\text{m}$ , and 50  $\mu\text{m}$ ) with two different probe laser wavelengths, (a) is with 800 nm, and (b) is with 1030 nm.

图2 不同厚度(10  $\mu\text{m}$ , 20  $\mu\text{m}$ , 30  $\mu\text{m}$ , and 50  $\mu\text{m}$ )的硒化镓晶体和两种不同探测激光波长的电光取样技术频率响应函数曲线, (a)所用激光波长为800 nm, (b)所用激光波长为1030 nm。

## in the mid-infrared range

The mid-infrared few-cycle laser pulse to be detected has a standard Gaussian profile as

$$E(t) = A_0 \exp(-t^2/T^2) \cos(2\pi f t + \phi_0) \quad (6)$$

where  $A_0$  is the amplitude,  $T$  is connected to the pulse full-width at half-maximum (FWHM)  $\tau$  by  $T = \frac{\tau}{2\sqrt{\ln(2)}}$ ,  $f$  is the central frequency of the laser,

$f = \frac{c}{\lambda}$  with  $\lambda$  at several micrometer, and  $\phi_0$  is the initial pulse phase. The interactions of such pulse with the atoms and molecules are determined by its phase (the carrier-envelope-phase, CEP) directly<sup>[22]</sup>. Thus, the precise measurement of the time waveform of such pulse is important.

In the measurement by the EOS detection, the few-cycle laser pulse to be detected is treated as an input signal for the EOS detection system with a special frequency response. The frequency spectrum of a few-cycle laser pulse is calculated from the Fourier transform of its time waveform:

$$E_{input}(f) = \mathcal{F}\{E_{input}(t)\}. \quad (7)$$

Then, the signal in the frequency domain after the EOS detection system is given by:

$$E_{output}(f) = E_{input}(f) \cdot G_{EOS}(f). \quad (8)$$

The signal in the time domain after the detection is obtained from the inverse Fourier transform of its frequency spectrum:

$$E_{output}(t) = \mathcal{F}^{-1}\{E_{output}(f)\}. \quad (9)$$

Thus, the changing of a few-cycle laser pulse before and after the EOS detection is obtained by comparing the signals above, including the time domain and the frequency domain. The dispersion induced by the EO crystal can be omitted if its thickness is very small.

We calculated the changing of a 2- $\mu\text{m}$  few-cycle laser pulse by the EOS detection technique from Eqs. (5)-(9). Here, the few-cycle laser pulse has 1.5 cycles in the envelope, and the probe laser has a wavelength of 1030 nm. Figure 3(a) shows the time waveforms before and after the EOS detection with two thicknesses of GaSe (10  $\mu\text{m}$  and 20  $\mu\text{m}$ ); Fig. 3(b) shows their frequency spectra, respectively. Although the probe laser pulse will be extended longer because of the group velocity dispersion induced by the crystal, this value is small since the crystal is very thin<sup>[23]</sup>. As shown in Fig. 3: (1) the amplitude of the pulses after detection become small although the waveforms don't change; (2) the frequency distributions are changed obviously, including a red-shift of the central frequency and a changing of the bandwidth; and (3) the thickness of the GaSe crystal affects these changings directly. Thus, this detection method in fact results in some pulse distortions, making the detected pulse red-shift of the central frequency and narrowing of the bandwidth. These are similar to the terahertz pulse distortions induced by the EOS detection<sup>[24]</sup> and the air coherent detection<sup>[25]</sup> reported before.

Then, the central wavelength of the few-cycle laser pulse is changed to 3  $\mu\text{m}$ . The results are plotted with time waveforms in Fig. 4(a) and frequency spectra in Fig. 4(b). The similar phenomena are observed from the Fig. 4 but with obvious changings of the spectra when the thickness of the GaSe crystal is 20  $\mu\text{m}$  (as the red line shown in Fig. 4(b)). This is because the central frequency of the few-cycle laser pulse is changed when its wavelength is changed. The frequency response is not flat in the bandwidth of the few-cycle laser pulse, therefore some frequency components are enlarged while some are reduced. Consequently, this makes the detection accuracy is also a function of the laser wavelength. Thus, it is necessary to carry out more calculations to find the relationships between the shift of the central frequency and the changing of the bandwidth depending on the laser wavelength.

In order to ensure the accuracy of the measurement of EOS technique with the GaSe crystal, the thickness of the GaSe is kept at 10  $\mu\text{m}$ . Change the wavelength of the few-cycle laser pulse from 2  $\mu\text{m}$  to 10  $\mu\text{m}$  (the central frequency 150 THz~30 THz), and then calculate (a) the shift of the central frequency and (b) the changing of the bandwidth, as shown in Fig. 5. It is found that: (1) when the laser wavelengths increase (its frequency decreases), these changings decrease gradually since the frequency response curve are flatter in the low frequency region; (2) for the 2- $\mu\text{m}$  laser pulse, its red-shift of the central frequency is 25 THz, while for the 10- $\mu\text{m}$  laser this shift can be neglected; and (3) the narrowing of the bandwidth presents similar property. Thus, the EOS detection technique with 10  $\mu\text{m}$  GaSe crystal as a sensor can measure the waveform of the mid-infrared pulse with some small pulse distortions.

When the GaSe crystal is changed to 20  $\mu\text{m}$  thick (or even thicker), the EOS detection will generate more dips in the frequency spectra and great changings in the time waveforms because there are more dips in the frequency response curves, as shown in the Fig. 2(b). Therefore, the thickness of GaSe crystal as a sensor in the mid-infrared band should not be larger than 10  $\mu\text{m}$ .

The calculations above mainly deal with the few-cycle laser pulse with 1.5 cycle in an envelope. It is found similar results when the laser pulse has 2 and 3 cycles in an envelope. Therefore, these studies have common conclusions for the mid-infrared few-cycle laser pulse detection by the EOS technique. Same method can be used to investigate the pulse distortions when the wavelength of the probe laser is changed. For example, the Er-doped fiber laser system usually offers femtosecond laser pulses with a wavelength of 1550 nm.

The mid-infrared few-cycle laser pulse has advantages in many areas of the high field physics, and its precise measurement will benefit these applications. The EOS detection measures the time waveform the pulse field, which is the most advantage comparing with the spectrometry measurement. The EOS with a thin GaSe crystal can work well in such bandwidth although with some small pulse distortions, such as a red-shift of the central fre-



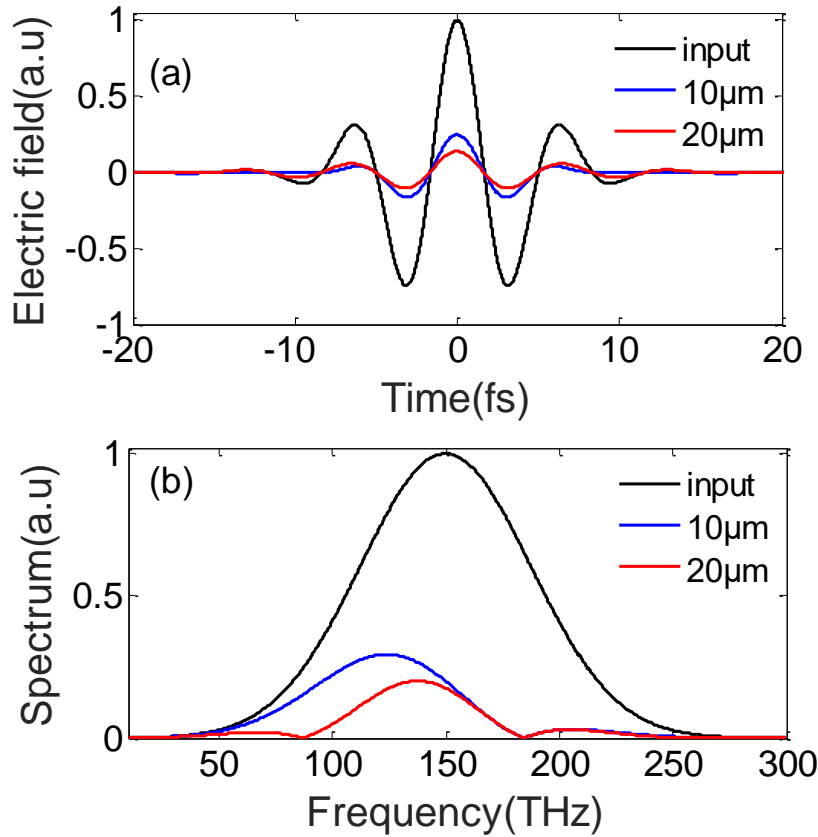


Fig. 3 The pulse distortions of few-cycle laser pulse with a wavelength of  $2\ \mu\text{m}$  induced by the EOS with different thickness. (a) shows the time waveforms, and (b) shows their frequency spectra, respectively.

图3 波长为  $2\ \mu\text{m}$  的少周期激光脉冲经过使用不同厚度的晶体电光取样探测后的脉冲畸变。(a)为时域波形变化,(b)为其对应的频谱变化。

quency and narrowing of the bandwidth.

#### 4 Discussions

Several groups have reported the measurement of time waveforms of the near- and mid-infrared few-cycle laser pulses with EOS detection technology. The frequency spectra through Fourier transform of the time waveforms are compared with the measurement results from the spectrometer, showing some differences in the central frequency and bandwidth. Here, a summary of their experimental parameters and measurement results are listed in the Table 1. We can see that the GaSe crystal is mainly used in the mid-infrared band while the BBO is in the near-infrared band. Our calculations in the Fig. 5 give the details of the changings of central frequency and bandwidth with a thin GaSe crystal.

Although the EOS detection technology has been used in the terahertz science for more than a decade, its applications in the few-cycle laser pulse measurement will bring obvious advantage that the phase of the laser pulse can be obtained directly. Thus, the EOS technology might be a key one in the few-cycle laser pulse since it has simple experimental schematic. The detection bandwidth of this technology with different crystals and probe laser pulses are different. These two parameters determine the frequency performance of the measurement.

Some different crystals (such as  $\text{LiNbO}_3$ ) might be used as EO crystal in the near- and mid-infrared band with good performance.

#### 5 Conclusions

In conclusion, the performance of the EOS detection technology with the thin GaSe crystal in the measurement of the mid-infrared few-cycle laser pulse are studied with numerical calculations. The frequency response and band limitation of the EOS with different thickness of GaSe sensor are given in the paper. Then, the shift of the central frequency and the changing of the bandwidth of the few-cycle laser induced by the EOS detection are investigated in detail. It is found this method induces a red-shift of the central frequency and narrowing of the bandwidth during the detection. These changings decrease when the laser wavelength increase from  $2\ \mu\text{m}$  to  $10\ \mu\text{m}$  with a  $10\text{-}\mu\text{m}$  thick GaSe crystal. Especially the pulse distortions can be neglected when the laser wavelength is longer than  $4\ \mu\text{m}$ . Therefore, the EOS detection technique with such thin GaSe crystal can work well in this bandwidth. This work offers a reference for the application of the EOS technique in the mid-infrared band and helps to estimate the accuracy of the experimental measurement.

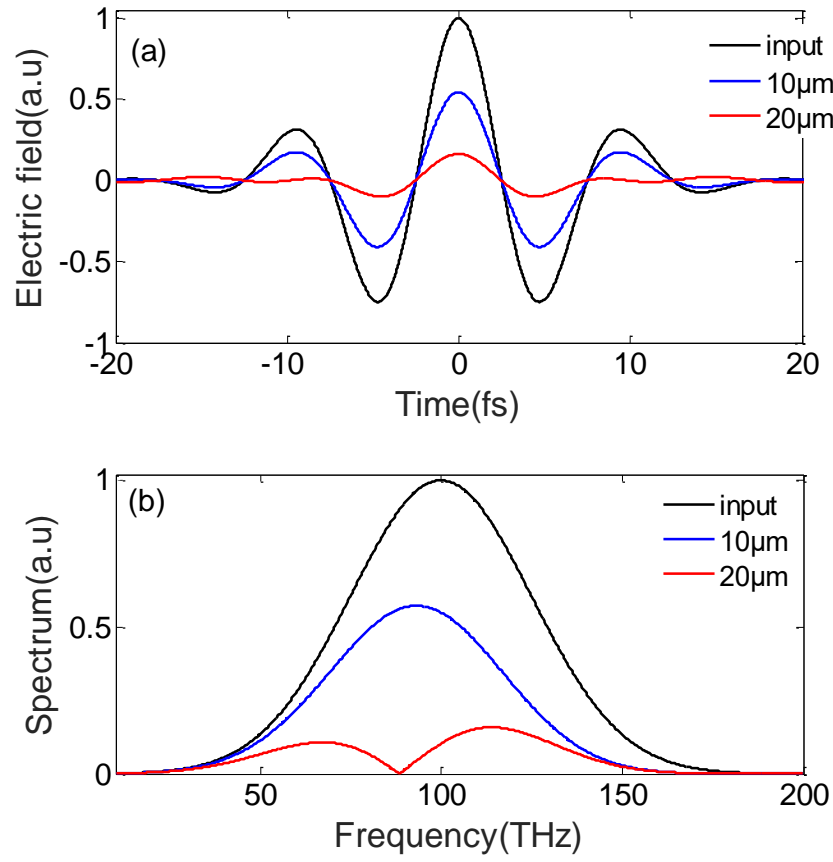


Fig. 4 The pulse distortions of few-cycle laser pulse with a wavelength of  $3 \mu\text{m}$  induced by the EOS with different thickness. (a) shows the time waveforms, and (b) shows their frequency spectra.

图4 波长为  $3 \mu\text{m}$  的少周期激光脉冲经过使用不同厚度的晶体电光取样探测后的脉冲畸变。(a)为时域波形变化,(b)为其对应的频谱变化。

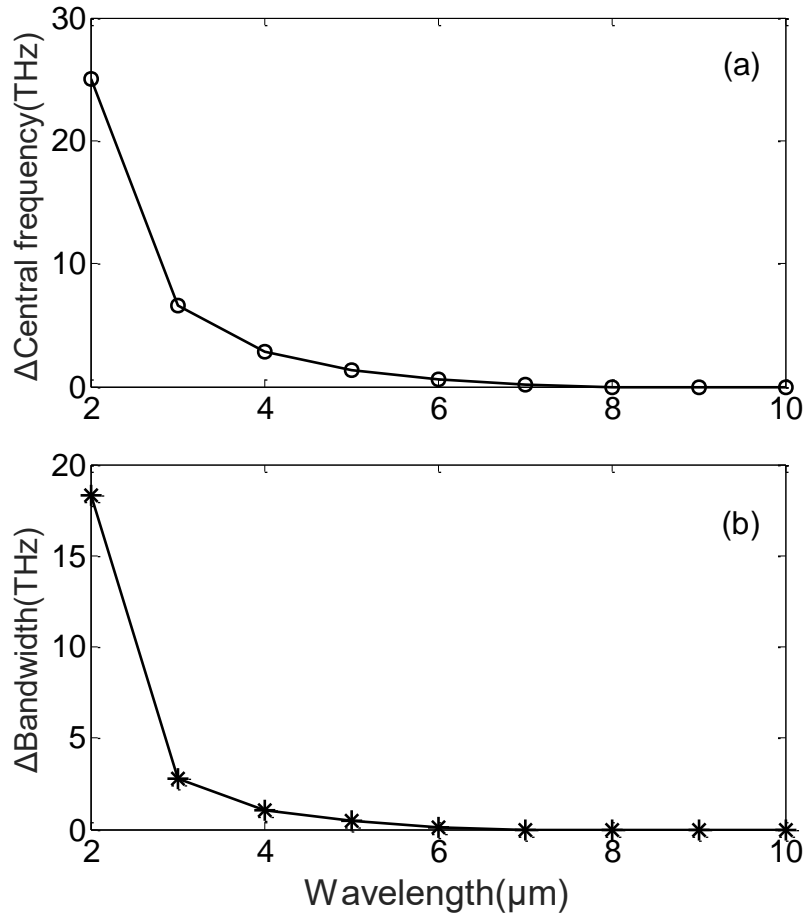


Fig. 5 The changing of (a) the central frequency and (b) the bandwidth depending on the wavelength of the few-cycle laser pulse from 2  $\mu\text{m}$  to 10  $\mu\text{m}$ .

图5 波长从2  $\mu\text{m}$ 到10  $\mu\text{m}$ 的少周期激光脉冲(a)中心频率和(b)带宽的变化规律。

**Table 1 Summary of experimental parameters and results of EOS applications in the near- and mid-infrared band.**

EO crystal	EO crystal Thickness ( $\mu\text{m}$ )	Probe laser wavelength (nm)	Probe laser duration (FWHM, fs)	Measurement wavelength ( $\mu\text{m}$ )	Ref.
GaSe	37	790	10	~20	[8]
GaSe	30	780	10	~8.88	[9]
GaSe	250, 500	1050	15	8–12	[11]
GaSe	180	1030	10	~8.6	[12]
GaSe	100	1030	16	9	[13]
BBO	100	500–950	4	1.2	[26]
BBO	5	300–600	2.8 $\pm$ 0.1	0.6–3	[27]
BBO	100	0.5–1	4	2	[28]
GaSe	250	1560	11	6.6–11	[29]

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