Invited Paper

Terahertz pulse manipulation based on difference frequency generation

Dehua Li [1, 2*], Jianjun Ma [3], Wei Zhou [1, 2], Peng Zhao [1, 2], Junxiang Yu [1, 2], and Zhaoxin Li [1, 2]
[1] Terahertz Science and Technology Research Center, Shandong University of Science and Technology, 579 Qianwangang Rd, Huangdao, Qingdao, 266590, China
[2] Qingdao Key Laboratory of Terahertz Technology, Qingdao, 579 Qianwangang Rd, Huangdao, 266590, China
[3] Department of Physics, New Jersey Institute of Technology, 322 King Blvd., Newark, New Jersey, 07102, USA
[*1] Email: jcbwl@sdust.edu.cn

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Abstract: This paper presents a method of terahertz (THz) pulse shaping in which both the amplitude and polarity of the THz pulse can be manipulated. A pair of temporally separated and orthogonally polarized collinear propagating femtosecond pulses with the same central frequencies are focused onto a (110) oriented ZnTe crystal. By adjusting the relative time delay between the pulses, the amplitude and polarity of the generated THz pulse based on difference frequency generation (DFG) can be controlled. Theoretical derivation and simulation based on one-dimensional propagation equation of the THz wave have been carried out under simplified conditions of perfect phase matching, plane-wave approximation and no absorption. Considering the crystal absorption and phase mismatching condition in the DFG process, THz power spectrums from ZnTe, GaAs, GaP, InP, and CdTe crystals are conducted and compared. Besides, the crystal length dependence of THz waveform is discussed.

Keywords: Terahertz pulse, Pulse manipulation, Difference frequency generation, Crystal orientation

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1. Introduction

Data processing based on terahertz (THz) technologies has a strong potential for a variety of interesting applications such as the exciton transitions of quantum systems [1, 2], biological and medical imaging [3, 4], rotational and vibrational molecular dynamics [5, 6], high-speed optical signal processing [7, 8], ultra-fast wireless digital interconnects [9], and so on. Advantages of waveform processing at the THz frequency include ultra-high bandwidth, high security and low attenuation. Recent major advances in femtosecond pulse manipulator have opened promising new directions in THz manipulation techniques. The femtosecond pulse shaping technique developed as a result has provided a powerful tool for the THz manipulation and the knowledge derived from their study has greatly advanced the fundamental understanding of THz waveform manipulation. However, there are still considerable limitations which need to be overcome and new methodologies need to be explored for some breakthroughs being achieved in this field.

In order to realize arbitrary THz waveform, optical nonlinear processing is seen as one of the

key technologies and many methods are investigated, such as optical rectification (OR), DFG. In this paper, a new scheme for THz temporal profile manipulation is reported, which is based on difference frequency mixing of a pair of temporally separated and orthogonally polarized pulses with the same central frequencies and femtosecond duration. Theoretical analysis shows that this is an efficient method to control the THz pulse amplitude and polarity by changing the relative time delay between the polarized femtosecond pulses in a (110)-cut ZnTe crystal. Nevertheless, to investigate the effectiveness of this method, numerical calculation and simple simulation using one-dimensional equation of the THz field are carried out with simplified conditions that phase matching is perfect, and absorption and other third-order nonlinear effects are neglected. In order to enhance the THz pulse shaping efficiency, GaAs, GaP, InP, and CdTe crystals, which possess the same cubic structure and -43*m* point group as ZnTe, are chosen as candidate crystals for THz pulse generation. Taking into account of the crystal absorption and phase mismatching condition, THz power spectrums from these crystals are presented and the performances of these crystals in the pulse shaping process are compared.

2. Theoretical calculation

To describe the evolution of the THz pulse in crystal, Maxwell's equations are used and the one-dimensional propagation equation is derived as [10]

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \mu_0 \varepsilon \frac{\partial^2 E(z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 P(z,t)}{\partial t^2} \tag{1}$$

where $E(z,t)$ stands for the electric field of the generated THz waves, and $P(z,t)$ refers to the nonlinear polarization as a driving force for the nonlinear process. ε is the dielectric constant, and μ_0 denotes the free space magnetic permeability.

Considering the slowly-varying amplitude approximation which is widely adopted in nonlinear optics [11], Eq. (1) can be simplified to

$$\frac{\partial E(z,t)}{\partial z} = i \frac{\mu_0}{2k_\Omega} \frac{\partial^2 P(t)}{\partial t^2} \exp(i\Delta k z) \tag{2}$$

where $k_\Omega = \Omega n_T / c$ is the wavevector of generated THz wave at frequency Ω, and Δk is due to the phase mismatching. The electric field can be derived by numerical integration of Eq. (2) as

$$E(z,t) = i \frac{\mu_0 z}{2k_\Omega} \frac{\partial^2 P(t)}{\partial t^2} \exp(i\Delta k z/2) \sin c(\Delta k z/2) \tag{3}$$

For the temporally separated optical pulses with identical intensity Gaussian temporal

envelope, femtosecond pulse duration and infinite plane wavefront are assumed in the theory. The THz electric field can be

$$E_1(z,t) = \frac{1}{2}\left[E_0 \exp(-t^2/\tau^2)\exp(i\omega_0 t) + c.c. \right]\exp(-ikz) \tag{4}$$

where, ω_0 is the central frequency, and τ stands for the pulse width defined by the full width at half maximum of the intensity. The second (time delayed) pulse is

$$E_2(z,t) = \frac{1}{2}\left\{ E_0 \exp\left[-(t+\Delta t)^2/\tau^2 \right]\exp\left[i\omega_0 (t+\Delta t) \right] + c.c. \right\}\exp(-ikz) \tag{5}$$

with Δt standing for the relative time delay. One (110) oriented ZnTe crystal is used as EO crystal resulting from its virtual transparency in the frequency region of 0.1-3 *THz* [12] and perfect phase matching conditions for THz wave generation [13].

As shown in Fig.1, these two orthogonal polarized pulses are coincident on the ZnTe crystal, with $E_1(t) \rightarrow [\bar{1}10]$, $E_2(t) \rightarrow [001]$. Previously studies has proved that no THz is generated through OR when the linearly polarized optical field is aligned along the [001] orientation [14].The pump field in the *x'y'z'* coordinate can be expressed as

$$\begin{aligned} \mathbf{E}'(t) &= \left[E_x'(t) \quad E_y'(t) \quad E_z'(t) \right] \\ &= \left[-E_1(t)/\sqrt{2} \quad E_1(t)/\sqrt{2} \quad E_2(t) \right] \end{aligned} \tag{6}$$

Then the nonlinear polarization is

$$\begin{bmatrix} P_x'(t) \\ P_y'(t) \\ P_z'(t) \end{bmatrix} = \begin{bmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{bmatrix} \begin{bmatrix} E_x'^2 \\ E_y'^2 \\ E_z'^2 \\ 2E_y'E_z' \\ 2E_x'E_z' \\ 2E_x'E_y' \end{bmatrix} \tag{7}$$

Rotating the *x'y'z'* coordinate to the *xyz* coordinate as illustrated in Fig. 1, the nonlinear polarization in the *xyz* coordinate becomes

$$\begin{bmatrix} P_x(t) \\ P_y(t) \\ P_z(t) \end{bmatrix} = d_{14} \begin{bmatrix} -E_1^2(t) \\ -2E_1(t)E_2(t) \\ 0 \end{bmatrix} \tag{8}$$

Based on Eqs. (3) and (8), THz electric field in time domain can be derived as

$$\begin{aligned} E_x(z,t) &= i\frac{\mu_0 z}{2k}\frac{\partial^2 P_x(t)}{\partial t^2}\exp(i\Delta kz/2)\sin c(\Delta kz/2) \\ &= -i\frac{\mu_0 z}{4k}d_{eff}E_0^2\exp(-2t^2/\tau^2)\times\frac{4(4t^2-\tau^2)}{\tau^4}\times\exp(i\Delta kz/2)\sin c(\Delta kz/2) \\ &\quad -i\frac{\mu_0 z}{4k}d_{eff}E_0^2\exp(-2t^2/\tau^2)\times\left[4\left(\frac{4t^2-\tau^2}{\tau^4}-\omega_0^2\right)\cos(2\omega_0 t)+4\omega_0\times\frac{4t}{\tau^2}\sin(2\omega_0 t)\right] \\ &\quad \times\exp(i\Delta kz/2)\sin c(\Delta kz/2) \end{aligned} \tag{9}$$

$$\begin{aligned} E_y(z,t) &= i\frac{\mu_0 z}{2k}\frac{\partial^2 P_y(t)}{\partial t^2}\exp(i\Delta kz/2)\sin c(\Delta kz/2) \\ &= -i\frac{\mu_0 z}{2k}d_{eff}E_0^2\exp\left[-\frac{t^2+(t+\Delta t)^2}{\tau^2}\right]\times\frac{4\left[(2t+\Delta t)^2-\tau^2\right]}{\tau^4} \\ &\quad \times\cos(\omega_0\Delta t)\exp(i\Delta kz/2)\sin c(\Delta kz/2) \\ &\quad -i\frac{\mu_0 z}{2k}d_{eff}E_0^2\exp\left[-\frac{t^2+(t+\Delta t)^2}{\tau^2}\right]\times\exp(i\Delta kz/2)\sin c(\Delta kz/2) \\ &\quad \times\left\{4\left[\frac{(2t+\Delta t)^2-\tau^2}{\tau^4}-\omega_0^2\right]\cos\left[\omega_0(2t+\Delta t)\right]+4\omega_0\frac{2(2t+\Delta t)}{\tau^2}\sin\omega_0(2t+\Delta t)\right\} \end{aligned} \tag{10}$$

Here, i implies that there is a phase delay of $\pi/2$, Δt corresponds to the relative time delay and the effective nonlinear optical coefficient d_{eff} is used to replace the tensor element d_{14} for the approximation. Both the second harmonic term and sum frequency term in Eqs. (13) and (14) can be filtered out by a silicon wafer in experiment. So the THz electric field E_{Tx} induced by OR in x direction and E_{Ty} induced by DFG in y direction could be obtained as

$$E_{Tx}(z,t) = -i\frac{\mu_0 z}{4k_\Omega}d_{eff}E_0^2\exp(-2t^2/\tau^2)\times\frac{4(4t^2-\tau^2)}{\tau^4}\exp(i\Delta kz/2)\sin c(\Delta kz/2) \tag{11}$$

$$E_{Ty}(z,t) = -i\frac{\mu_0 z}{2k_\Omega}d_{eff}E_0^2\exp\left[-\frac{t^2+(t+\Delta t)^2}{\tau^2}\right]\times\frac{4\left[(2t+\Delta t)^2-\tau^2\right]}{\tau^4}\cos(\omega_0\Delta t)\exp(i\Delta kz/2)\sin c(\Delta kz/2) \tag{12}$$

and Δk is the phase mismatch defined by

$$\Delta k = k_{\Omega} + k_{\omega} - k_{\Omega+\omega} \tag{13}$$

where k_{ω}, $k_{\Omega+\omega}$ are wavevectors of incident optical pulses at different frequencies respectively. c refers to the speed of light in vacuum, the refractive index n_T of the THz wave is assumed to be a constant (for ZnTe, n_T =3.17 [15]).

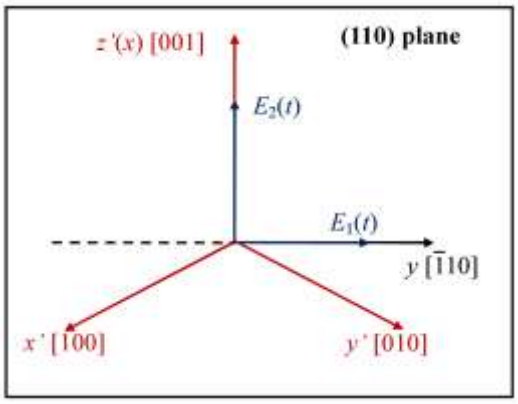

Fig. 1 Geometry of (110) oriented ZnTe crystal. (The plane of the page is the (110) plane of the crystal).

When Δk=0, Eqs. (11) and (12) can be simplified to

$$E_{Tx}(z,t) = -i\frac{\mu_0 z}{4k_{\Omega}} d_{eff} E_0^2 \exp(-2t^2/\tau^2) \times \frac{4(4t^2-\tau^2)}{\tau^4} \tag{14}$$

$$E_{Ty}(z,t) = -i\frac{\mu_0 z}{2k_{\Omega}} d_{eff} E_0^2 \exp\left[-\frac{t^2+(t+\Delta t)^2}{\tau^2}\right] \times \frac{4\left[(2t+\Delta t)^2-\tau^2\right]}{\tau^4} \cos(\omega_0 \Delta t) \tag{15}$$

The generation of THz waves through OR, as expressed in Eq. (14), has gained widespread acceptance and can be isolated by a wire grid polarizer. So THz pulses induced by DFG as shown in Eq. (15) could be obtained solely. Seeing from Fig. 2, the radiated THz wave induced by DFG has the same pulse width as the THz wave generated by OR, which confirms that there is THz wave generated from the crystal due to DFG.

According to Eq. (15), the amplitude and polarity of the generated THz wave can be manipulated by adjusting the relative time delay Δt between the temporally separated and orthogonally polarized pulses. Figure 3 shows the THz waveform versus to the relative time delay varying from Δt=0 ($\Delta\varphi$=0) to Δt=4/3 *fs* ($\Delta\varphi$=π) providing that the incident optical pulses have identical intensity, pulsewidth is 100 *fs* and central wavelength is 800 *nm*. The amplitude of the THz wave with inverse polarity is maximized for Δt=0 ($\Delta\varphi$=0) and Δt=4/3 *fs* ($\Delta\varphi$=π). In other words, the generated terahertz waveforms have the largest amplitude and inverse polarity when the phase shift between the two incident pulses is 0 and π.

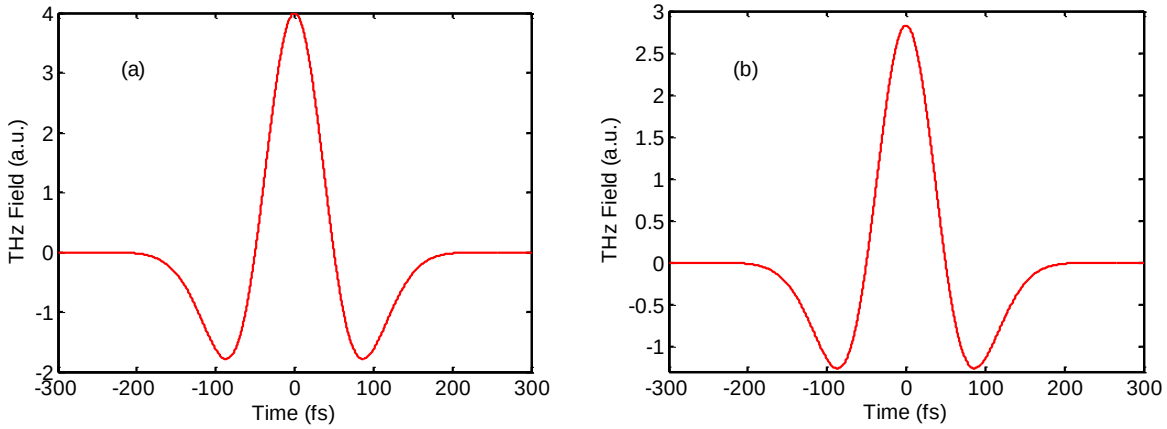

Fig. 2 Curves of terahertz waves generated from optical rectification(a) and difference frequency mixing (b) when Δt=1/3 *fs*.

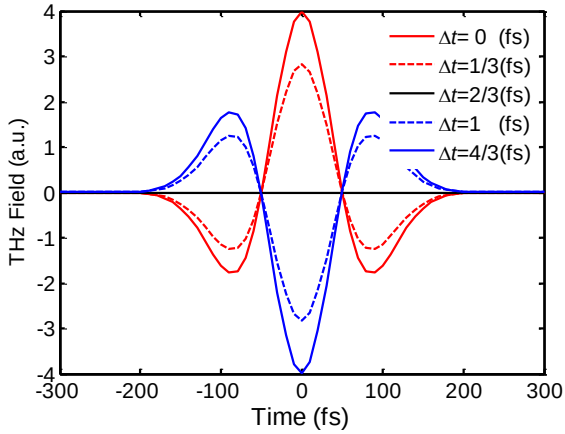

Fig. 3 Curves of terahertz waveforms when two femtosecond pulses are incident with the relative time delay between them ranging from Δt=0 (Δφ=0) to Δt=4/3 *fs* (Δφ=π).

To improve the THz pulse shaping efficiency in this method, other zinc blende crystals (such as GaAs, GaP, In P, and CdTe) with larger nonlinear coefficient are chosen as the candidate crystals. These crystals show strong coupling between photons and the transverse optical (TO) phonon resonances in the Reststrahlens's band, which results in the absorption and dispersion for the pulses propagating in the crystals [16]. From the dielectric response of a harmonic oscillator for the TO-phonon line applied to these zinc blende crystals, the crystal absorption in the THz frequency, which has great influence on the spectral bandwidth of generated THz pulse, can be expressed as [17, 18]

$$\alpha_T(f)=\frac{4\pi f}{c}\,\mathrm{Im}\left(\varepsilon_{el}+\frac{\varepsilon_{st} f_{TO}^2}{f_{TO}^2-f^2+2i\gamma f}\right) \tag{16}$$

where, f_{TO} refers to the strong TO-phonon resonance of the crystals at room temperature. f is the THz frequency, ε_{st} and γ are the oscillator strength and the line width of the TO-phonon mode,

respectively. c refers to the velocity of light in vacuum.

Considering the crystal dispersion for the optical and THz wave, the phase mismatching is conducted from the dielectric response function of the harmonic oscillator in crystals

$$\Delta k = \frac{\omega_T \left| n_{opt}(\lambda) - \lambda \frac{dn_{opt}(\lambda)}{d\lambda}\bigg|_{\lambda_{opt}} - \mathrm{Re}\left(\varepsilon_{el} + \frac{\varepsilon_{st} f_{TO}^2}{f_{TO}^2 - f^2 + 2i\gamma f} \right) \right|}{c} \tag{17}$$

Where ω_T is the angular frequency of THz waves and λ is the optical wavelength. The dispersion property in the optical refractive index $n_{opt}(\lambda)$ can be expressed as [19]

$$n_{opt}(\lambda) = \sqrt{A + \frac{B\lambda^2}{\lambda^2 - C^2}} \tag{18}$$

Here, A, B and C^2 are parameters related to the crystal properties.

The THz wave spectrum can be obtained by the Fourier Transform of THz propagation equation as

$$E(z,\omega_T) = -i\frac{\mu_0 d_{eff}\omega_T c E_0^2 \tau}{2\pi} \exp\left(-\frac{\Delta t^2}{2\tau^2} - \frac{\tau^2\omega_T^2}{8} \right) \exp(i\omega_T \Delta t) \frac{\exp\left[\left(-\alpha_L + i\Delta k \right) z \right] - \exp\left(-\frac{\alpha_T}{2} z \right)}{\left(-\alpha_L + \frac{\alpha_T}{2} + i\Delta k \right)} \cos(\omega_0 \Delta t) \tag{19}$$

Herein ω_0 is the central frequency of the incident pump pulses, and τ donates the pulsewidth at the $1/e^2$ level in intensity. α_L and α_T denote crystal absorption for the pump and terahertz wave, and Δt corresponds to the temporal separation.

Using the parameters shown in Table 1, the calculated power spectrums of the THz waves in 0.1 *mm* crystals by Eq. (19) are presented in Fig. 4(a). The GaP crystal owns the widest bandwidth in the THz spectrum, which makes it the best choice for THz pulse shaping. Fig. 4(b) shows the THz power spectrum in 0.5 *mm* crystals. It can be seen that the bandwidth of the first peak of the spectrum is becoming narrower than in Fig. 4(a) because of the phase mismatching and crystal absorption, but the peak of the power spectrum is much higher in the 0.5 *mm* crystals owning to the higher conversion efficiency.

The ratio between the peak of the oscillatory tail with the first dominant peak of the power spectrum reflects the radiation efficiency of THz waves. In the Fig. 4 (a) and (b), that ratio is becoming larger in the longer GaAs and GaP crystals which attributes to the phase mismatching of the optical and THz waves. However, the same ratio could be seen in 0.1 *mm* and 0.5 *mm* ZnTe (1:4) for its perfect phase matching condition with 0.8 μm optical wavelength, which could

compensate its smaller nonlinear coefficients than in Gap and GaAs crystals. So, the ZnTe crystal shows better performance.

Tab. 1 Parameters of the dielectric respond function of crystals [17-26]

	ZnTe	GaAs	GaP	InP	CdTe
ε_{el}	7.4	11.0	8.7	9.61	7.1
ε_{st}	2.7	2.0	1.8	2.8	3.1
f_{T0} (THz)	5.3	8.0	10.98	9.0	4.23
γ (THz)	0.045	0.035	0.01	0.05	0.1
A	4.27	8.95	2.680	7.255	5.68
B	3.01	2.054	6.40	2.316	1.53
C^2 (μm^2)	0.142	0.390	0.09	0.3922	0.366
d_{14} (pm/V)	90	368.7	147-220	143.5	108

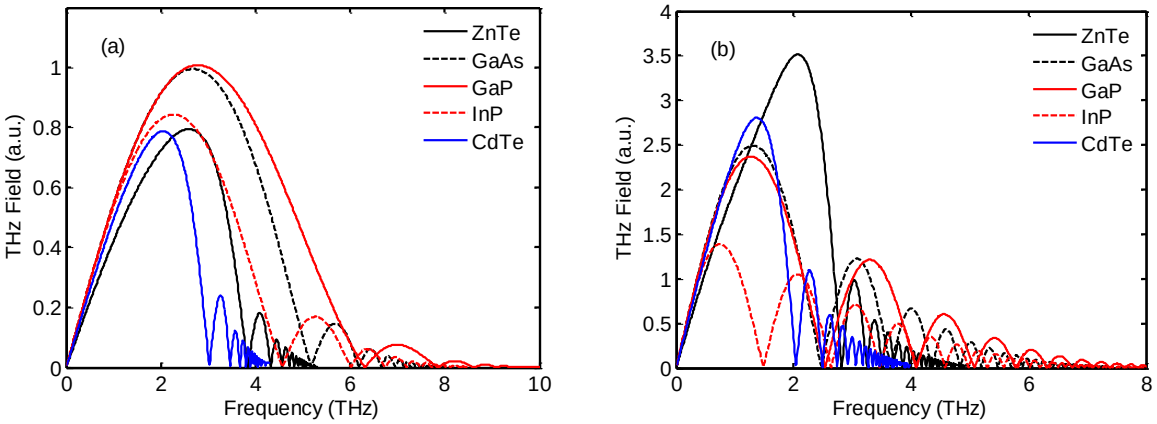

Fig. 4 Calculated Power spectrum for five crystals of 0.1 *mm* (a) and 0.5 *mm* (b) lengths with pulse duration of 100-*fs* and central wavelength at 0.8 μm in ZnTe, 1.56 μm in GaAs and GaP crystal.

The crystal length influences the amplitude of the THz wave propagating in it due to phase mismatching and crystal absorption. Based on Eq. (19), the spectral shapes of the THz field in ZnTe are mapped in Fig. 5 which illustrate that the THz spectrum is sensitive to the crystal length. The spectral amplitude at the dominant peak is proportional to the crystal length, while the spectral width is inversely proportional to the crystal length because of phase mismatching, which agrees well with the results in [27].

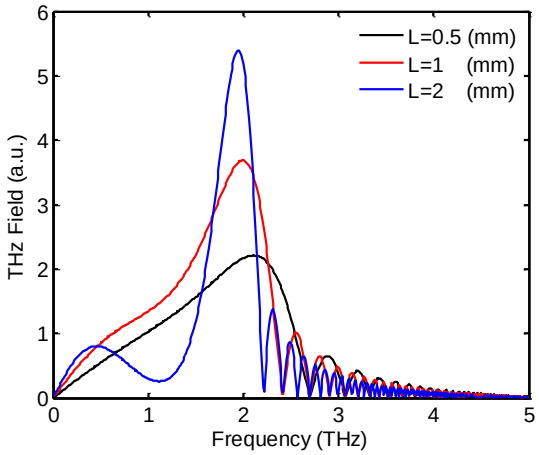

Fig. 5 THz power spectrum with respect to the crystal length in ZnTe

3. Conclusions

The theoretical analysis based on one-dimensional propagation equation of the THz waves and the computer simulations using Matlab are carried out, and the results show that amplitude and polarity of generated THz waves can be manipulated using DFG of a pair of temporally separated and orthogonally polarized shaped femtosecond pulses with the same frequencies. For the sake of simplicity of the calculations, the perfect phase matching and no loss are assumed. Under such conditions, manipulation of amplitude and polarity of the generated THz waves can be realized by adjusting the relative time delay between the two orthogonal polarized incident pump pulses. Four different zinc blende crystals with larger nonlinear coefficients than ZnTe are used to compare the THz pulse shaping efficiency. Thin (0.1 *mm*) GaP crystal shows good performance for broadband THz pulse generation in the pulse shaping process, while thick (0.5 *mm*) ZnTe crystal owns higher THz spectral peak with broader bandwidth compared with GaAs and GaP crystals. Due to absorption and phase mismatching between the THz wave and pump lasers, increasing the crystal length results in narrower THz spectral bandwidth and higher peak spectral amplitude.

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