Theoretical research on cascaded terahertz difference-frequency generation based on sphalerite crystals*

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A theoretical model of cascaded terahertz (THz) difference-frequency generation is established based on one-dimensional coupled-wave equations. The relationships between sphalerite crystals' wave vector mismatches and difference-frequency pump waves are analyzed. To produce terahertz wave with the frequency of 1.5 THz, 80-order cascaded difference-frequency is applied. By introducing crystal absorption, we calculate the optimum crystal length and pump frequency under actual circumstances. It is found that Stokes waves dominate the terahertz waves output in cascaded progress, and cascaded difference-frequency can increase the photon conversion efficiency obviously.

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The difference-frequency terahertz (THz) source attracts great research interest, because it is simple, flexible and has wide tuning range without threshold[1-3]. Not only the traditional anisotropic nonlinear crystal, such as $ZnGeP_2$ and GaSe[4-6], but also 4-dimethylamino-N-methyl-4-stilbazolium tosylate (DAST), N-benzyl-2-methyl-4-nitroaniline (BNA) and other new organic crystals[7-9] are widely used for difference-frequency THz sources. Sphalerite crystals (GaAs, GaP, ZnTe, CdTe, etc.) are widely used in the study of narrow-band THz sources for their low absorption coefficients and large nonlinear coefficients[10-12]. However, the conversion efficiency of THz difference-frequency generation is low, which is typically less than 10^{-5}[4]. Over the years, people have been working for improving the conversion efficiency[13-17].

In this paper, we study the influence of wave vector mismatch and sphalerite crystals' absorption on cascaded difference-frequency generating THz wave by using GaAs, GaP, ZnTe,CdTe as example. The optimal crystal length, the optimal pump light frequency and the corresponding maximum value of photon flux are calculated in cascaded difference-frequency THz generation. The results provide a theoretical basis and experimental parameters for cascaded difference-frequency THz generation based on sphalerite crystals.

There is no difference between the second-order nonlinear difference-frequency THz generation and the usual difference-frequency near-infrared light generation in theory. Two near-infrared laser beams with frequencies of ω_1 and ω_2 are allowed to beat together in a nonlinear optical medium, and a light wave with frequency of ω_T is generated, where

$$\omega_T=\omega_1-\omega_2 \ . \quad (1)$$

If the light waves with frequencies of ω_2 and ω_T meet the phase matching condition and their intensities are strong enough, they will also generate a wave with frequency of ω_3 by difference-frequency. Similarly, ω_3 and ω_T can continue to have difference frequency when they satisfy the phase matching condition. If this process continues, a series of light waves can be generated through the cascaded effect, while the energy of ω_T always increases. According to this theory, we can greatly enhance the conversion efficiency from light waves to THz waves.

One-dimensional coupled-wave equations of the optical and THz waves are derived using the wave equation[17]:

$$\frac{\partial^2 E(z,t)}{\partial z^2}-\varepsilon\mu\frac{\partial^2 E(z,t)}{\partial t^2}-\mu\sigma\frac{\partial E(z,t)}{\partial t}=\mu\frac{\partial^2 P(z,t)}{\partial t^2} , \quad (2)$$

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where $E(z,t) = E_L(z,t) + E_T(z,t)$ is the electric field composed of an optical frequency component of $E_L(z,t)$ and a THz frequency component of $E_T(z,t)$. ε is the dielectric constant, μ is the magnetic permeability, which is assumed to be in vacuum and independent of frequency, σ is the conductivity, which is related to the absorption coefficient, and $P(z,t)=2d_{\mathrm{eff}}\times[E_L(z,t)]^2$ is the nonlinear polarization where d_{eff} is the effective nonlinear coefficient. The electric fields of the pump light are expressed using a slowly varying envelope $e_L(z,t)$ as

$$E_L(z,t)=\frac{1}{2}e_L(z,t)\exp(-\mathrm{i}\omega_0 t)+c.c. \quad (3)$$

Move the multiplier $\exp(-\mathrm{i}\omega_0 t)$ to the other side, so

$$e_L(z,t)=\int E_L(z,\omega)\exp[\mathrm{i}k(\omega)z+\mathrm{i}\omega_0 t-\mathrm{i}\omega t]\mathrm{d}\omega . \quad (4)$$

Because the impact of dispersion is relatively small in the vicinity of phase matching conditions, we can obtain

$$k(\omega_0)+k(\omega)=k(\omega_0+\omega) . \quad (5)$$

Therefore,

$$e_L(z,t)=\int E_L(z,\omega)\exp[\mathrm{i}k(\omega_0+\omega)z-\mathrm{i}\omega t]\mathrm{d}\omega , \quad (6)$$

and then,

$$E_L(z,t)=\int E_L(z,\omega)\exp[\mathrm{i}k(\omega)z-\mathrm{i}\omega t]\mathrm{d}\omega , \quad (7)$$

$$E_T(z,t)=\int E_T(z,\omega_T)\exp[\mathrm{i}k(\omega_T)z-\mathrm{i}\omega_T t]\mathrm{d}\omega_T . \quad (8)$$

From Eqs.(2), (7) and (8), we can obtain one-dimensional coupled-wave equations as follows,

$$\frac{\partial}{\partial z}E_L(z,\omega)=-\frac{\alpha_L}{2}E_L(z,\omega_L)+\mathrm{i}\frac{2\mu d_{\mathrm{eff}}(\omega+\omega_T)^2}{k(\omega+\omega_T)}\times \int E_L(z,\omega+\omega_T)E_T(z,\omega)\mathrm{e}^{\mathrm{i}\Delta k_L z}\mathrm{d}\omega_T . \quad (9)$$

Similarly,

$$\frac{\partial}{\partial z}E_T(z,\omega)=-\frac{\alpha_T}{2}E_T(z,\omega)+\mathrm{i}\frac{\mu d_{\mathrm{eff}}\omega_T{}^2}{2k\omega_T}\times \int E_L(z,\omega+\omega_T)E_L^*(z,\omega)\mathrm{e}^{\mathrm{i}\Delta k_T z}\mathrm{d}\omega , \quad (10)$$

where α_L and α_T are crystal absorption coefficients for pump waves and THz waves, respectively.

According to Ref.[18], difference-frequency pump wavelengths are different for different crystals under the complete phase-matching conditions. If we want to realize the THz waves output with the frequency of f_T, the frequencies of two pump waves can be obtained under the complete phase-matching conditions. Ref.[18] gives the dispersion equations of the different crystals. The wave vector mismatch versus the pump frequency (corresponding to the wavelength of λ_{p2}) in the case of 1.5 THz wave output is shown in Fig.1. We can find that for the different crystals of GaAs, GaP, ZnTe and CdTe, the optimal pump frequencies are 217.5 THz, 302.5 THz , 270 THz and 280.5 THz, respectively, where the wave vector mismatch is zero approximately. In the cascaded case, the wave vector mismatch of the front and back orders is small. In the same circumstance, the wave vector mismatch of ZnTe is the smallest, and that of CdTe is the biggest.

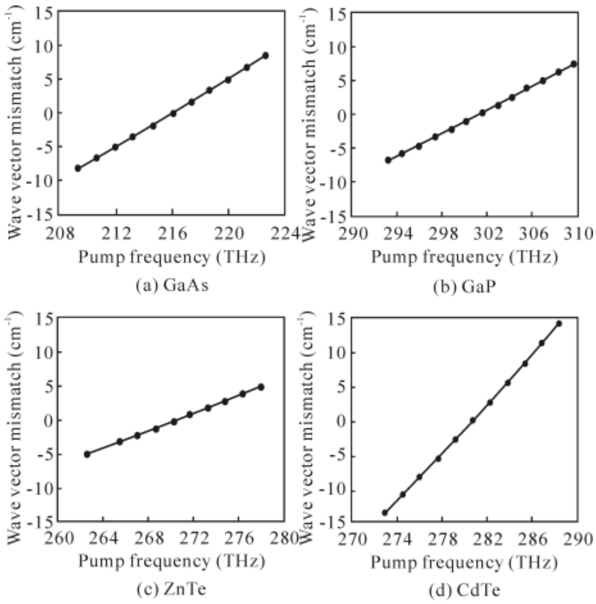

Fig.1 Wave vector mismatch versus pump frequency

In this part, we consider the ideal case of complete wave vector matching and no absorption, i.e., $\Delta k = 0$, $\alpha=0$. Suppose that the incident pump waves' indensity is $I = 50$ MW/mm^2 and the length of crystals is 15 mm. Selecting 80-order cascaded difference-frequency approximation, and then we obtain the surface plot which shows the development of Stokes and anti-Stokes cascades for generation of 1.5 THz radiation, as shown in Fig.2. The photon flux can be expressed as

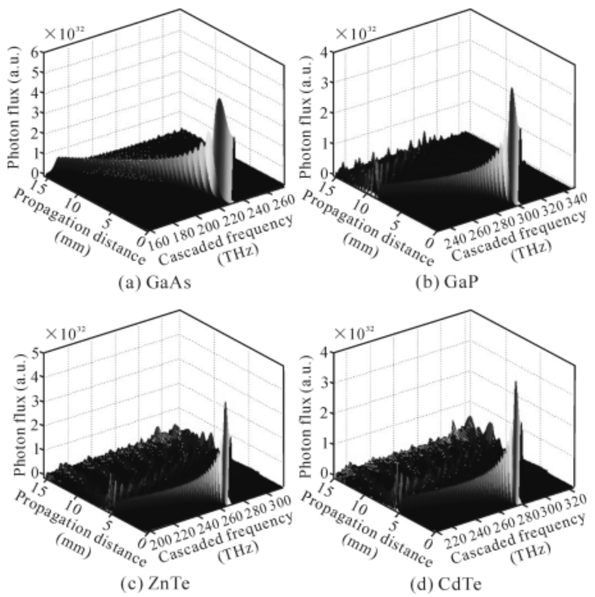

Fig.2 Developments of Stokes and anti-Stokes cascades for generation of 1.5 THz radiation

$$F_i = \frac{|A_i|^2}{2\eta}\sqrt{\frac{\varepsilon_0}{\mu_0}} \quad , \qquad (11)$$

where $A_i = \sqrt{\frac{n_i}{\omega_i}} E_i$. And Fig.3 shows the relation between the THz wave photon flux and propagation distance in the progress of cascaded difference-frequency.

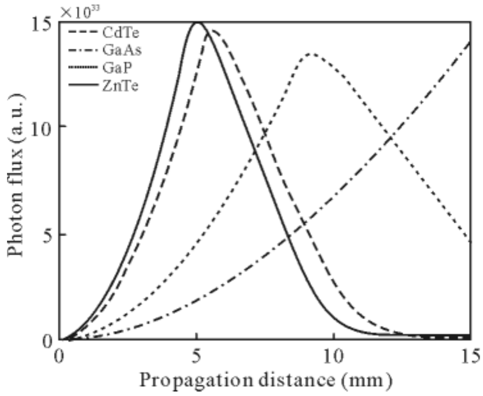

Fig.3 THz wave photon flux versus interaction length for generation of 1.5 THz radiation

According to the above analyses, the longer the propagation distances are, the better in the ideal case. However, only the situation in GaAs keeps with the rule. That is because we apply 80-order cascaded difference-frequency approximation. In this context, after completing 80-order cascaded difference frequency, sum-frequency starts on stage, and then the intensity of THz wave reduces, as shown in GaP, ZnTe and CdTe. When the propagation distance in GaP is greater than 8 mm, the intensity of THz wave becomes small. And the similar cases exsit in CdTe and ZnTe when the propagation distances are greater than 5 mm and 5.5 mm, respectively. From this perspective, the bigger the cascaded order is, the better in simulation. However, when we consider the wave vector mismatch and crystal absorption, the cascaded order and the optimal crystal length are not the bigger the better yet.

In order to comparatively analyze all the cases, the relationships between THz photon flux and propagation distance in the different cases are shown in Fig.4(a)-(d).

As we know, the absorption coefficients of GaAs and GaP in both optical and THz ranges are relatively small. So in Fig.4(a) and (b), the slopes of the dashed lines are smaller than those of the dotted lines, and all of the slopes in the coordinate ranges are positive. However, the slopes of dashed lines for ZnTe and CdTe are negative when the propagation distance is greater than 3 mm approximately, as shown in Fig. 4(c) and (d). In order to obtain the greater THz photon flux, the absorption effect is the smaller the better.

In this paper, the wave vector mismatch is expressed as

$$\Delta k_i = k_i - k_{i+1} - k_{\mathrm{T}} \quad , \qquad (12)$$

where

$$k_i = 2\pi\left(\frac{n_i}{\lambda_i} - \frac{n_{i+1}}{\lambda_{i+1}} - \frac{n_{\mathrm{T}}}{\lambda_{\mathrm{T}}}\right) \quad . \qquad (13)$$

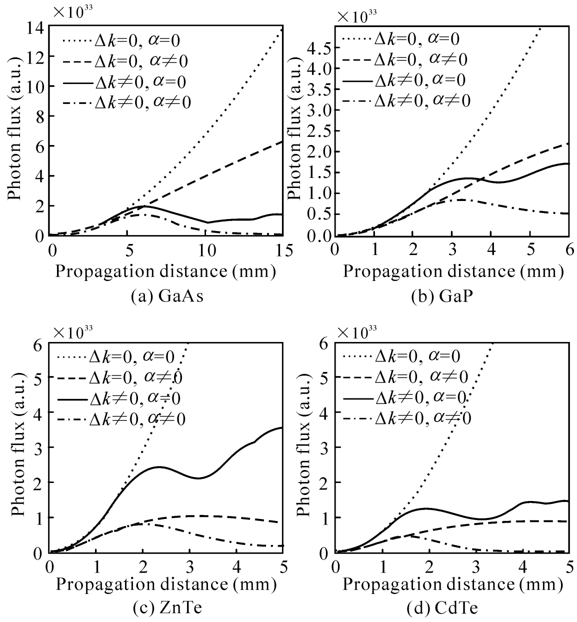

Fig.4 THz photon flux versus interaction length in GaAs, GaP, ZnTe and CdTe for different cases

The solid lines in Fig.4 denote the case of considering the wave vector mismatch only. We can see that the effect of the wave vector mismatch is obvious for cascaded difference frequency, and could not be neglected. In order to realize high-order cascaded difference frequency, it is necessary to compensate for mismatch.

We can see from the solid lines in Fig.4, every crystal has an optimal length with which can realize the maximum THz wave output when the pump light intensity and frequency are given. When inputting the two pump waves with frequencies of 219.5 THz and 218 THz, the optimal length of crystal GaAs is 5.9215 mm. When inputting the two pump waves with frequencies of 302 THz and 300.5 THz, the optimal length of crystal GaP is 3.2372 mm. When inputting the two pump waves with frequencies of 270 THz and 228.5 THz, the optimal length of crystal ZnTe is 2.0576 mm. When inputting the two pump waves with frequencies of 280 THz and 278.5 THz, the optimal length of crystal CdTe is 1.5073 mm.

In order to compare the photon conversion efficiencies of difference frequency and cascaded difference frequency, the diagrams of THz wave photon flux versus the frequency of the pump wave for different crystals in different cases are shown in Fig.5. All the crystals' lengths are 4 mm.

We can see from Fig.5, for a certain crystal, the photon conversion efficiencies of difference frequency and cascaded difference frequency are quite different at the same pump frequency. We also find that the optimal pump frequency is slightly higher than the so called zero-mismatch frequency.

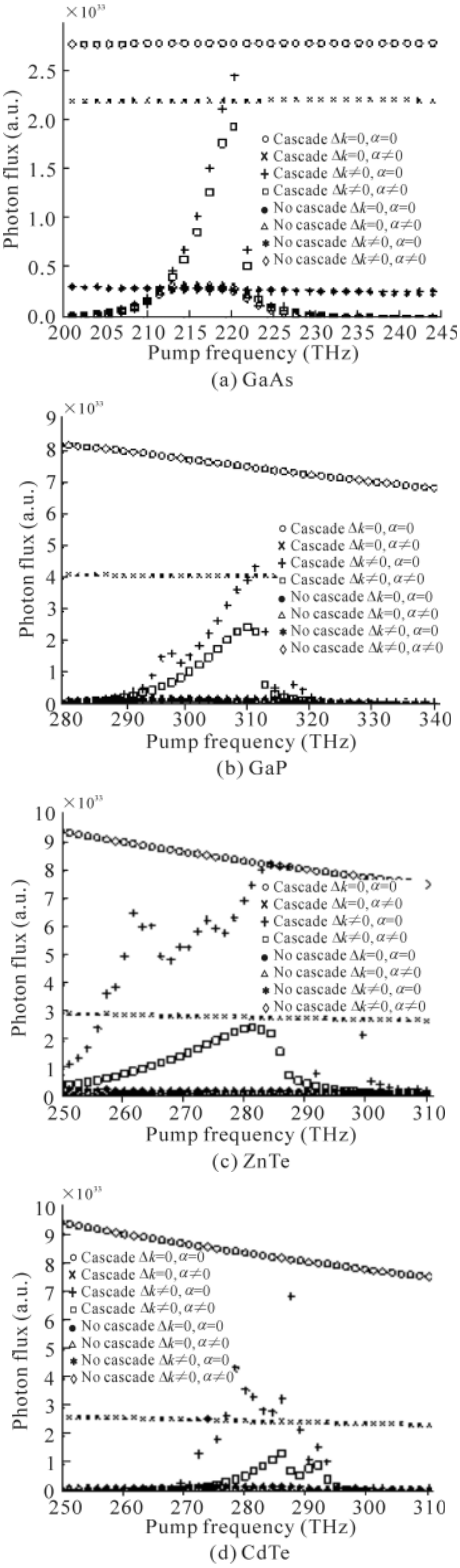

Fig.5 THz photon flux versus the pump frequency in GaAs, GaP, ZnTe and CdTe for different cases

From Fig.1, the zero-mismatch frequencies of GaAs, GaP, ZnTe and CdTe are 217.5 THz, 302.5 THz, 270 THz and 280.5 THz, respectively, in the no cascaded case. However, in the cascaded case, the optimal pump frequencies of those crystals are 223 THz, 310 THz, 281.5 THz and 287.5 THz, respectively. To some extent, the frequencies have high-frequency shifts.

In order to obtain the THz wave with the frequency of 1.5 THz, we make the frequencies of pump waves for GaAs, GaP, ZnTe and CdTe equal to 217.5 THz, 302.5 THz, 207 THz and 280.5 THz, respectively, and then those wave vector mismatches are zero approximately. When the frequency of output THz wave changes, the optimal pump frequencies should be calculated again. For the output THz wave intensity, the absorption coefficients are the smaller the better. From the comparison of the four crystals in no cascaded and cascaded cases, we find that cascaded difference frequency obviously increases the photon conversion efficiency for sphalerite crystals. For example, the photon conversion efficiencies of GaAs and GaP are increased by over 10 times.

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