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High-efficiency terahertz wave generation with multiple frequencies by optimized cascaded difference frequency generation*

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High-efficiency terahertz (THz) wave generation with multiple frequencies by optimized cascaded difference frequency generation (OCDFG) is investigated at 100 K using a nonlinear crystal consisting of a periodically poled lithium niobate (PPLN) part and an aperiodically poled lithium niobate (APPLN) part. Two infrared pump waves with a frequency difference ω_{T1} generate THz waves and a series of cascaded optical waves in the PPLN part by cascaded difference frequency generation (CDFG). The generated cascaded optical waves with frequency interval ω_{T1} then further interact in the APPLN part by OCDFG, yielding the following two advantages. First, OCDFG in the APPLN part is efficiently stimulated by inputting multi-order cascaded optical waves rather than the only two intense infrared pump waves, yielding unprecedented energy conversion efficiencies in excess of 37% at 1 THz at 100 K. Second, THz waves with M times ω_{T1} are generated by mixing the mth-order and the $(m+M)$th-order cascaded optical waves by designing poling period distributions of the APPLN part.

Keywords: terahertz wave generation, optimized cascaded difference frequency generation, aperiodically poled lithium niobate

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1. Introduction

Terahertz (THz) waves have recently received great attention due to a wide variety of interesting applications, such as THz imaging and spectroscopy for materials science,[1] security inspections[2] and nondestructive inspections,[3] THz fundamental scientific applications for inspecting vibrational modes of molecules, polarons, excitons, phonons and magnons,[4,5] THz industrial applications for pharmaceutical industry,[6] food industry[7] and communication industry.[8] For such applications, a high-power THz source is essential. Among many optical methods for THz wave generation, cascaded difference frequency generation (CDFG) holds great promise as an approach with quantum conversion efficiency beyond the Manley–Rowe limit.[9–14] In many cases, CDFG is not well phase-matched. Phase mismatches increase rapidly with the increase of cascading orders, which limits the evolution of optical spectrum to higher cascading orders.[14] The above problem is solved by optimized cascaded difference frequency generation (OCDFG) where aperiodically poled lithium niobate (APPLN) with optimized poling period distributions is utilized to guide the phase mismatches of cascaded Stokes processes (CSP) to zero one by one from the first order to the high orders, yielding energy conversion efficiencies in excess of 30% from optical waves to THz waves.[9]

Though the phase mismatches of OCDFG are optimized by designing poling period distributions of APPLN, a large part of the pump photons are transferred to cascaded Stokes waves, while a small part is transferred to cascaded anti-Stokes waves as the phase mismatches of a few low-order cascaded anti-Stokes processes (CaSP) are small. The above CaSP depress the energy conversion efficiency. Moreover, it is hard to generate multiple frequencies or tunable THz waves with only two fixed pump frequencies by CDFG. THz wave parametric oscillator or THz wave parametric generator can produce tunable THz sources with a pump laser and a seed laser, but cascaded optical processes are challenging to implement due to noncollinear phase-matched.[15] In this work, we propose an approach to generate high-efficiency THz waves with multiple frequencies by OCDFG. The nonlinear crystal consists of a PPLN part and an APPLN part. The two pump lasers with frequency difference ω_{T1} generate cascaded optical waves with interval frequency ω_{T1} in the PPLN part by CDFG. The cascaded optical waves with M times ω_{T1} frequency difference can efficiently interact by designing poling period distributions of APPLN, thus generating THz waves with a frequency of

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M times ω_{T1}. Taking full advantage of inputting multi-order cascaded optical waves and modulating phase mismatches of OCDFG, unprecedented energy conversion efficiencies in excess of 37% at 1 THz are realized at a temperature of 100 K, respectively.

2. Theoretical model

According to the basic principle of CDFG, the input two pump waves ω_0 and ω_1 generate a series of cascading optical waves and THz waves ($\omega_{T1} = \omega_0 - \omega_1$), as shown in Fig. 1(a). The red line indicates the interaction between two cascaded optical waves, and the yellow line indicates the generation and consumption of THz photons. The interval frequency between adjacent cascaded optical waves is ω_{T1}. As phase mismatches between adjacent cascaded optical waves can be set to zero one by one from the first order to high orders by designing poling period distributions of APPLN, THz wave with a frequency of ω_{T1} can be efficiently generated. If phase mismatches between the mth-order and the $(m+2)$th-order cascaded optical waves are set to zero one by one from the first order to high orders by designing poling period distributions, THz wave with a frequency of ω_{T2} ($\omega_{T2} = 2\omega_{T1}$) can be efficiently generated, as shown in Fig. 1(b). Similarly, THz waves with frequencies of ω_{T3} ($\omega_{T3} = 3\omega_{T1}$) and ω_{T4} ($\omega_{T4} = 4\omega_{T1}$) can be generated by designing poling period distributions, as shown in Figs. 1(c)–1(d), respectively. It can be deduced that THz wave with a frequency of M times ω_{T1} can be generated through the mixing of the mth-order and the $(m+M)$th-order cascaded optical waves by designing poling period distributions.

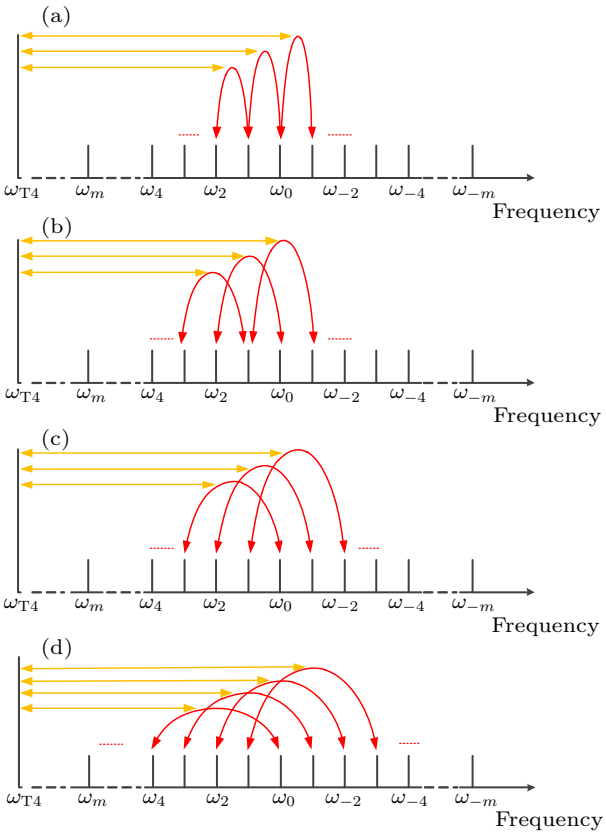


Fig. 1. Schematic diagram of OCDFG generating THz waves with multiple frequencies.

Nonlinear crystal consists of a PPLN part and an APPLN part, as shown in Fig. 2. Two infrared pump lasers ω_0 and ω_1 generate a series of cascading optical waves in the PPLN part by CDFG. The generated cascaded optical waves with frequency interval ω_{T1} interact in the APPLN part generating THz waves with a frequency of M times ω_{T1} by OCDFG. The coupled wave equations of OCDFG can be derived from those of CDFG, given by

$$\frac{dE_{TM}}{dz} = -\frac{\alpha_{TM}}{2}E_{TM} + j\kappa_{TM}\sum_{-\infty}^{+\infty}E_m E_{m+M}^* e^{j\Delta k_{mM}z}, \quad (1)$$

$$\frac{dE_m}{dz} = -\frac{\alpha_m}{2}E_m + j\kappa_m[E_{m-M}E_{TM}^* e^{j\Delta k_{m-Mz}} + E_{m+M}E_{TM}e^{-j\Delta k_{mMz}}], \quad (2)$$

$$\kappa_m = \frac{\Omega_m d_{eff}}{cn_m}, \quad (3)$$

$$\kappa_{TM} = \frac{\Omega_{TM} d_{eff}}{cn_{TM}}, \quad (4)$$

$$\Delta k_{mM} = k_m - k_{m+M} - k_{TM} + k_{\Lambda M}, \quad (5)$$

$$I = \frac{1}{2}nc\varepsilon_0|E|^2, \quad (6)$$

$$\eta(z) = \frac{n(\omega_{TM})|E_{TM}(\omega_{TM},z)|}{\sum n(\omega_m)|E_{(\omega_m,0)}|}, \quad (7)$$

where the subscript M is the positive integer, indicating the M times of ω_{T1}. The subscripts m and TM represent mth-order cascaded optical wave and THz wave with M times ω_{T1}, respectively. Ω is the angular frequency, E is the amplitude of the electric field, α is the absorption coefficient, κ is the coupling coefficient, n is the refractive index, k is the wave vector, Δk is the phase mismatch, d_{eff} is the nonlinear coefficient, c is the speed of light in vacuum, I is the power density, ε_0 is the vacuum dielectric constant, and $\eta(z)$ is energy conversion efficiency. The first term of Eq. (1) represents the absorption of THz wave, and the second term represents the sum of THz wave intensities generated by ω_m and ω_{m+M} in OCDFG. The first term of Eq. (2) represents the absorption of the mth-order cascaded optical wave, the second term describes the generation of ω_m by the interaction between ω_{m-M} and ω_{TM}, and the third term describes the consumption of ω_m by the OCDFG generating ω_{m+M} and ω_{TM}.

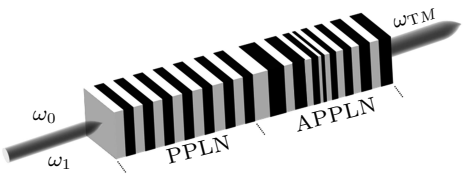


Fig. 2. Nonlinear crystal consisting of a PPLN part and an APPLN part.

3. Numerical simulations

In the calculations, the incident two pump frequencies ω_0 and ω_1 are 291.76 THz and 291.26 THz, respectively. ω_{T1} is 0.5 THz. The theoretical refractive index values of MgO:LiNbO$_3$ in infrared[16] and THz regions [17] are calculated by wavelength-and temperature-dependent Sellmeier equation, respectively. At 100 K, the absorption coefficients of cascaded optical waves are 0 cm^{-1}, and the absorption coefficients of THz wave at 0.5 THz, 1.0 THz, 1.5 THz, 2.0 THz are 130 cm^{-1}, 210 cm^{-1}, 370 cm^{-1}, 650 cm^{-1}, respectively.[17] The nonlinear optical coefficient of the PPLN is 336 pm/V at 291.76 THz,[13] and the laser damage threshold of MgO:LiNbO$_3$ with the pulse duration of 100 ps is $\sim$ 10 GW/cm^2.[18] The poling period of PPLN Λ_{PPLN} is set to 237.17 μm for the phase-matched first-order CSP at 100 K.

Cascaded optical spectra are generated by CDFG in the PPLN part with two pump intensities I_0 and I_1, as shown in Fig. 3. As shown in Fig. 3(a), with lower pump intensities of 30 MW/cm^2, pump photons do not diffuse to high-order cascaded optical waves. As pump intensities increase to 300 and 3000 MW/cm^2, the diffusion range of cascaded optical spectra is enlarged. The two pump lasers are converted into a series of cascaded optical waves, which provides a possibility to generate THz waves with multiple frequencies.

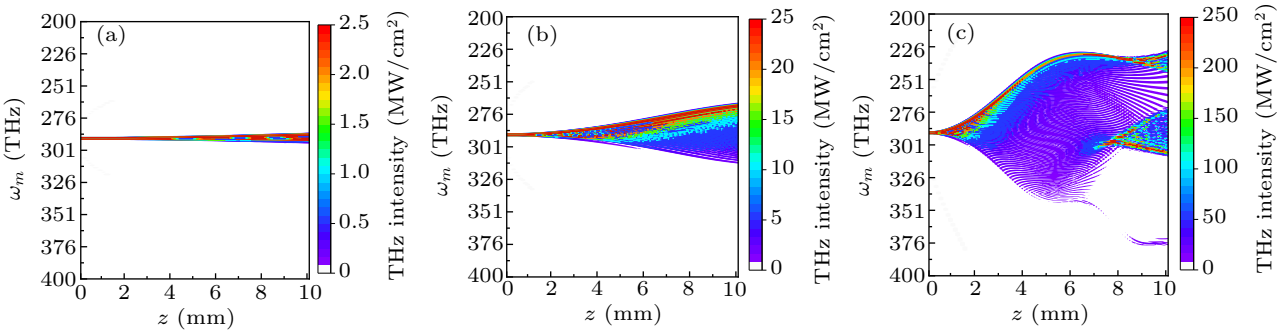


Fig. 3. Cascaded optical spectra generated by CDFG in the PPLN part. (a) $I_0 = I_1 = 30$ MW/cm^2, (b) $I_0 = I_1 = 300$ MW/cm^2, (c) $I_0 = I_1 = 3000$ MW/cm^2.

As cascaded optical waves generated in the PPLN part stimulate the APPLN part, the mth-order cascaded optical wave can interact with the $(m+M)$th-order cascaded optical wave, providing the phase mismatches are small enough. The phase mismatches Δk_{mM} can be compensated by selecting suitable poling periods. Figure 4 shows the distributions of $|\Delta k_{mM}|$ with different poling periods. From the figure, we find that the phase mismatches $|\Delta k_{mM}|$ increase with the increase of M. The phase mismatches $|\Delta k_{m1}|$, $|\Delta k_{m2}|$, $|\Delta k_{m3}|$, and $|\Delta k_{m4}|$ can be small enough by choosing suitable poling periods, which indicates that THz waves with frequencies of 0.5, 1.0, 1.5, 2.0 THz can be efficiently generated. Moreover, if the variable poling periods keep the $|\Delta k_{mM}|$ minimal at each-order CSP from 291.76 THz to 60.26 THz, the phase mismatches $|\Delta k_{mM}|$ of CSP are equal to zero one by one from the first order to high orders, whereas the phase mismatches $|\Delta k_{mM}|$ of CaSP are enlarged, as shown in Fig. 4. The optimal poling periods of APPLN $\Lambda_{\mathrm{APPLN}-M}$ is given by

$$\Lambda_{\mathrm{APPLN}-M} = \frac{c}{-n_m \times \left(\omega_0 - \omega_{\mathrm{TM}} \times N \times z/L\right) + n_{m+M} \times \left(\omega_0 - \omega_{\mathrm{TM}} - \omega_{\mathrm{TM}} \times N \times z/L\right) + n_{\mathrm{TM}} \times \omega_{\mathrm{TM}}}, \tag{8}$$

where N denotes the number of cascaded orders, L is the maximum crystal length. The $\Lambda_{\mathrm{APPLN}-M}$ described by Eq. (8) leads the phase mismatches $|\Delta k_{mM}|$ of CSP to zero one by one from the first order to high orders.

The number of cascaded optical waves No generated from the PPLN part influences the output of THz waves from the APPLN part by OCDFG. As shown in Fig. 3, the number No depends on the length of the PPLN part. Figure 5 shows energy conversion efficiency η by OCDFG in the APPLN part versus the number No. In this figure, $\omega_{T1} = 0.5$ THz, and $\Lambda_{\mathrm{APPLN}-1}$ is calculated with Eq. (8). From the figure, we find that the energy conversion efficiency η increases with the two pump intensities.The maximum η of 1.87%, 14.32% and 36.14% is reached in Figs. 5(a), 5(b), and 5(c), respectively. The energy conversion efficiency η is significantly improved as the number No increases. Compared with η at No = 2, the maximum η increases by 3.9, 2.2, and 1.94 times in Figs. 5(a), 5(b), and 5(c), corresponding to the number No of 15, 27, and 28, respectively. A large number of pump photons are transferred to cascaded anti-Stokes waves via CaSP with No = 2 as the phase mismatches of a few low-orders CaSP is small, which depresses the η. With No > 10, the photon number of each-order cascaded optical wave is moderate, which leads the energy transfer from the low-order cascaded Stokes waves to high-order cascaded Stokes waves sufficiently.

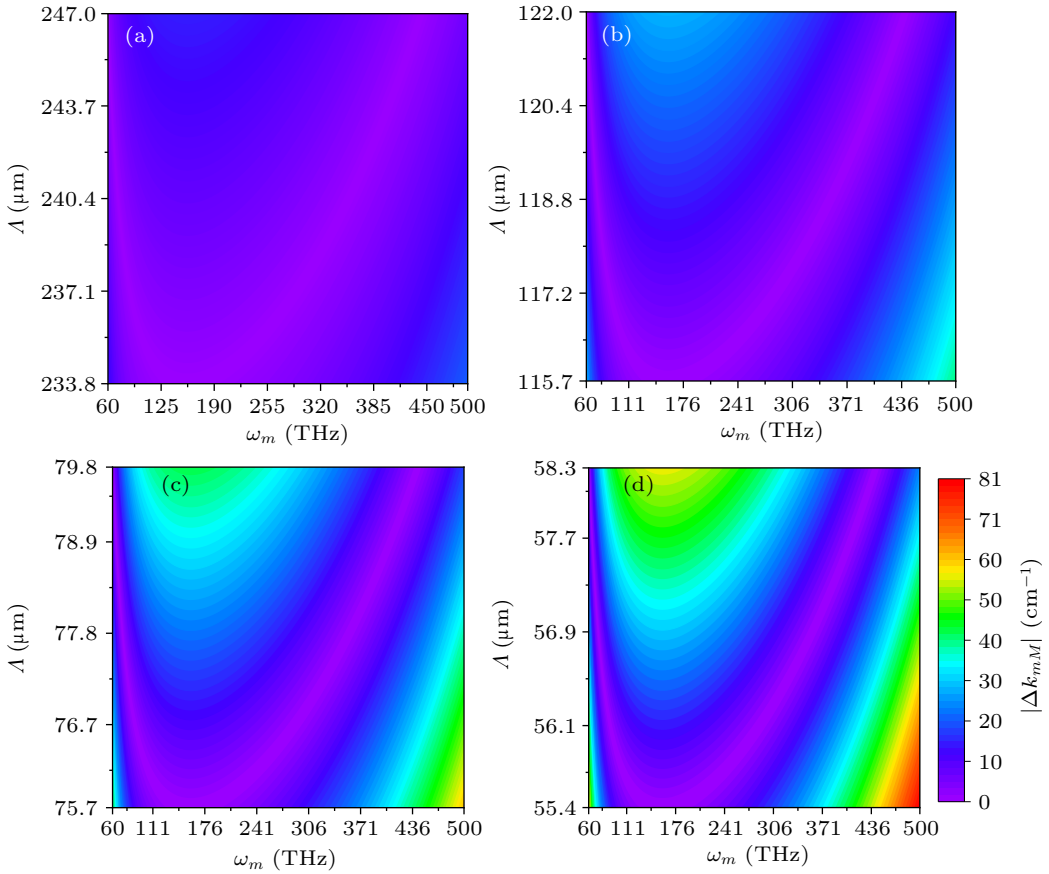


Fig. 4. The poling periods Λ and phase mismatches $|\Delta k_{mM}|$ versus cascaded optical frequency. (a) $M = 1$, $\omega_{\Gamma 1} = 0.5$ THz, (b) $M = 2$, $\omega_{\Gamma 2} = 1.0$ THz, (c) $M = 3$, $\omega_{\Gamma 3} = 1.5$ THz, (d) $M = 4$, $\omega_{\Gamma 4} = 2.0$ THz.

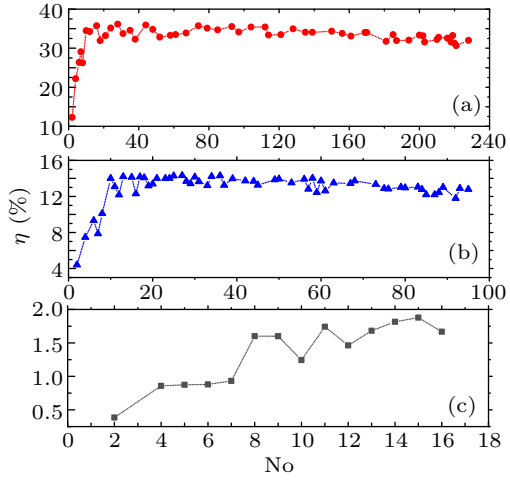


Fig. 5. Energy conversion efficiency η by OCDFG in the APPLN part versus No. THz wave frequency is 0.5 THz. (a) $I_0 = I_1 = 3000$ MW/cm^2, (b) $I_0 = I_1 = 300$ MW/cm^2, (c) $I_0 = I_1 = 30$ MW/cm^2.

Figure 6 shows energy conversion efficiency η by OCDFG in the APPLN part versus the number No at 0.5, 1.0, 1.5, 2.0 THz, respectively. Both the two pump intensities are 3000 MW/cm^2. From the figure, we find that η increases rapidly with No, then remains relatively stable, and finally decreases smoothly. All the energy conversion efficiencies η exceed 20% with No > 25. The optimal No at 0.5, 1.0, 1.5, 2.0 THz is 28, 44, 57, and 66, respectively. η depends on THz wave absorption coefficients α_{TM}, coupling coefficients κ_{TM}, and phase mismatches $|\Delta k_{mM}|$. The maximum η at 1.0 THz slightly exceeds that at 0.5 THz, which attributes to the larger coupling coefficients at 1.0 THz. The η at 1.5 THz and 2.0 THz are lower than those at 0.5 THz and 1.0 THz, which attributes to the larger THz wave absorption coefficients and phase mismatches at 1.5 THz and 2.0 THz.

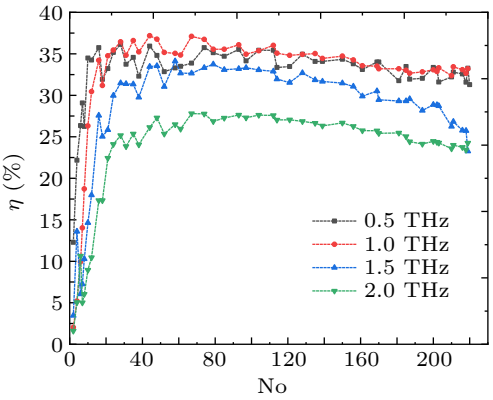


Fig. 6. Energy conversion efficiency η by OCDFG in the APPLN part versus the number No at 0.5, 1.0, 1.5, 2.0 THz, respectively. $I_0 = I_1 = 3000$ MW/cm^2.

The cascaded optical spectra and THz wave intensities generated in the PPLN part by CDFG and in the APPLN part by OCDFG are shown in Fig. 7. The length of the PPLN part corresponds to the optimal No shown in Fig. 6. From the figure, we find that the diffusions of cascaded optical waves

in Figs. 7(a)–7(d) show almost identical trends. The diffusion ranges are roughly the same, and the highest-order cascaded Stokes wave diffuses to around 80 THz. Most of the pump photons are transferred to the Stokes range, while a small number of pump photons are transferred to the anti-Stokes range. Compared with the diffusions of cascaded optical waves in Fig. 3(c), the diffusion ranges are wider in the Stoke range and the pump photons transferred to Stokes range are more in Figs. 7(a)–7(d), which indicates that THz generation by OCDFG is superior to that by CDFG. As shown in Fig. 7(e), THz wave at 0.5 THz is generated in the PPLN part with CDFG and in the APPLN part with OCDFG, while THz waves at 1.0, 1.5, and 2.0 THz are generated in the APPLN part by OCDFG. THz wave intensities increase rapidly to the maximum and then decrease fast, which is consistent with the diffusion trends of cascaded optical waves in Figs. 7(a)–7(d). The maximum THz wave intensities are 2168.5 MW/cm^2 at 0.5 THz, 2231.0 MW/cm^2 at 1.0 THz, 2046.6 MW/cm^2 at 1.5 THz, and 1667.8 MW/cm^2 at 2.0 THz, corresponding to the energy conversion efficiency η of 36.1%, 37.1%, 34.1%, and 27.8%, respectively.

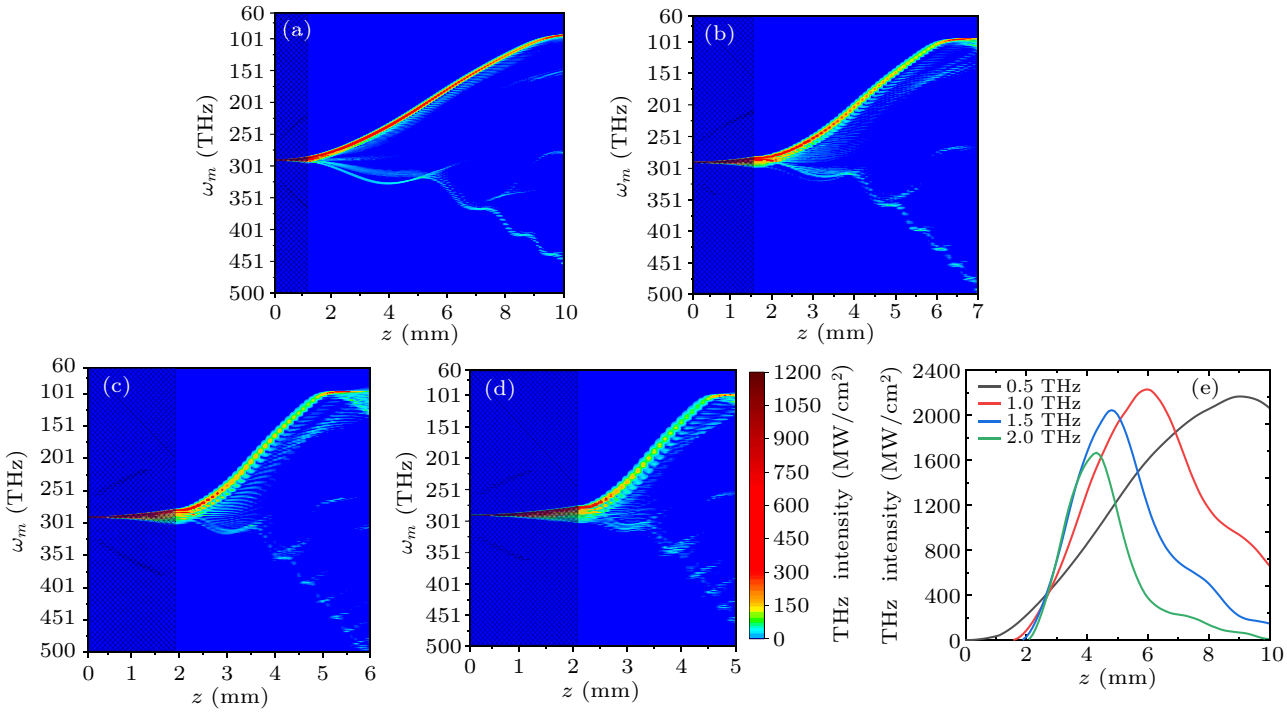


Fig. 7. The cascaded optical spectra in the PPLN part generated by CDFG and in the APPLN part generated by OCDFG, $I_0 = I_1 =$ 3000 MW/cm^2. The shaded region represents the PPLN part and the non-shaded region represents the APPLN part. (a) $\omega_{T1} = 0.5$ THz, No = 28, and the length of the PPLN part is 1.2 mm, (b) $\omega_{T2} = 1.0$ THz, No = 44, and the length of the PPLN part is 1.6 mm, (c) $\omega_{T3} = 1.5$ THz, No = 57, and the length of the PPLN part is 1.9 mm. (d) $\omega_{T4} = 2.0$ THz, No = 66, and the length of the PPLN part is 2.1 mm, (e) THz wave intensities at 0.5, 1.0, 1.5, and 2.0 THz.

4. Discussion

In this work, we design a scheme to efficiently generate THz waves with multiple frequencies by OCDFG. The frequency ω_{T1} can be adjusted by changing the two pump frequencies of ω_0 and ω_1. As a result, THz waves with M times ω_{T1} can be generated. Most of the THz frequencies can be generated, providing ω_{T1} is small enough. Ravi *et al.* reported a THz wave generation scheme by CDFG with PPLN with energy conversion efficiency η of 8% at 77 K.[12] Our group reported a THz wave generation scheme by OCDFG using APPLN with energy conversion efficiency η of 33.4% at 10 K.[9] In Ref. [9], the OCDFG is stimulated by only two intense pump lasers, resulting in insufficient energy transfer from pump photons to high-order Stokes photons. Moreover, part of pump photons are transferred to anti-Stokes waves, which consumes THz photons. In this work, the OCDFG is stimulated by multi-order cascaded optical waves, resulting in sufficient energy transfer from the mth order cascaded Stokes wave to the $(m+M)$th-order cascaded Stokes wave, yielding the maximum η in excess of 37% at 1 THz at 100 K.

In practice, the experimental device is mainly composed of an APPLN crystal and two intense infrared pump lasers. The two pulsed Yb:YAG lasers with a wavelength around 1030 nm are preferred.[12] As the APPLN crystal with gradually changing poling periods is inconvenient to manufacture, the APPLN crystal with stepwise changing poling periods, which can provide similar performance, is easy to manufacture and has been extensively used for THz wave generations.[19,20] The high performance Yb:YAG laser and APPLN manufacturing technology make the high-efficiency THz wave generation with multiple frequencies by OCDFG practically feasible.

5. Conclusion

We present a scheme of generating multiple frequencies THz waves by OCDFG at 100 K. By introducing multi-order cascaded optical waves instead of two intense pump lasers to stimulate the APPLN part, THz waves with multiple frequencies are generated through the mixing of the mth-order and the $(m+M)$th-order cascaded optical waves. Moreover, energy transfer from the low-order cascaded Stokes waves to the high-order cascaded Stokes waves sufficiently, yielding unprecedented energy conversion efficiencies in excess of 37% at 1 THz at 100 K. The scheme paves the way for high-efficiency THz wave generation with multiple frequencies by only two pump lasers.

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