

Investigation on high-order cascaded parametric processes in terahertz parametric oscillator with a noncollinear phase-matching scheme

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Abstract: Cascaded optical parametric processes in terahertz parametric oscillator with a noncollinear phase-matching scheme based on bulk lithium niobate were observed. The spectra of the first-, second-, and third-order Stokes waves were observed. By analyze the spectra of the first-, second-, and third-order Stokes waves it is found that the frequency difference between adjacent-order Stokes waves is approximately equivalent, which means that the high-order cascaded parametric processes have realized. With high-order cascaded terahertz parametric processes one pump photon can generate several THz photons, which means that the quantum conversion efficiency can be enhanced.

Key words: terahertz wave; terahertz wave parametric oscillator;
cascaded optical parametric oscillation

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非共线相位匹配太赫兹波参量振荡器级联参量过程的研究

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摘 要: 在铌酸锂晶体非共线相位匹配太赫兹波参量振荡器中观察到了级联光学参量效应。实验中测量到了一阶、二阶和三阶斯托克斯光。通过分析一阶、二阶和三阶斯托克斯光谱发现相邻阶斯托克斯光频率差相等, 表明在太赫兹波的产生过程中发生了级联光学参量效应。在高阶级联光学参量过程中, 一个泵浦光子可以产生多个太赫兹光子, 表明在太赫兹波产生过程中量子转换效率会有效提高。

关键词: 太赫兹波; 太赫兹波参量振荡器; 级联光学参量振荡

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0 Introduction

Terahertz (THz) radiation, widely defined as electromagnetic radiation in the frequency range 0.1–10 THz, offers researchers many intriguing possibilities, ranging from fundamental science through to applications in communications, imaging and spectroscopy, security and defense, and non-destructive evaluation^[1–6]. Among many electronic and optical methods for the THz – wave generation, THz – wave parametric oscillator (TPO) and difference-frequency generation (DFG) in a second-order nonlinear medium exhibit many advantages, such as narrow linewidth, coherent, wide tunable range, high-power output and room temperature operation^[7–9]. The TPO with both noncollinear phase-matching configuration and quasi-phase-matching configuration can perform well^[10–11]. Unfortunately, the quantum conversion efficiency of the TPO and DFG is extremely low as the THz – wave is intensely absorbed by the gain medium. To improve the low quantum conversion efficiency, Sowade et al. present a continuous-wave optical parametric terahertz light source with a scheme of cascaded processes which can increase the output power of a terahertz system by more than one order of magnitude^[12]. Walsh et al. report the use of quasi-phase-matching techniques based on periodically-poled MgO:LiNbO₃ with a scheme of cascaded difference frequency generations for the generation of nanosecond duration pulses of terahertz radiation in intracavity optical parametric oscillators^[13]. Liu et al. analyze the threshold and conversion efficiency of a cascaded continuous-wave TPO^[14]. With high-order cascaded parametric processes one pump photon can generate several THz photons, suggesting that the overall quantum conversion efficiency for the THz radiation can overcome the Manley-Rowe limit.

In this paper, we report on the observation of the high-order Stokes waves from cascaded parametric processes with a noncollinear phase-matching scheme.

We present a comprehensive study of the effect.

1 Experimental setup and results

The experimental setup comprised a single-resonant optical parametric oscillator with a Fabry-Perot cavity pumped by a multi-longitudinal-mode Q-switched Nd:YAG laser at 1064 nm, as shown in Fig.1.

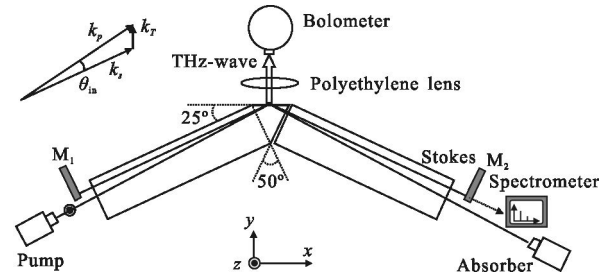
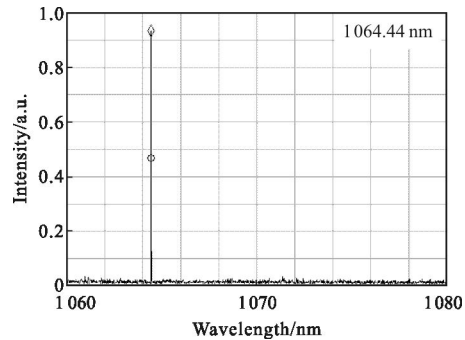


Fig.1 Experimental setup of the MgO:LiNbO₃-TPO. Fabry-perot cavity and the MgO:LiNbO₃ crystal are mounted onto a rotating stage

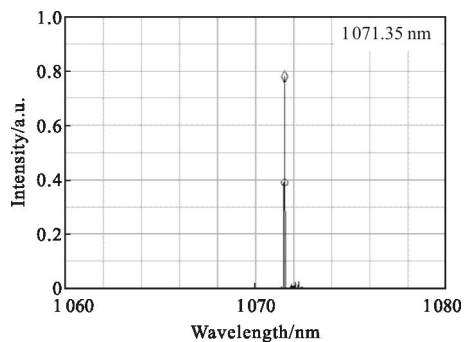
The pulse width and the repetition rate of the pump wave were 15 ns and 10 Hz, respectively. The pump beam was collimated by a lens pair to a diameter of about 1.9 mm. The nonlinear gain mediums were composed of two 5 mol% MgO doped LiNbO₃ crystals. The dimensions of the rectangular crystal were 50(x)×10(y)×5(z) mm³. The other pentagonal crystal was composed of a rectangular crystal with dimensions of 50(x)×10(y)×5(z) mm³ and an isosceles triangle crystal with an apex angle of 50°. The incident angle of the Stokes wave to the exit facet of the THz–wave was 65°, which ensured that the THz–wave almost perpendicularly exited from the LiNbO₃ crystal due to noncollinear phase matching condition. All the crystal surfaces were antireflection coated with a residual reflectivity smaller than 1% at the Stokes wavelengths. The polarizations of the pump wave, Stokes wave and THz–wave were all along the z–axis of the LiNbO₃ crystal. The TPO cavity for the Stokes wave consisted of two plane-parallel mirrors, M₁ and M₂. M₁ was highly reflecting (>99.8%) and M₂ was coated with a reflectivity of 85%. The cavity with a length of 210 mm was symmetric. The pump wave passed through the

cavity at the edge of M_1 and M_2 . The cavity mirrors and the $\text{MgO}:\text{LiNbO}_3$ crystal were mounted on a rotating stage. The wavelength of the first-order Stokes wave, and hence the wavelength of the THz-wave can be tuned by rotating the stage continuously since the angle between the pump wave vector and the optical axis of the Stokes wave cavity are changing continuously.

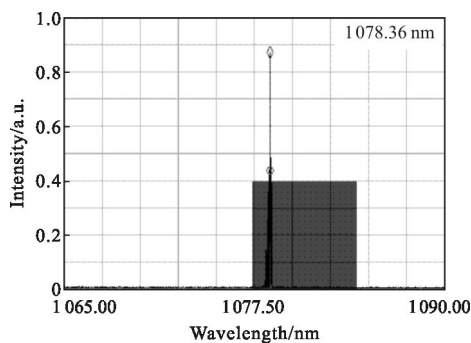
By using a single optical fiber positioned beyond M_2 , feeding into an optical spectrum analyzer (Agilent, model 86142B), the wavelengths of the high-order Stokes waves can be measured. During the experiments we observe the spectra of the pump wave, the first-, second-, and third-order Stokes waves, as shown in Fig.2. The wavelengths of the



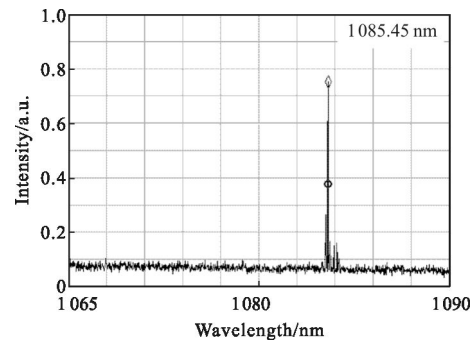
(a) Pump wave



(b) First-order Stokes wave



(c) Second-order Stokes wave



(d) Third-order Stokes wave

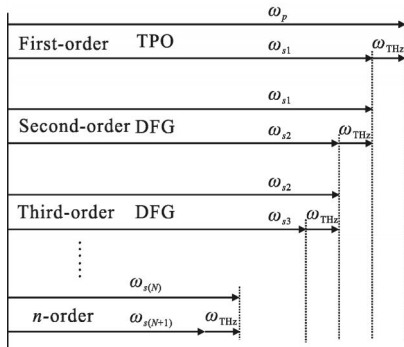
Fig.2 Spectra of the pump wave and stokes waves captured by an agilent 86142B optical spectrum analyzer

pump wave, the first-, second-, and third-order Stokes waves are 1064.44 nm, 1071.35 nm, 1078.36 nm and 1085.45 nm, respectively. The frequency difference of 1.818 THz between the pump wave and the first-order Stokes wave immediately gives the frequency of the THz-wave generated by the first-order parametric process. The difference frequencies between the first-order and the second-order Stokes wave, the second-order and the third-order Stokes wave are 1.820 THz and 1.817 THz, respectively. The spectral lines exhibit a constant frequency shift between the adjacent-order Stokes waves.

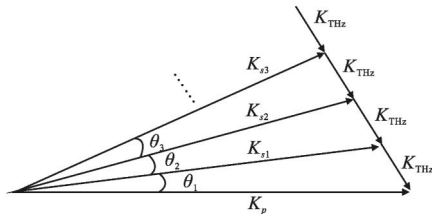
2 Investigation of the high-order Stokes waves

The near equality of the frequency differences mentioned above is consistent with the second-order Stokes wave being generated by the process of DFG in which the THz-wave, which is parametrically generated as a result of the interaction between the pump-wave and the first-order Stokes wave, then subsequently interacts with the first-order Stokes wave so as to generate the second-order Stokes wave. The cascaded optical processes can be continued as the high-power and high conversion efficiency THz-wave radiations based on DFG have been realized^[15-17]. The energy diagram and phase-matching diagram of cascaded optical parametric processes is shown in Fig.3. The first-order parametric process converts the pump

wave ω_p into the THz-wave ω_T and the first-order Stokes wave ω_{s1} . The second-order Stokes wave ω_{s2} is generated by the process of DFG in which the first-order Stokes wave ω_{s1} interacts with the THz-wave ω_T . The THz-wave ω_T which is initially generated in the first-order parametric process is amplified within the process of DFG. The third-order Stokes wave ω_{s3} is generated by the process of DFG in which the THz-wave ω_T interacts with the second-order Stokes wave ω_{s2} . With high-order cascaded optical parametric processes from every pump photon several terahertz photons are created rather than only one.



(a) Energy diagram of cascaded optical parametric processes



(b) Phase-matching diagram of cascaded optical parametric processes (The lengths of the wavevectors do not scale)

Fig.3 Cascaded optical parametric processes

The propagation direction of the first-order Stokes wave is determined by the optical axis of the Stokes wave resonant cavity formed by mirrors M_1 and M_2 . The propagation direction of the second-order and third-order Stokes wave is undefined by any external constraints, and is determined by the phase matching condition alone, as shown in Fig.3 (b). For the first-order THz-wave parametric process, two requirements have to be fulfilled: the energy conservation condition:

$$\frac{1}{\lambda_p} = \frac{1}{\lambda_{s1}} + \frac{1}{\lambda_T} \quad (1)$$

and the phase-matching condition:

$$\vec{k}_p = \vec{k}_{s1} + \vec{k}_T \quad (2)$$

Here, λ is the wavelength, while \vec{k} is the wave-vector, the subscripts p , $s1$ and T denote the pump wave, first-order Stokes wave and THz-wave, respectively. The magnitudes of the corresponding wave vectors (within the nonlinear gain medium) are calculated using the following equation:

$$k_j = \frac{2\pi n_j}{\lambda_j} \quad (3)$$

where the subscript j indicates the wave of interest, and n_j is the refractive index for that wave. The phase-matching condition can be rewritten as

$$k_T^2 = k_p^2 + k_{s1}^2 - 2k_p k_{s1} \cos \theta_1 \quad (4)$$

where θ_1 is the angle between the pump wave and the first-order Stokes wave.

For the second-order process, the energy conservation condition is

$$\frac{1}{\lambda_{s1}} = \frac{1}{\lambda_{s2}} + \frac{1}{\lambda_T} \quad (5)$$

and the phase-matching condition is

$$k_T^2 = k_{s1}^2 + k_{s2}^2 - 2k_{s1} k_{s2} \cos \theta_2 \quad (6)$$

Here, the subscript $s2$ denotes the second-order Stokes waves. θ_2 is the angle between the first-order Stokes wave and the second-order Stokes wave.

For the third-order process, the energy conservation condition is

$$\frac{1}{\lambda_{s2}} = \frac{1}{\lambda_{s3}} + \frac{1}{\lambda_T} \quad (7)$$

and the phase-matching condition is

$$k_T^2 = k_{s2}^2 + k_{s3}^2 - 2k_{s2} k_{s3} \cos \theta_3 \quad (8)$$

Here, the subscript $s3$ denotes the third-order Stokes waves. θ_3 is the angle between the second-order Stokes wave and the third-order Stokes wave.

The theoretical values of the pump and Stokes wavelengths are calculated using a wavelength- and temperature-independent Sellmeier equation for 5% MgO-doped congruent lithium niobate (MgO:cLN) in

the IR^[18] and THz range, respectively^[19]. The Sellmeier equation for MgO:cLN of Gayer et al^[18]. in the IR range in a temperature range from 20 °C to 200 °C can be written as

$$n_e^2 = a_1 + b_1 f \frac{a_2 + b_2 f}{\lambda^2 - (a_3 + b_3 f)^2} + \frac{a_4 + b_4 f}{\lambda^2 - a_5} - a_6 \lambda^2 \quad (9)$$

where $a_1=5.756$, $a_2=0.0983$, $a_3=0.202$, $a_4=189.32$, $a_5=12.52$, $a_6=1.32 \times 10^{-2}$, $b_1=2.86 \times 10^{-6}$, $b_2=4.7 \times 10^{-8}$, $b_3=6.113 \times 10^{-8}$, $b_4=1.516 \times 10^{-4}$, and $f=(T-24.5) \times (T+570.82)$, T is the crystal temperature in °C. The Sellmeier equation for MgO:cLN of Kiessling et al^[19]. in the THz range in a temperature range from 30 °C to 200 °C can be written as

$$n_{\text{THz}}^2 = A_0 + B_0 f + \frac{A_1 + B_1 f}{\lambda^2 - (A_2 + B_2 f)^2} \quad (10)$$

where $A_0=24.326$, $A_1=31298$, $A_2=48.084$, $B_0=2.14 \times 10^{-5}$, $B_1=0.15824$, $B_2=-3.097 \times 10^{-4}$.

According to the Eqs.(4)–(8), the frequency tuning of the first-, second-, third-order Stokes waves can be realized by varying the angle θ_1 between the pump wave and the first-order Stokes wave, as is shown in Fig.4. The insert symbols are the experimental values. We measure 8 groups of experimental value at different angles θ_1 . From the figure we find that the experimental values agree well with the theoretical values calculated from the cascaded parametric processes. At any angle θ_1 the frequency difference between the first-order and second-order Stokes waves is approximately equal to the difference between the second-order and third-order Stokes waves, which

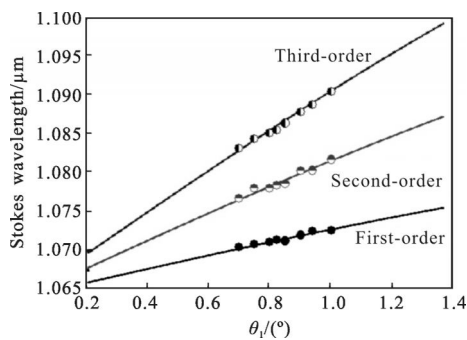


Fig.4 Wavelengths of the first-, second-, third-order Stokes waves versus the angle θ_1 (The solid lines show theoretical values, the symbols are measured values)

means that the high-order cascaded parametric processes have realized.

In contrast to the use of PPLN for THz-wave generation in the collinear phase-matching scheme, the use of bulk lithium niobate in the noncollinear phase-matching scheme is convenient for the generation of the tuning THz-wave by tuning the phase-matching angle between the pump wave and the first-order Stokes wave.

3 Conclusion

Cascaded optical parametric processes in terahertz parametric oscillator with a noncollinear phase-matching scheme based on bulk lithium niobate are observed. By analyze the spectra of the first-, second-, and third-order Stokes waves we find that the first-order parametric process is optical parametric oscillation and the high-order parametric processes are difference frequency generation. The frequency difference between adjacent-order Stokes waves is approximately equivalent. With high-order cascaded terahertz parametric processes one pump photon can generate several THz photons.

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