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Investigation of pump-wavelength dependence of terahertz-wave parametric oscillator based on $LiNbO_3$*

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This paper investigates the performances of terahertz-wave parametric oscillators (TPOs) based on the $LiNbO_3$ crystal at different pump wavelengths. The calculated results show that TPO characteristics, including the frequency tuning range, the THz-wave gain and the stability of THz-wave output direction based on the Si-prism coupler, can be significantly improved by using a short-wavelength pump. It also demonstrates that a long-wavelength-pump allows the employment of a short TPO cavity due to an enlarged phase-matching angle, that is, an increased angular separation between the pump and oscillated Stokes beams under the THz-wave generation at a specific frequency. The study provides an useful guide and a theoretical basis for the further improvement of TPO systems.

Keywords: terahertz-wave parametric oscillator, THz-wave radiation, pump-wavelength dependence

PACC: 4265K, 4265C, 7136, 7830G

1. Introduction

During the past few years, more and more researchers have shown considerable interests in terahertz-wave generation and detection from the perspectives of both fundamental research and application.[1] Following the THz-wave generation using photoconduction and optical rectification,[2,3] technologies based on the nonlinear optical parametric process are gradually beginning to play important roles in a variety of applications including monochromatic THz imaging, biomedical diagnostics, and THz spectroscopy.[4−7] This is largely due to their excellent and distinctive characteristics such as tunability, coherence and narrow line-width. In recent years, terahertz-wave parametric oscillators (TPOs) have been proved to be one of the techniques with most potential for the efficient generation of coherent, widely tunable and monochromatic THz waves. The TPOs have a number of advantages including compactness, simplicity, broad tunability and room-temperature operation. In addition, compared with the different frequency mixing schemes employing the second-order nonlinearities in a nonlinear crystal, a TPO system only requires a fixed-wavelength pump source and a low-cost nonlinear crystal such as $LiNbO_3$ (LN) crystal. Over the past decade, a great deal of outstanding progress in LN-TPO has been made by several research groups.[7−16]

In previous studies, Q-switched Nd:YAG lasers operating at the fundamental frequency of 1.064 μm were usually employed as the pump sources for LN-TPOs.[5−15] Widely tunable and coherent THz waves can be generated through the so-called angle-tuning method, as shown in Fig.1, where frequency-tuning is achieved by rotating the TPO cavity slightly to vary the incidence angle, $\theta_{\rm ext}$, relative to the TPO resonator axis. It is well known that the conventional Raman scattering process is independent of the incident pump wavelength, and inherently related to the difference between the initial and the final energy levels of the material. However, tunable THz-wave generation with TPOs, which is relevant to the near-forward stimulated Raman scattering process,[17] is determined by both dispersion characteristics of the A_1-symmetry polariton mode of LN and the three-wave phase-matching condition. Hence it is clear that

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the choice of the pump wavelength has a great effect on the operation of TPO.

This paper theoretically investigates the dependence of the operation of LN-TPO on the pump wavelength in detail. The theoretical analyses demonstrate that the employment of the short-wavelength pump can yield better performance of TPO in the frequency tuning range, THz-wave gain and the stability of THz-wave output direction of TPO base on the Si-prism coupler. Our calculated results also show that a long-wavelength pump allows the employment of a short TPO cavity due to an enlarged phase-matching angle, that is, a larger angular separation between the pump and oscillated Stokes beams under the THz-wave generation at a specific frequency. In addition, with a long-wavelength pump scheme we can avoid the occurrence of the photorefractive damage in nonlinear crystals.

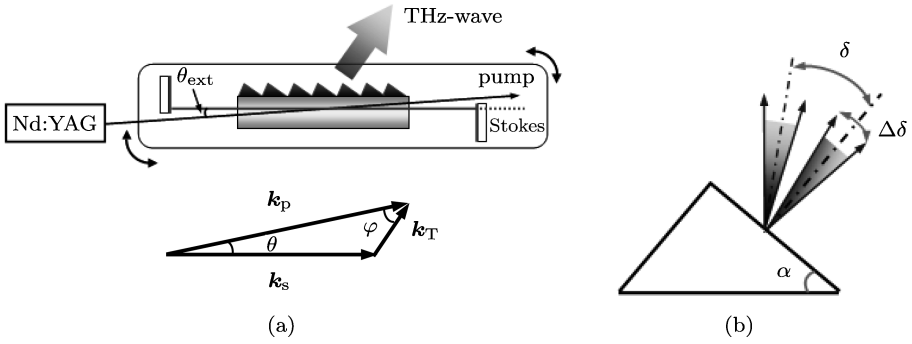


Fig.1. (a) Experimental setup of a typical TPO with a Si-prism array coupler, and (b) schematic diagram of a Si-prism.

2. Pump-wavelength dependence of THz-wave tunability

As is well known, the operation principle of a tunable LN-TPO is based on the process of the small-angle tunable parametric scattering (stimulated polariton scattering) occurred in the vicinity of the lowest A_1-symmetry polariton mode in LN. It is the dispersion characteristics of the infrared- and Raman-active mode of LN that account partly for the tuning capability of TPO. Figure 2 shows the theoretical dispersion characteristics of the 248 cm^{-1} polariton mode in LN, calculated from the infrared reflectivity data of LN by using[17]

$$k^2_{\text{Polariton}}(\nu) = \left(\omega^2/c^2\right)\varepsilon = 4\pi^2\nu^2\varepsilon(\nu), \qquad (1)$$

where, $k_{\text{Polariton}}$ is the polariton mode wavevector, c is the velocity of light in vacuum and $\varepsilon(\nu)$ is the complex dielectric constant given by

$$\varepsilon(\nu) = \varepsilon_\infty + \sum_j \frac{S_j\nu^2_{0j}}{\nu^2_{0j} - \nu^2 - \mathrm{i}\nu\Gamma_j}, \qquad (2)$$

where ν_{0j}, S_j and Γ_j are respectively the jth eigenfrequency, oscillator strength and damping coefficient of the lowest A_1-symmetry phonon mode, ε_∞ is the high-frequency dielectric constant. Since the polariton is a quasi-particle resulting from strong coupling of electromagnetic waves with transverse optical vibration modes (TO modes) when their frequencies and wavevectors are nearly identical, its dispersion curve is partly phonon-like in the high-frequency region, where the predominant scattering is similar to the Raman scattering process, and partly photon-like in the low-frequency region, where the predominant scattering process can be thought of as a parametric process.

In this stimulated scattering process, the generated THz-wave radiation frequency (ω_T) and the Stokes radiation frequency (ω_s) together with the pump frequency (ω_p) satisfy the energy conservation law

$$\omega_p = \omega_s + \omega_T, \qquad (3)$$

and the noncollinear phase-matching condition $\boldsymbol{k}_p = \boldsymbol{k}_s + \boldsymbol{k}_T$ (see the inset of Fig.1), which can be rewritten

in the form of cosine law as

$$k_{\mathrm{T}}^2 = k_{\mathrm{p}}^2 + k_{\mathrm{s}}^2 - 2k_{\mathrm{p}}k_{\mathrm{s}}\cos\theta, \tag{4}$$

where k_{p}, k_{s} and k_{T} are the wavevectors of the pump, Stokes, and THz waves, respectively, and θ is the phase-matching angle between the pump and Stokes beams in the crystal.

As can be seen, in Eq.(4) there are two independent variables, θ and $\boldsymbol{k}_{\mathrm{p}}$. By varying the angle θ within a selected range under a certain fixed pump wavelength (the so-called angle-tuning method) and using Eq.(4), we can obtain a family of phase-matching curves representing the locus of all points satisfying both the energy and wavevector matching conditions. The intersections of these phase-matching curves and the dispersion curve of the polariton mode are expected to determine the allowed frequencies of the THz wave and the simulated Stokes wave participating in the scattering process under small angle condition.[16] It should be stressed that the principle of the angle-tuning method of TPO is inherently distinguishable from an optical parametric oscillation system, in which the tuning of output frequency is achieved by the birefringent wavevector matching.

The other parameter in Eq.(4) that plays an important role in the noncollinear phase-matching condition is $\boldsymbol{k}_{\mathrm{p}}$, which is determined by the pump wavelength. Figure 2 shows the relation between the dispersion curve of the 248 cm^{-1} polariton mode in $LiNbO_3$ and phase-matching curves at a phase-matching angle of $\theta = 0.7^\circ$ under pump wavelengths of 532 nm, 694.3 nm, and 1064 nm, respectively. The corresponding theoretical Stokes shifts are 108 cm^{-1}, 83.28 cm^{-1} and 54.61 cm^{-1}, respectively. In other words, the frequencies of the THz waves available are about 3.24 THz, 2.5 THz and 1.64 THz, respectively. Hence, it is clear that the tuning characteristics of a LN-TPO are different at the different pump wavelengths. From another point of view, we can see that it is possible for a TPO to generate tunable THz waves by continuously varying the pump wavelength if the incident angle θ_{ext} of the pump relative to the TPO resonator axis is held to be fixed at a certain value. We have defined this tuning method of TPO as the pump-wavelength tuning technique.[18]

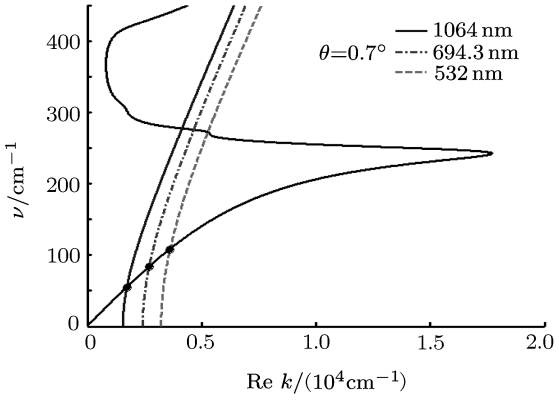


Fig.2. Relation between the dispersion curve of the 248 cm^{-1} polariton mode in LN and the phase-matching curves for the different pump wavelengths at a fixed phase-matching angle of 0.7° designated in terms of wave number.

Figure 3 shows the relationship between the phase-matching angle θ and pump wavelength for generating 1–3 THz THz-waves. We can see that for the generation of a THz wave at a specific frequency, the required phase-matching angle θ decreases with the pump wavelength, indicating the increase of the parametric interaction length of the three waves and the improvement in the TPO performance. Although the opposite condition occurs at the longer wavelength pump, it is quite possible for the larger incident angle θ_{ext} (together with θ which satisfies Snell's Law[19]) to allow the shorter resonator employed to completely separate the pump and Stokes beams at the resonator mirror in the noncollinear TPO geometry. In addition, for THz-wave generation within the same frequency-tuning range, the changes of phase-matching angle, $\Delta\theta$, increase with the pump wavelength, as shown in Fig.4.

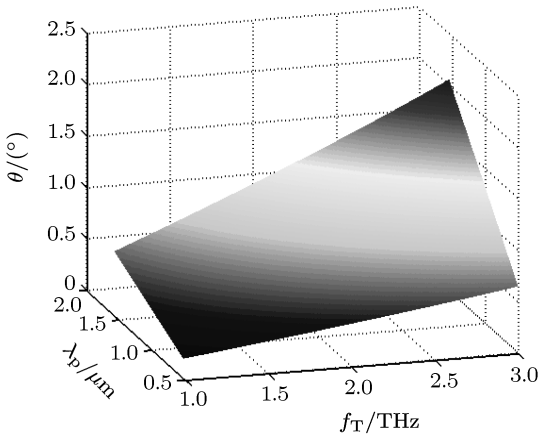


Fig.3. Phase-matching angle θ as a function of the pump wavelength λ_{p} and the THz-wave frequency f_{T}.

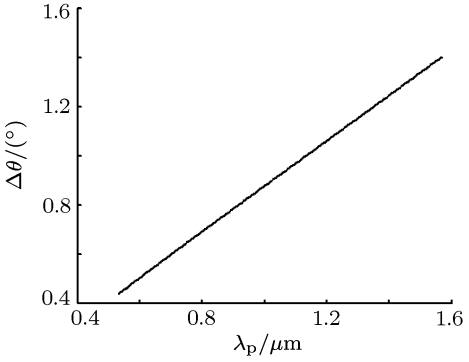


Fig.4. Changes $\Delta\theta$ of phase-matching angle vs pump wavelength λ_p under THz-wave generation of 1–3 THz.

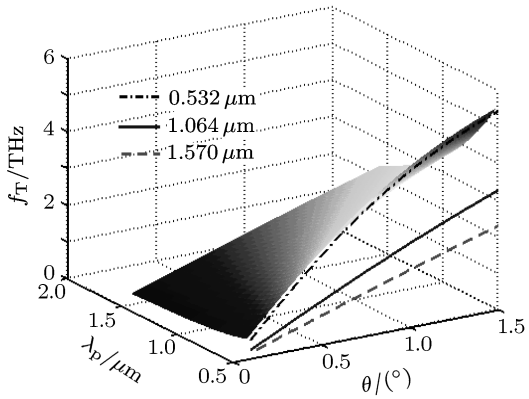


Fig.5. THz-wave frequency f_T as a function of phase-matching angle θ under different pump wavelengths λ_p. Three projection curves in $\theta - f_T$ plane represent the cases under the pump wavelength of 0.532 μm, 1.064 μm and 1.57 μm, respectively.

Figure 5 shows the THz-wave tuning range under different pump wavelengths when θ is adjusted over a range of 0.1°–1.5°. The three projection curves in the $\theta - f_T$ plane from the three-dimensional picture correspond to three pump wavelengths of 0.532 μm, 1.064 μm and 1.57 μm, respectively. We can see that a short-wavelength pump results in a wider THz-wave frequency tuning range for a given angle-tuning range. In other words, the relatively rapid frequency tuning of a LN-TPO can be achieved in short-wavelength pump scheme. By comparison, as for the case of long wavelength pump, although the frequency tuning of TPO is slightly insensitive to the variation of the phase-matching angle, the fine-frequency tuning of THz wave may be easily achievable.

3. Pump wavelength dependence of THz-wave parametric gain

It can be seen clearly from the above analyses that the frequency tuning characteristics of TPOs are highly dependent on the pump wavelength. Besides, the THz-wave parametric gain characteristics of the stimulated polarition scattering as well as its absorption inside the crystal are also responsible for the tuning capability of a TPO. The analytical expressions of the exponential gain for the THz and Stokes waves are given by[11,14,20]

$$g_T = g_S \cos\varphi = \frac{\alpha_T}{2}\left\{\left[1 + 16\cos\varphi\left(\frac{g_0}{\alpha_T}\right)^2\right]^{\frac{1}{2}} - 1\right\}, \tag{5}$$

where ϕ is the phase-matching angle between the pump and the THz wave, α_T is the absorption coefficient of the THz wave in the nonlinear crystal, and g_0 is the parametric gain in the low-loss limit. In CGS units system, g_0 and α_T can be written as

$$g_0 = \sqrt{\frac{\pi\omega_s\omega_T I_p}{2c^3 n_p n_s n_T}}\left(d'_E + \sum_j \frac{S_j\omega_{0j}^2 d'_{Qj}}{\omega_{0j}^2 - \omega_T^2}\right), \tag{6}$$

$$\alpha_T = 2\left|\mathrm{Im}k_T\right| = 2\frac{\omega_T}{c}\mathrm{Im}\left(\varepsilon_\infty + \sum_j \frac{S_j\omega_{0j}^2}{\omega_{0j}^2 - \omega_T^2 - \mathrm{i}\omega_T\Gamma_j + M(\omega_T)}\right)^{\frac{1}{2}}, \tag{7}$$

where I_p is the pump intensity, n_p, n_s and n_T denote the indexes of refraction at the wavelengths of pump beam, Stokes wave and THz wave respectively. The nonlinear coefficients d'_E and d'_{Qj} represent the second- and the third-order nonlinear processes, respectively. The $M(\omega_T)$ is the frequency-dependent damping constant of the low-frequency modes below the 248 cm^{-1} A_1-symmetry mode of LN.

Figure 6(a) shows the calculated THz-wave gain coefficient as a function of pump wavelength and THz frequency at a pump intensity of $I_{\rm p} = 200$ MW/cm^2, calculated by using Eq.(5). As can be seen, the THz-wave gain coefficient increases with the decrease of pump wavelength and the peak value of the gain curve increases more remarkably in the short pump wavelength region than in the long one. Hence, it is found that a short-wavelength-pump scheme in TPOs can provide efficient improvements in the conversion efficiency and effective reduction of the TPO oscillation threshold. In addition, it is also found that by using a short-wavelength pump with a relatively low pump intensity, one can achieve a THz-wave gain which is only achieved by use of a higher pump intensity at the often used 1.064-μm pump (see Fig.6(b)). However, the high-power, short-wavelength pump can easily bring about the remarkable photorefractive damage inside the LN. The occurrence of this undesirable phenomenon will not only severely destroy the phase-matching condition, but also have an obviously negative effect on the spatial coherence and spot quality of THz wave in its direction of propagation. Therefore, for practical operation, measures should be taken to relieve this problem to a certain extent.

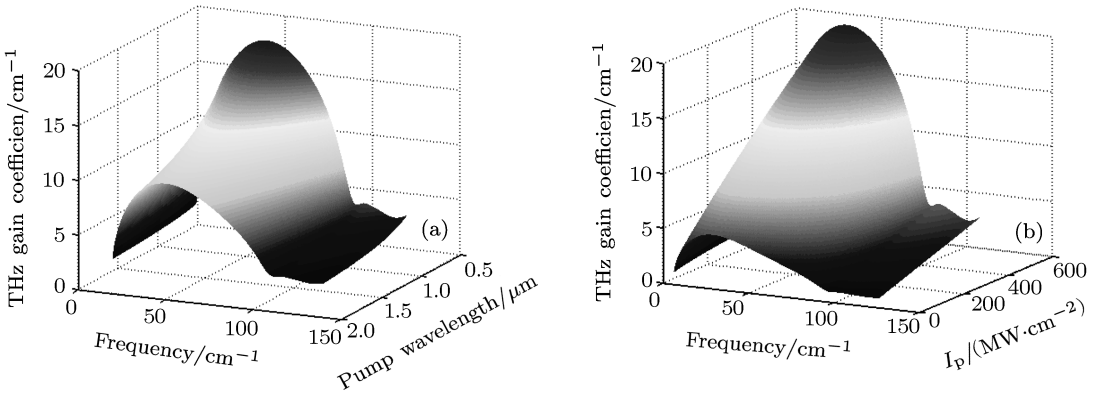


Fig.6. Three-dimensional plot of the calculated THz-wave gain coefficient as a function of THz-wave frequency: (a) pump wavelength at the fixed pump intensity of $I_{\rm p} = 200$ MW/cm^2; (b) pump intensity under the pump wavelength of 1.064 μm.

4. Characteristics of THz-wave output direction based on Si-prism

In order to efficiently extract THz waves generated in LN and to substantially improve the output directionality of THz radiation from TPOs, the arrayed prisms fabricated from silicon with low dispersion in the THz region are often employed as the THz-wave coupler.[9,12] Generally, a proper base angle α of Si-prisms is chosen to make the THz wave emerge almost normally to the exit surface of the Si-prism ($\delta \approx 0°$) (see Fig.1(b)) to facilitate the confirmation of the output direction of THz wave. According to Eqs.(3) and (4) as well as Snell's law, we calculate the pump-wavelength dependence of the base angle α of Si-prisms for the THz-wave tuning range of 100–600 μm at different pump wavelengths, as shown in Fig.7(a). Here, the refractive index of Si is assumed to be 3.4 in the THz region. The prism base angle α increases with the decrease of pump wavelength, and is approximately a constant over the whole THz-wave tuning range, especially in the short-wavelength-pump scheme. We define and calculate the optimum prism base angle, α', for different pump wavelengths when the radiation angle δ is set to be zero for the THz-wave generation at a wavelength of 200 μm, as shown in Fig.7(b). Figure 8 shows the calculated radiation angle δ under the optimum prism base angles corresponding to respective pump wavelengths. It can be found that the radiation angle is highly dependent on the pump wavelength, and the changes of the radiation angle, $\Delta\delta$, almost increase linearly with the pump wavelengths under the same THz-wave tuning range, as shown in Fig.9. Therefore, a long-wavelength-pump scheme will lead to a large radiation angle of THz wave, and the stability of the output direction may be slightly unsatisfactory. It should be noted, however, that since the TPO cavity is angle-tuned in the direction opposite to the radiation

angle δ, the actual changes of radiation angle are much less than the values calculated here.[9]

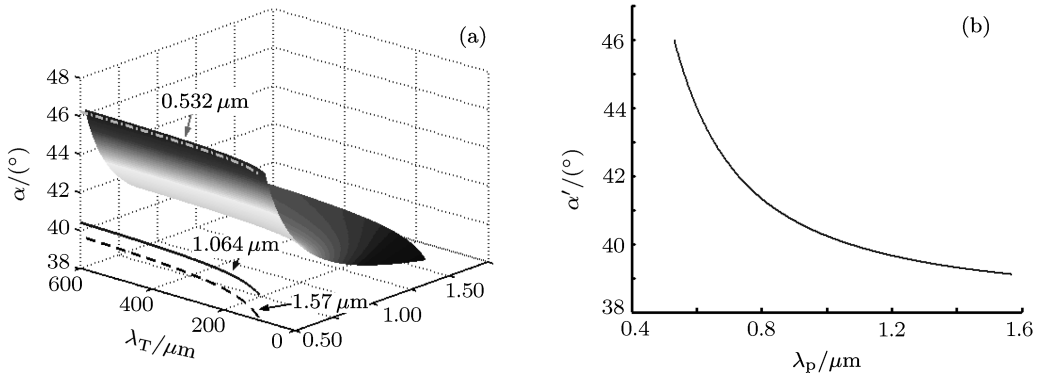


Fig.7. (a) Calculated base angle α of Si-prism as a function of THz and pump wavelengths (three projection curves in $\lambda_T - \alpha$ plane correspond to the cases under the pump wavelength of 0.532 μm, 1.064 μm and 1.57 μm, respectively); (b) the optimum base angle α' of prism vs pump wavelength under $\delta = 0°$ at $\lambda_T = 200$ μm as a reference.

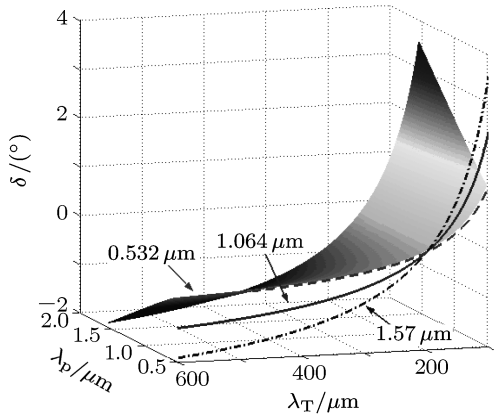


Fig.8. Three-dimensional plot of the calculated radiation angle based on optimum base angle of prism as a function of pump wavelength λ_p and THz wavelengths λ_T. Three projection curves in $\lambda_T - \delta$ plane show the cases under the pump wavelength of 0.532 μm, 1.064 μm and 1.57 μm, respectively.

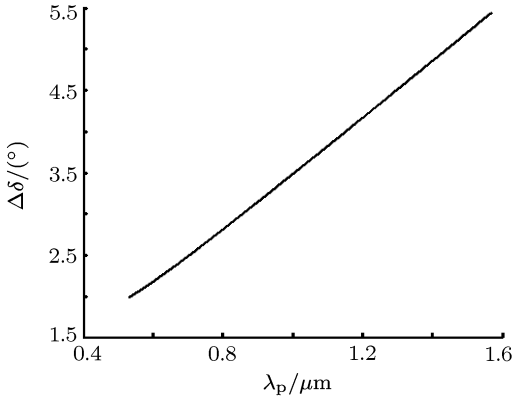


Fig.9. Changes of radiation angle $\Delta\delta$ of THz wave vs pump wavelength λ_p under the same THz-wave tuning range.

On the other hand, if the Si-prism with a specific base angle not corresponding to respective pump wavelengths is used in TPO, the output direction of THz wave will diverge remarkably away from the normal direction to the exit surface of the Si-prism. Figure 10 shows the radiation angle of THz wave under the often used prism with a base angle of $\alpha = 40°$ corresponding to the 1.064-μm pump. It can be seen that the radiation angle δ at short pump wavelengths is much larger than that at long ones. However, it is noteworthy that the changes $\Delta\delta$ of radiation angle within the THz-wave tuning range are almost the same as those under the optimum prism base angle,

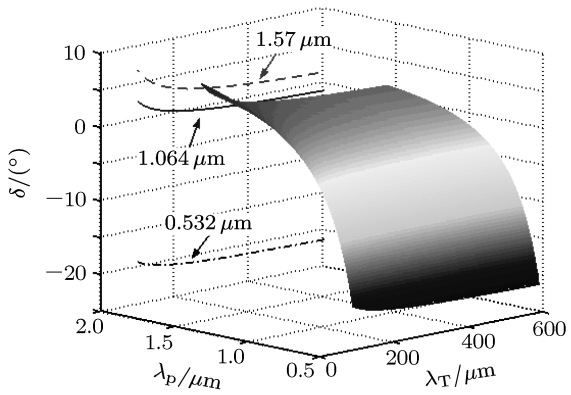


Fig.10. Calculated radiation angle δ of THz wave for the Si-prism coupler with base angle of $\alpha = 40°$ under different pump wavelengths λ_p. Three projection curves in $\lambda_T - \delta$ plane correspond to the cases under the pump wavelengths of 0.532 μm, 1.064 μm and 1.57 μm, respectively.

as shown in Fig.11. It means that the choice of prism base angle affects the radiation angle δ severely, but has little effect on the stability of the output direction of THz wave within the THz-wave tuning range.

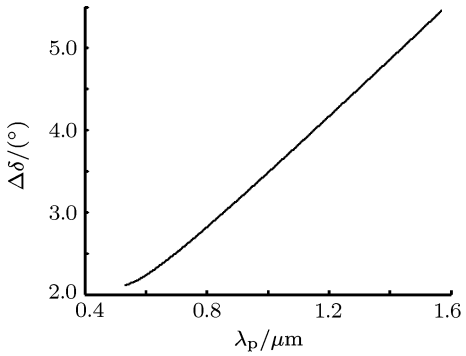


Fig.11. Changes of radiation angle $\Delta\delta$ of THz wave vs pump wavelength λ_p under the base angle $\alpha = 40°$.

It should be emphasized that since some parameters, such as the lattice vibration parameters and refractive indices of the LN crystal in the different regions, used in this paper are mainly cited from the Refs.[17] and [21], and are probably different from those adopted in other publications,[8−15] there may be a slight, but negligible, discrepancy between our results and others. However, we think that the final conclusions in our paper still apply.

5. Conclusions

In summary, we have studied the pump-wavelength dependence of the performance of LN-TPOs. The simulations indicate that a short-wavelength-pump scheme can not only increase the THz-wave gain, but also lead to a small phase-matching angle under the THz-wave generation at a specific frequency, which is helpful to increase the spatial overlap of the three parametric beams. Furthermore, the stability of THz-wave output direction of TPO based on the Si-prism coupler can also be remarkably improved. By comparison, a long-wavelength-pump scheme can avoid the photorefractive damage in crystals to some extent. Although large incident angles of the pump beam with respect to the TPO resonator axis is needed for the generation of a THz wave at a certain frequency in this scheme, short TPO cavities can be adopted in this noncollinear phase-matching geometry, which is of great advantage to the improvement in the conversion efficiency and the reduction of the oscillation threshold.

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