Theoretical analysis of stimulated polariton scattering from the A_1-symmetry modes of $KNbO_3$ crystal*

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Stimulated polariton scattering (SPS) based on noncollinear phase matching scheme from the A_1-symmetry modes of $KNbO_3$ crystal is investigated for generating terahertz (THz) wave. Frequency tuning characteristics of THz wave by varying the phase matching angle and pump wavelength are analyzed. The expression of the effective parametric gain length under the noncollinear phase matching condition is deduced. Parametric gain and absorption characteristics of THz wave in $KNbO_3$ are theoretically simulated. The characteristics of $KNbO_3$ for parametric oscillator (TPO) are compared with those of MgO:$LiNbO_3$. The analysis results indicate that $KNbO_3$ is an excellent optical crystal for TPO to enhance the output of THz wave.

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Over the past two decades, with the ever-increasing number of applications for terahertz (THz) radiation, such as imaging, biology and medicine, communications, security technologies and quality control[1-6], there is growing demand for THz sources with excellent performance. Among many electronic and optical methods for THz wave generation, terahertz parametric oscillator (TPO)[7] based on stimulated polariton scattering (SPS) processes exhibits many advantages, such as narrow linewidth, coherent, wide tunable range, high power output and room temperature operation. In SPS, the interaction of a fundamental laser field with a polariton mode of a crystal generates THz wave and Stokes wave. Typically, the refractive index of THz wave is substantially larger than that for the optical pump wave, and phase matching is impossible for collinear interactions. Noncollinear phase matching for THz generation can perform well[8-11]. However, noncollinear phase matching configuration, in which the pump wave, the Stokes wave and the THz wave are all non-parallel with each other, significantly reduces parametric gain. So it is of vital importance to increase the effective parametric gain length in the noncollinear phase matching configuration.

A frequently employed material for TPO is the nonlinear optical crystal of MgO:$LiNbO_3$, because of its relatively large second-order optical nonlinearity and its wide transparency range[12]. Unfortunately, the quantum conversion efficiency of the TPO is extremely low as the THz wave is intensely absorbed by MgO:$LiNbO_3$ crystal. At the frequency of 1.5 THz, the absorption coefficient is about 45 cm^{-1}[13]. $KNbO_3$ crystal is an attractive material for the nonlinear optical interaction between optical and THz waves due to its wide transmission range (0.4—4.5 μm)[14], a high nonlinear coefficient (d_{33}=27.4 pm/V at 1 064 nm)[15] and a relatively high optical damage threshold of 350 MW/cm^2[16]. $KNbO_3$ has four infrared- and Raman-active phonon modes called as A_1-symmetry modes, which are located at 190 cm^{-1}, 290 cm^{-1}, 299 cm^{-1} and 687 cm^{-1}[17]. When pump excitation is sufficiently strong, THz wave can be generated from the efficient parametric scattering of laser light via SPS.

In this paper, we theoretically study the characteristics of $KNbO_3$ for TPO with a noncollinear phase matching scheme. We analyze the frequency tuning characteristics of the THz wave by varying the phase matching angle and pump wavelength. The expression of the effective parametric gain length under the noncollinear phase matching

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condition is deduced. Gain and absorption characteristics of the THz wave in $KNbO_3$ and MgO:$LiNbO_3$ are investigated.

A surface-emitted TPO with a noncollinear phase matching scheme comprises a single-resonant optical parametric oscillator with a Fabry-Perot cavity, as shown in Fig.1. The configuration was first reported by T. Ikari et al[18]. The nonlinear optical crystal is $KNbO_3$ crystal. The resonant cavity for the Stokes wave consists of two plane-parallel mirrors of M_1 and M_2 with high reflectance. The pump wave with nanosecond pulsewidth passes through the cavity at the edge of M_1 and M_2, and the Stokes wave propagates along the x axis of $KNbO_3$. THz wave vector perpendicular to the output surface is achieved by setting the incident angles of pump wave to the crystal surface. The polarizations of pump wave, Stokes wave and THz wave are all along the z-axis of the $KNbO_3$ crystal. θ is the angle between the vectors of the pump wave and the Stokes wave within the crystal, and φ is the angle b etween the vectors of the pump wave and the THz wave within the crystal.

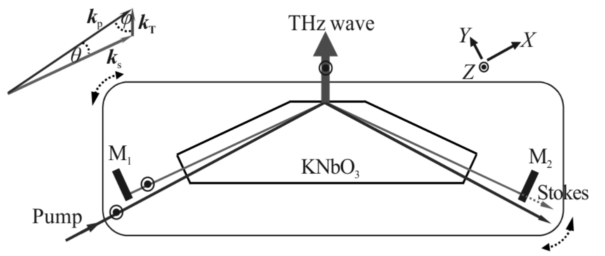

Fig.1 Schematic diagram of the surface-emitted TPO using $KNbO_3$ with a noncollinear phase matching scheme

The theoretical values of the refractive index are calculated by Sellmeier equation for $KNbO_3$ in the infrared range at 22°C[14] and in the THz range[17], respectively. In this paper, the theoretical parameters for $KNbO_3$ are taken from Ref.[17].

For the tunable THz wave generation, two requirements have to be fulfilled, which are the energy conservation law of $\omega_p=\omega_s+\omega_T$ and the noncollinear phase matching condition of $\boldsymbol{k}_p = \boldsymbol{k}_s + \boldsymbol{k}_T$, as shown in the insets of Fig.1. Here, ω_p, ω_s and ω_T are the angular frequencies, while $\boldsymbol{k}_p$, $\boldsymbol{k}_s$ and $\boldsymbol{k}_T$ are the wavevectors of the pump, Stokes and THz waves, respectively. The phase matching condition can be rewritten as $\boldsymbol{k}_T^2 = \boldsymbol{k}_p^2 + \boldsymbol{k}_s^2 - 2\boldsymbol{k}_p\boldsymbol{k}_s\cos\theta$. Varying one of the parameters in noncollinear phase matching condition, such as the angle θ and the pump wavelength λ_p, we can obtain a family of the phase matching curves. Fig.2 shows the dispersion curve of the A_1-symmetry polariton modes in $KNbO_3$ and the phase matching curves for the 1 064 nm laser pump. When the phase matching curves are superimposed on the dispersion curve of the A_1-symmetry polariton modes, the points of the intersection of these curves are expected to determine the allowed frequencies and wave vectors of THz wave. As the angle θ is changed continuously, the frequency tuning of the THz wave is realized simultaneously, which is the basic principles of the so-called angle-tuning method of TPO. When the angle θ varies from 0° to 7.3°, the phase matching curves and the dispersion curve of the A_1-symmetry polariton modes intersect, which means THz wave can be generated.

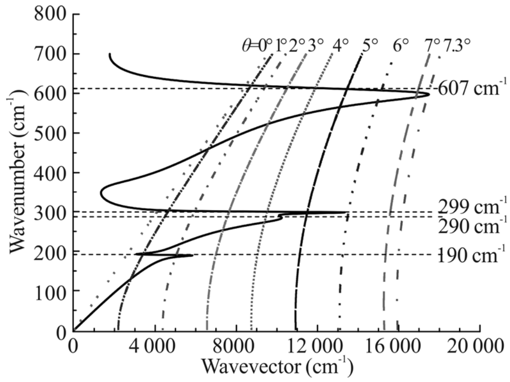

Fig.2 Dispersion curve of the A_1-symmetry polariton modes in $KNbO_3$ and the phase matching curves for the 1 064 nm laser pump

According to the noncollinear phase matching condition, the tuning of THz wave can be realized by varying the pump wavelength λ_p. Fig.3 shows the dispersion curve of the A_1-symmetry polariton modes in $KNbO_3$ and the phase matching curves at a fixed phase matching angle θ of 1° when the pump wavelengths are 400 nm, 633 nm, 1 064 nm, 1 550 nm and 3 000 nm, respectively. We find from Fig.3 that when the pump wavelength λ_p changes, the intersection points between the phase matching curves and the dispersion curve of the A_1-symmetry polariton modes change, which means that the frequency tuning of the THz wave is realized simultaneously. The shorter the pump wavelength is, the higher frequency region the intersection point will shift to. That is to say, the higher frequency THz wave can be achieved.

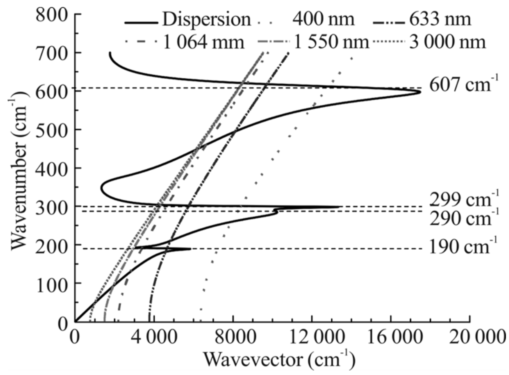

Fig.3 Dispersion curve of the A_1-symmetry polariton modes in $KNbO_3$ and the phase matching curves at room temperature at a fixed phase matching angle θ of 1° when the pump wavelengths are 400 nm, 633 nm, 1 064 nm, 1 550 nm and 3 000 nm

Next, we deduce the expression of the effective parametric gain length under the noncollinear phase matching condition based on the theoretical model proposed in Ref.[19]. Assuming that these three mixing waves have Gaussian profiles, the Stokes spot size is narrowed by the gain polarization and broadened by the diffraction simultaneously. The balance determines the final Stokes wave spot size. The relationship between the pump wave radius w_p and the Stokes wave radius w_s is given by

$$\left(\frac{\pi}{2L\lambda_s}\right)^2\left(\frac{w_p^2}{w_p^2+2w_s^2}\right)^3+\frac{w_p^2w_s^2}{w_p^2+2w_s^2}-\frac{w_p^2}{2}=0, \tag{1}$$

where λ_s is the wavelength of the Stokes wave, and L is the optical cavity length. The walkoff length l_ω is given by

$$l_\omega=\frac{\sqrt{\pi}}{2}\frac{w_p}{\theta}\sqrt{\frac{w_p^2+w_s^2}{w_p^2+w_s^2/2}}, \tag{2}$$

where θ is used as a substitute for the double refraction walkoff angle. The effective parametric gain length L_{eff} is given by

$$L_{eff}=l_\omega erf\left(\frac{\sqrt{\pi}}{2}\frac{l}{l_\omega}\right), \tag{3}$$

where l is the crystal length which is the propagation length of Stokes wave within the $KNbO_3$ crystal. The effective parametric gain length L_{eff} versus the pump wavelength λ_p is shown in Fig.4 when frequencies of THz wave are 1 THz, 2 THz, 3 THz, 4 THz and 5 THz, respectively. We find that as the pump wavelength increases, the effective parametric gain length gradually decreases. The reason is that as the pump wavelength increases, the phase matching angle θ is enlarged. The pump wave in the short wavelength region can effectively lengthen the effective parametric gain length. Compared with those with frequencies of 2 THz, 3 THz, 4 THz and 5 THz, the effective parametric gain length with frequency of 1 THz is maximum. This is because the phase matching angle θ is minimum as THz wave frequency is 1 THz.

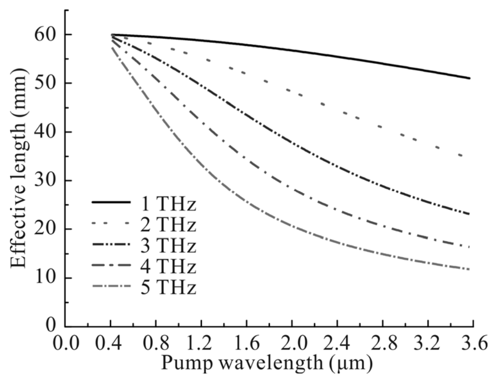

Fig.4 The effective parametric gain length versus the pump wavelength when the length of resonant cavity is 80 mm, l=60 mm and w_p=1 mm

Fig.5 shows the effective parametric gain length versus the radius of pump wave when frequencies of THz wave are 1 THz, 2 THz, 3 THz, 4 THz and 5 THz, respectively. We find that the effective parametric gain length increases rapidly and smoothly with the increase of radius of pump wave. The pump wave with a large beam radius can generate the Stokes wave and the THz wave with a large beam radius simultaneously, resulting in a long effective parametric gain length.

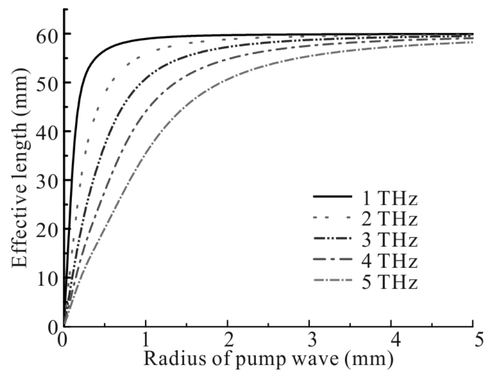

Fig.5 The effective parametric gain length versus the radius of pump wave when the length of resonant cavity is 80 mm, l=60 mm and λ_p=1 064 nm

Fig.6 shows the effective parametric gain length versus the crystal length when the frequencies of THz wave are 1 THz, 2 THz, 3 THz, 4 THz and 5 THz, respectively. We find from Fig.6 that the effective parametric gain length increases rapidly with the increase of crystal length as the frequencies are 1 THz, 2 THz, 3 THz, and increases smoothly as the frequencies are 4 THz and 5 THz. At lower frequency band of THz wave, the pump and Stokes waves are almost overlapped as the phase matching θ is small. On the contrary, the pump and Stokes waves separate fast as these two beams are partially overlapped at higher frequency band.

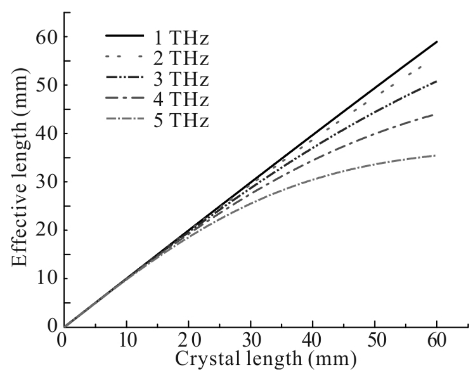

Fig.6 The effective parametric gain length versus the crystal length when the length of resonant cavity is 80 mm, w_p=1 mm and λ_p=1 064 nm

For the efficient generation of THz wave, parametric gain is another crucial parameter. According to the Ref.[20], the analytical expression of the THz wave parametric gain g_T under the noncollinear phase matching condition can be written as

$$g_T = \frac{\alpha_T}{2}\left\{\left[1+16\cos\varphi\left(\frac{g_0}{\alpha_T}\right)^2\right]^{\frac{1}{2}}-1\right\}, \tag{4}$$

$$g_0^2 = \frac{\pi\omega_s\omega_T}{2c^3 n_p n_s n_T} I_P \left(d_E' + \sum_j \frac{S_j\omega_0^2 d_{Qj}'}{\omega_{0j}^2-\omega_T^2}\right)^2, \tag{5}$$

$$\alpha_T = 2\frac{\omega_T}{c}\mathrm{Im}\left(\varepsilon_\infty + \sum_j \frac{S_j\omega_{0j}^2}{\omega_{0j}^2-\omega_T^2-\mathrm{i}\omega_T-\Gamma_j}\right)^{\frac{1}{2}}, \tag{6}$$

where α_T is absorption coefficient in THz region, ω_{0j}, S_j and Γ_j denote eigenfrequency, oscillator strength of the polariton modes and the bandwidth of the jth A_1-symmetry phonon mode in the $KNbO_3$ crystal, respectively. I_p is the power density of pump wave, g_0 is the low-loss parametric gain. n_p and n_s are the refractive indices of the pump and Stokes waves, respectively. d_E' and d_Q' are nonlinear coefficients related to pure parametric (second-order) and Raman (third-order) scattering processes, respectively. Far below the lowest A_1-symmetry polariton mode 190 cm^{-1}, Eq.(5) can be rewritten as

$$g_0^2 = \frac{\pi\omega_s\omega_T}{2c^3 n_p n_s n_T} I_P \left(d_E' + \sum_j S_j d_{Qj}'\right)^2. \tag{7}$$

Nonlinear coefficients d_E' and d_Q' relate to electro-optic coefficient r, i.e.,

$$d_E' + \sum_j S_j d_{Qj}' = r n_T^4. \tag{8}$$

So the low-loss parametric gain coefficient g_0 is rewritten as

$$g_0^2 = \frac{\pi\omega_S\omega_T}{2c^3 n_p n_s n_T} I_P (r n_T^4)^2. \tag{9}$$

According to Eqs.(4)—(9), we show the parametric gain coefficient g_T at a pump intensity of 100 MW/cm^2 and the absorption coefficient α_T in $KNbO_3$ and $MgO:LiNbO_3$ as shown in Fig.7. From Fig.7, we find that as the frequencies of THz wave are far below the lowest A_1-symmetry polariton mode 190 cm^{-1}, both the parametric gain coefficients g_T in $KNbO_3$ and $MgO:LiNbO_3$ are in the order of several per centimeter. The gain coefficients in $KNbO_3$ are larger than those in $MgO:LiNbO_3$ at most region. Meanwhile, the absorption coefficients α_T of THz wave in $KNbO_3$ are much lower than those in $MgO:LiNbO_3$. Compared with $MgO:LiNbO_3$, $KNbO_3$ has larger gain coefficients at most region and lower absorption coefficients, so $KNbO_3$ is a suitable selection for TPO to enhance the output of THz-wave.

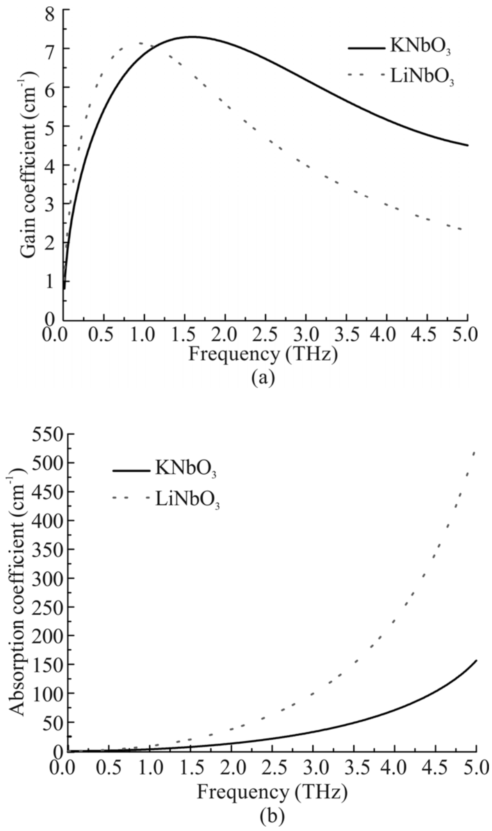

Fig.7 THz wave (a) parametric gain coefficients g_T and (b) absorption coefficient α_T in $KNbO_3$ and $MgO:LiNbO_3$ at room temperature with λ_p=633 nm and I_p=100 MW/cm^2

SPS from the A_1-symmetry modes of $KNbO_3$ crystal for generating THz wave with a noncollinear phase matching scheme is investigated. The wide tuning THz wave can be generated by varying the phase matching angle and pump wavelength. The pump wave with shorter wavelength and larger beam radius can effectively lengthen the effective parametric gain length under the noncollinear phase matching condition. Compared with $MgO:LiNbO_3$, $KNbO_3$ has larger gain coefficients at most region and lower absorption coefficients, so $KNbO_3$ is an excellent optical crystal for TPO.

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