



## Article

# Simultaneous Generation of Two Pairs of Stokes and Terahertz Waves from Coupled Optical Parametric Oscillations with Quasi-Phase-Matching

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**Abstract:** We present a theoretical investigation on simultaneous generation of two pairs of Stokes and terahertz (THz) waves from coupled optical parametric oscillations (OPOs) with a quasi-phase-matching (QPM) scheme. The two pairs of Stokes and THz waves are generated by stimulated polariton scattering (SPS) from periodically-inverted GaP. By analyzing the QPM conditions of coupled OPOs we find that the two THz waves with any frequency below the transverse optical (TO) mode frequency of GaP can be simultaneously generated with a suitable pump wavelength. We calculate the photon flux densities of the two THz waves by solving the coupled wave equations. The calculation results indicate that the two THz waves can be efficiently generated with high pump intensities, particularly in lower THz frequency band.

**Keywords:** terahertz wave; stimulated polariton scattering; coupled optical parametric oscillation; GaP

## 1. Introduction

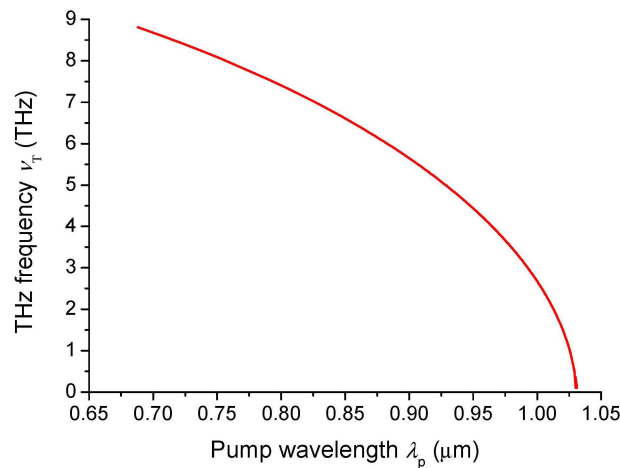
In recent years, it has been demonstrated that coupled optical parametric oscillations (OPOs) can simultaneously generate two pairs of signal and idler waves from periodically-inverted KTiOPO<sub>4</sub> (KTP) plates by a single pump wavelength [1,2]. The wavelengths for each pair of signal and idler waves satisfy quasi-phase-matching (QPM) conditions. The wavelengths of the signal and idler waves are near infrared when pump wavelength is 532 nm. Based on the research above, we consider that the frequencies of the two idler waves generated from coupled OPOs with periodically-inverted GaP by stimulated polariton scattering (SPS) can extend to terahertz (THz) frequencies. The frequency separation between the two THz waves can be tuned by selecting the thickness of each GaP plate. The generated two THz waves are useful for imaging and food inspection [3,4].

SPS which was used in MgO: LiNbO<sub>3</sub>, LiTaO<sub>3</sub>, and KTiOPO<sub>4</sub>, has proved to be an efficient scheme to generate THz waves [5–12]. SPS can also be used in zinc-blende structure crystals, since the crystals have a high nonlinear coefficient and a low THz wave absorption coefficient [13,14]. GaP which has an infrared- and Raman-active transverse optical (TO) mode with frequency of 367 cm<sup>−1</sup> is an attractive material for THz wave generation via SPS [15] since GaP has a high second-order nonlinear coefficient ( $d_{36} = 70.6$  pm/V at 1064 nm) [16] and a low THz wave absorption coefficient below TO mode frequency [15]. Moreover, collinear configuration can be realized in GaP by using cross-Reststrahlen band dispersion compensation phase-matching [17].

In this work, we theoretically study simultaneous generation of two pairs of Stokes and THz waves from coupled OPOs with periodically-inverted GaP by SPS. The QPM conditions of the coupled OPOs are analyzed. We calculate the photon flux densities of the two THz waves by solving the coupled wave equations.

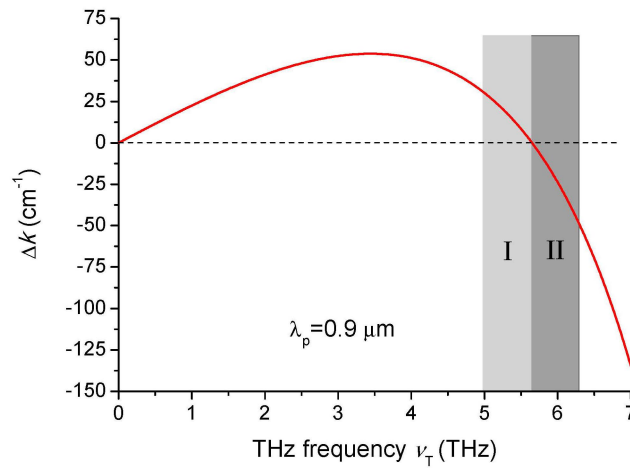
## 2. Phase-Matching Characteristics

In the SPS processes with cross-Reststrahlen band dispersion compensation phase-matching, pump and Stokes waves are in the near-infrared transmission window of GaP crystal, and THz wave is in the far-infrared transmission window, on the other side of the crystal's Reststrahlen band. Since the refractive indices of the three waves in GaP are approximately equal, the collinear phase-matching condition can be realized. Figure 1 shows THz frequencies  $\nu_T$  versus pump wavelengths  $\lambda_p$  with collinear phase-matching condition in GaP. The theoretical values of the refractive index are calculated using a Sellmeier equation for GaP in the infrared range [17] and in the THz range [15], respectively. From the figure we find that pump wavelengths  $\lambda_p$  in the range of 0.69–1.03  $\mu\text{m}$  fulfill collinear phase-matching, corresponding to the THz frequencies  $\nu_T$  in the range of 0.15–8.8 THz.



**Figure 1.** THz frequencies  $\nu_T$  versus pump wavelengths  $\lambda_p$  with collinear phase-matching condition in GaP.

Figure 2 shows phase mismatching  $\Delta k$  among collinear SPS processes in GaP when  $\lambda_p = 0.9 \mu\text{m}$ .  $\Delta k = \vec{k}_p - \vec{k}_s - \vec{k}_T$ , where  $\vec{k}_p$ ,  $\vec{k}_s$ , and  $\vec{k}_T$  are the wave vectors of pump, Stokes, and THz waves respectively. From the figure we find that  $\Delta k$  is zero at 5.65 THz, where phase-matching can realize. At area I where frequencies are slightly smaller than 5.65 THz,  $\Delta k$  is slightly larger than zero, whereas at area II where frequencies are slightly larger than 5.65 THz,  $\Delta k$  is slightly smaller than zero. At area I the phase mismatch  $\Delta k$  in optical parametric oscillation (OPO) can be compensated by grating vector  $\vec{k}_\Lambda$  of periodically-inverted GaP.  $k_\Lambda = 2\pi/\Lambda$  and  $\Lambda$  is the QPM period. Simultaneously, at area II, the phase mismatch  $\Delta k$  in the other OPO can also be compensated by the same grating vector  $\vec{k}_\Lambda$ . As a result, the two phase mismatch of the two OPOs can be simultaneously compensated by the same grating vector  $\vec{k}_\Lambda$ . The two OPOs will couple with each other with a same pump wavelength and a same QPM period.



**Figure 2.** Phase mismatching  $\Delta k$  versus THz frequencies  $\nu_T$  with collinear phase-matching condition in GaP,  $\lambda_p = 0.9 \mu\text{m}$ .

In order to achieve efficient conversion of coupled OPOs from pump wave to THz wave, a precise phase-matching condition must be satisfied. Figure 3 shows the QPM scheme of coupled OPOs with periodically-inverted GaP. In order to simultaneously satisfy the two QPM conditions, accurate value of grating vector  $\vec{k}_\Lambda$  is required. The two QPM conditions of the coupled OPOs are

$$\vec{k}_p - \vec{k}_{s1} - \vec{k}_{T1} + \vec{k}_\Lambda = 0 \quad (1)$$

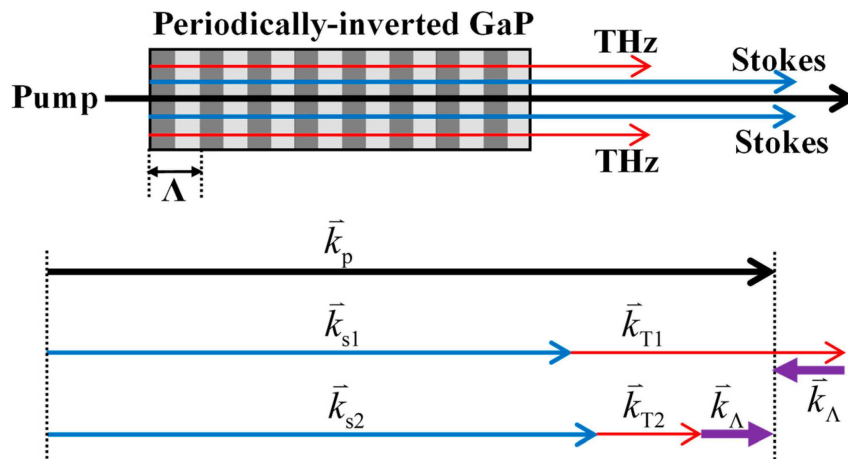
$$\vec{k}_p - \vec{k}_{s2} - \vec{k}_{T2} - \vec{k}_\Lambda = 0 \quad (2)$$

where  $\vec{k}_{s1}$  and  $\vec{k}_{s2}$  are the wave vectors of the two Stokes waves respectively,  $\vec{k}_{T1}$  and  $\vec{k}_{T2}$  are the wave vectors of the two THz waves respectively. The energy conservation condition has to be fulfilled,

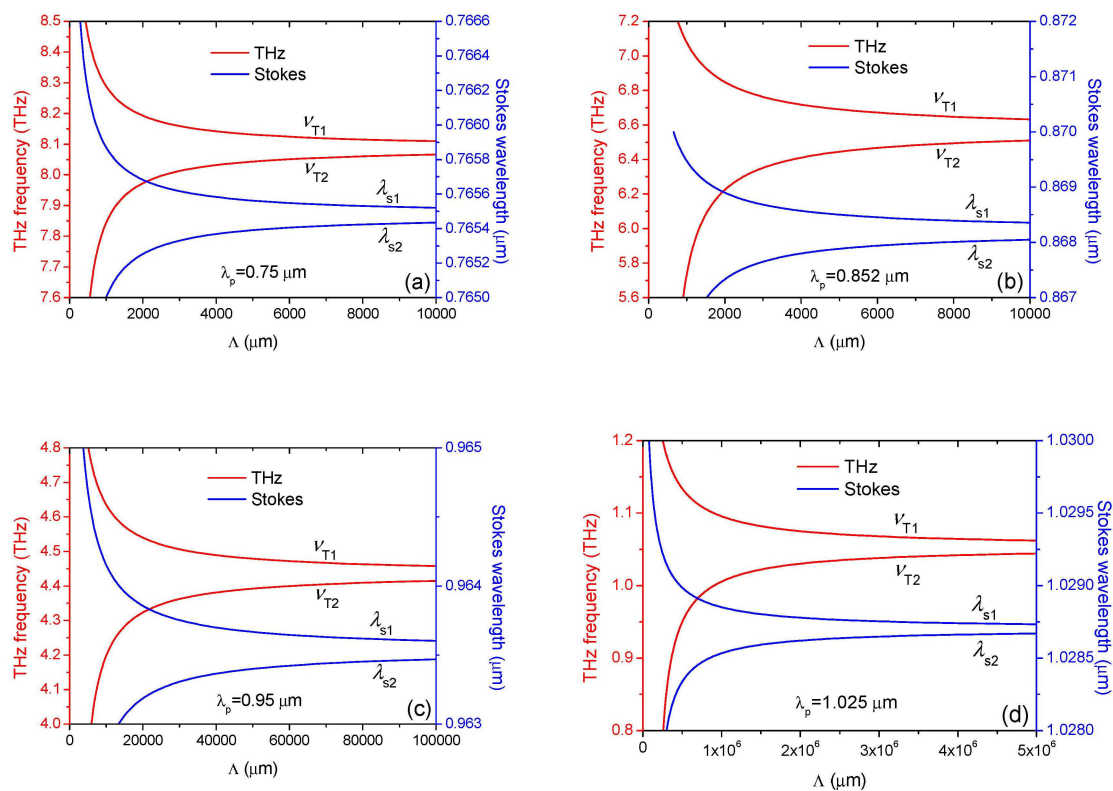
$$\frac{1}{\lambda_p} - \frac{1}{\lambda_{s1}} - \frac{1}{\lambda_{T1}} = 0 \quad (3)$$

$$\frac{1}{\lambda_p} - \frac{1}{\lambda_{s2}} - \frac{1}{\lambda_{T2}} = 0 \quad (4)$$

where  $\lambda_{s1}$  and  $\lambda_{s2}$  are the wavelengths of the two Stokes waves respectively,  $\lambda_{T1}$  and  $\lambda_{T2}$  are the wavelengths of the two THz waves respectively. If the two QPM conditions of the coupled OPOs are simultaneously satisfied, the two THz waves can be simultaneously generated with a single pump wave. Figure 4 shows THz frequencies and Stokes wavelengths versus QPM period  $\Lambda$  with coupled OPOs with different pump wavelengths.  $\nu_{T1}$  and  $\nu_{T2}$  are frequencies of the two THz waves,  $\nu_{T1} = c/\lambda_{T1}$  and  $\nu_{T2} = c/\lambda_{T2}$ ,  $c$  is velocity of light. From the figure we find that two pairs of Stokes and THz waves can be generated by coupled OPOs with a single pump wave. As QPM period  $\Lambda$  changes the THz frequencies and Stokes wavelengths are tuned. The difference frequencies between the two THz waves become small as QPM period  $\Lambda$  increases. The minimum difference frequencies are smaller than 0.01 THz. As pump wavelengths change from 0.75  $\mu\text{m}$  to 0.852, 0.95, and 1.025  $\mu\text{m}$ ,  $\nu_{T1}$  and  $\nu_{T2}$  decrease from 8.1 THz to 1.05 THz. From Figure 4 we conclude that the two THz waves with any frequency from 1.05 THz to 8.1 THz can be simultaneously generated by coupled OPOs with a suitable pump wavelength.



**Figure 3.** Schematic diagram of QPM scheme of the coupled OPOs with periodically-inverted GaP. Pump wave, Stokes waves, and THz waves are collinear.  $\vec{k}_p$  is the wave vector of pump wave and  $\vec{k}_{s1}$  and  $\vec{k}_{s2}$  are the wave vectors of the two Stokes waves respectively,  $\vec{k}_{T1}$  and  $\vec{k}_{T2}$  are the wave vectors of the two THz waves respectively,  $\vec{k}_\Lambda$  is the grating vector of periodically-inverted GaP.



**Figure 4.** THz frequencies and Stokes wavelengths versus QPM period  $\Lambda$  with coupled OPOs. (a)  $\lambda_p = 0.75 \mu\text{m}$ ; (b)  $\lambda_p = 0.852 \mu\text{m}$ ; (c)  $\lambda_p = 0.95 \mu\text{m}$ ; (d)  $\lambda_p = 1.025 \mu\text{m}$ .

### 3. THz Wave Photon Flux Density

The generated THz intensities by SPS processes depend on the nonlinear optical susceptibility and THz absorption coefficients. The nonlinear optical susceptibility in GaP in the THz frequency range is governed by the superposition of electronic and ionic contributions. Faust showed that the ionic and electronic contributions are of opposite sign, leading to a cancellation of both contributions below the TO mode frequency of GaP [18]. Sussman deduced coupled wave equations for SPS processes [19].

In this work we calculate THz intensity based on the theories of Faust and Sussman. The analytical expression of THz parametric gain coefficient  $g_T$  in SPS processes under the QPM condition in the international system of units can be written as [19,20]

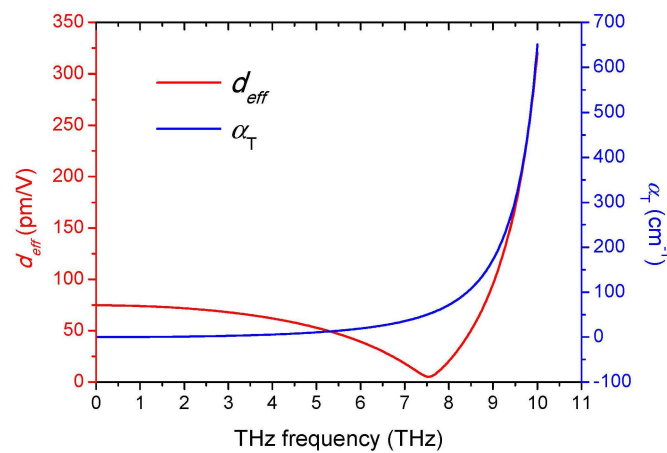
$$g_T = \frac{\alpha_T}{2} \left\{ \left[ 1 + 16 \left( \frac{g_0}{\alpha_T} \right)^2 \right]^{\frac{1}{2}} - 1 \right\} \quad (5)$$

$$g_0^2 = \frac{\omega_s \omega_T}{128 \pi^2 \epsilon_0 c^3 n_p n_s n_T} I_P d_{eff}^2 \quad (6)$$

$$d_{eff} = d_e \left( 1 + C_1 \frac{\omega_{TO}^2}{\omega_{TO}^2 - \omega_T^2 - i \omega_T \Gamma} \right) \quad (7)$$

$$\alpha_T = 2 \frac{\omega_T}{c} \text{Im} \left( \epsilon_\infty + \frac{S \omega_{TO}^2}{\omega_{TO}^2 - \omega_T^2 - i \omega_T \Gamma} \right)^{\frac{1}{2}} \quad (8)$$

where  $\omega_{TO}$ ,  $S$ , and  $\Gamma$  denote eigenfrequency, oscillator strength of the polariton modes, and the bandwidth of the TO mode in GaP crystal, respectively.  $\omega_s$  and  $\omega_T$  are the angular frequencies of Stoke and THz waves, respectively.  $\epsilon_0$  and  $\epsilon_\infty$  are vacuum dielectric constant and high-frequency dielectric constant, respectively.  $I_P$  is the power density of pump wave,  $g_0$  is the low-loss parametric gain.  $n_p$ ,  $n_s$ , and  $n_T$  are the refractive indices of the pump, Stokes and THz waves, respectively.  $\alpha_T$  is material absorption coefficient in THz region.  $d_e$  is the electronic second-order nonlinear coefficient, and  $d_{eff}$  is the bulk value of the second-order nonlinear coefficient involving electronic and ionic contributions.  $C_1$  is a coupling constant, and  $C_1$  is  $-0.53$  for GaP [18]. Figure 5 shows calculated  $d_{eff}$  and  $\alpha_T$  versus THz frequencies. In the calculation  $d_e$  is 70.6 pm/V with pump wavelength of 1.064  $\mu\text{m}$ . From the figure we find that  $\alpha_T$  monotonously increases with frequencies, and  $d_{eff}$  smoothly decreases first and then rapidly increases.  $d_{eff}$  is smaller than 70.6 pm/V below the TO mode frequency of GaP due to the cancellation of ionic and electronic contributions. The minimum value of  $d_{eff}$  is 5.0 pm/V at 7.54 THz. When approaching TO mode frequency,  $d_{eff}$  is larger than 70.6 pm/V because polaritons at this area induce giant ionic nonlinearities. By comprehensive consideration of  $d_{eff}$  and  $\alpha_T$  we conclude that the lower frequencies of THz wave in the range of 0–7.54 THz, the higher intensities of THz wave generated from SPS processes.

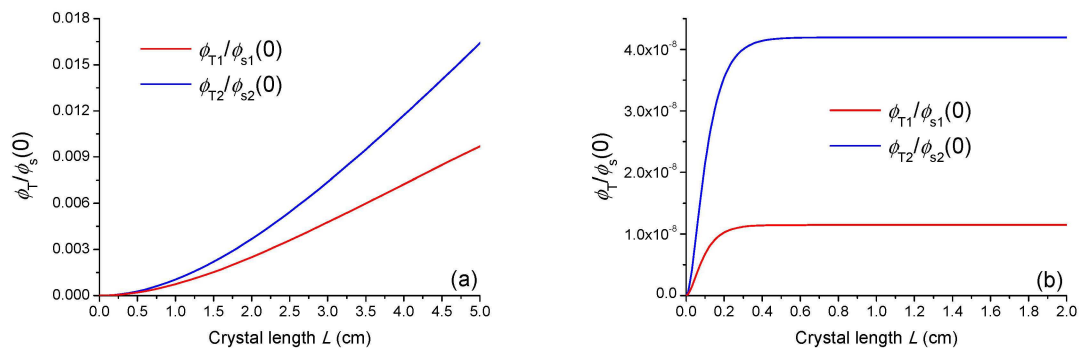


**Figure 5.** Calculated  $d_{eff}$  and  $\alpha_T$  versus THz frequencies.  $d_e$  is 70.6 pm/V with pump wavelength of 1.064  $\mu\text{m}$ .

With THz wave absorption, without phase mismatch and pump depletion, the coupled wave equations can be solved to give the THz photon flux density  $\phi_T$  with a general solution [21,22], given by

$$\phi_T = \phi_s(0)e^{-\alpha_T L/2} \frac{g_T^2}{g_T^2 + \left(\frac{\alpha_T}{4}\right)^2} \times \left| \sinh \left( \sqrt{g_T^2 + \left(\frac{\alpha_T}{4}\right)^2} L \right) \right|^2 \quad (9)$$

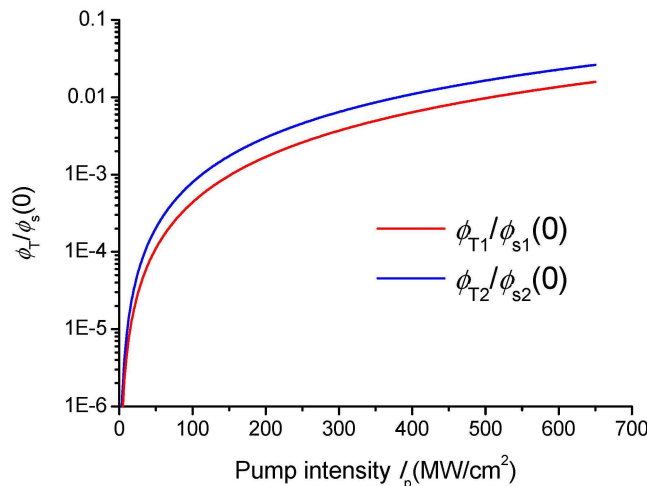
where  $L$  is the crystal length. The initial THz photon flux density  $\phi_T$  is assumed to be zero, and  $\phi_s(0)$  is the initial Stokes wave photon flux density. Figure 6 shows THz wave photon flux density  $\phi_T/\phi_s(0)$  from coupled OPOs by SPS processes.  $\phi_{T1}$  and  $\phi_{T2}$  are the photon flux densities of  $\nu_{T1}$  and  $\nu_{T2}$ , respectively,  $\phi_{s1}(0)$  and  $\phi_{s2}(0)$  are the initial photon flux densities of  $\lambda_{s1}$  and  $\lambda_{s2}$ , respectively. In the calculations pump intensity  $I_p$  is 500 MW/cm<sup>2</sup> since the damage threshold of GaP is 650 MW/cm<sup>2</sup> [23]. From Figure 6a we find that  $\phi_T/\phi_s(0)$  rapidly increases with crystal length  $L$  as  $\nu_{T1} = 1.13$  THz and  $\nu_{T2} = 0.95$  THz. As shown in Figure 5,  $d_{eff}$  is large and  $\alpha_T$  is small as THz wave frequencies are around 1 THz. The large  $d_{eff}$  and small  $\alpha_T$  enhance the rapid increase of  $\phi_T/\phi_s(0)$  with crystal length  $L$ . As the coefficient  $\alpha_T$  of 1.13 THz is larger than that of 0.95 THz,  $\phi_{T1}/\phi_{s1}(0)$  is smaller than  $\phi_{T2}/\phi_{s2}(0)$ .  $\phi_{T2}/\phi_{s2}(0)$  with value of 0.016 can be reached as crystal length  $L$  is 5 cm. From Figure 6b we find that  $\phi_T/\phi_s(0)$  rapidly increases with crystal length  $L$  and then keeps invariant as  $\nu_{T1} = 6.69$  THz and  $\nu_{T2} = 6.45$  THz. Compared with Figure 6a,  $d_{eff}$  becomes smaller and  $\alpha_T$  becomes larger, which induces an intensive decrease of  $\phi_{T1}/\phi_{s1}(0)$  and  $\phi_{T2}/\phi_{s2}(0)$  in Figure 6b. From Figure 6 we conclude that high-power THz waves can be generated in lower THz frequency band. The intensity of THz wave can be boosted by injecting an initial Stokes wave as a seed light.



**Figure 6.** THz wave photon flux density  $\phi_T/\phi_s(0)$  versus crystal length,  $I_p = 500$  MW/cm<sup>2</sup>. (a)  $\lambda_p = 1.025$   $\mu\text{m}$ ,  $\nu_{T1} = 1.13$  THz,  $\nu_{T2} = 0.95$  THz,  $\Lambda = 500,000$   $\mu\text{m}$ ; (b)  $\lambda_p = 0.852$   $\mu\text{m}$ ,  $\nu_{T1} = 6.69$  THz,  $\nu_{T2} = 6.45$  THz,  $\Lambda = 5000$   $\mu\text{m}$ .

As shown in Equation (6), low-loss parametric gain  $g_0$  is proportional to the pump intensity  $I_p$ . A pump wave with a pump intensity on the order of GW/cm<sup>2</sup> can enlarge the gain coefficient  $g_T$  by several orders of magnitude [24,25], which can enhance the intensity of THz wave. Figure 7 shows the THz wave photon flux density  $\phi_T/\phi_s(0)$  versus pump intensity  $I_p$ .  $\phi_{T1}/\phi_{s1}(0)$  and  $\phi_{T2}/\phi_{s2}(0)$  monotonously increase with the increase of pump intensity  $I_p$ .  $\phi_{T2}/\phi_{s2}(0)$ , with a value of 0.026, can be reached as pump intensity  $I_p$  is 650 MW/cm<sup>2</sup>. The intense THz wave can be generated using a pump wave with a high intensity.





**Figure 7.** THz wave photon flux density  $\phi_T/\phi_s(0)$  versus pump intensity  $I_p$ ,  $L = 5$  cm,  $\lambda_p = 1.025$   $\mu\text{m}$ ,  $\nu_{T1} = 1.13$  THz,  $\nu_{T2} = 0.95$  THz,  $\Lambda = 500,000$   $\mu\text{m}$ .

Compared with other SPS processes generating THz waves, the scheme in this work generating two pairs of Stokes and THz waves from coupled OPOs with periodically-inverted GaP by SPS processes has four advantages. First, two pairs of Stokes and THz waves are simultaneously generated only by a periodically-inverted GaP crystal and a pump laser. Second, since the two pairs of Stokes and THz waves are simultaneously generated by a same pump wave, the two pairs of Stokes and THz waves are phase-conjugate. Third, the two THz waves with any frequency below the TO mode frequency of GaP can be simultaneously generated with a suitable pump wavelength. Fourth, the intensities of the THz waves can be enhanced by injecting an initial Stokes wave as a seed light. The scheme can be applied to other crystals, such as GaAs, ZnTe, CdTe, DSTMS (4-*N,N*-dimethylamino-4'-*N'*-methylstilbazolium 2,4,6-trimethylbenzenesulfonate), and OH1 (2-[3-(4-hydroxystyryl)-5,5-dimethylcyclohex-2-enylidene]malononitrile). These crystals can generate THz wave by SPS with collinear phase-matching configuration. Moreover, these crystals can be periodically-inverted or periodically poled to satisfy QPM condition. Furthermore, THz absorption coefficients of these crystals are small.

#### 4. Conclusions

Two pairs of Stokes and THz waves can be simultaneously generated from coupled OPOs with SPS processes in periodically-inverted GaP. The two QPM conditions of the coupled OPOs are simultaneously satisfied. The two THz waves with any frequency below the TO mode frequency of GaP can be generated with a suitable pump wavelength. THz waves can be efficiently generated with high pump intensities, particularly in a lower THz frequency band.

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