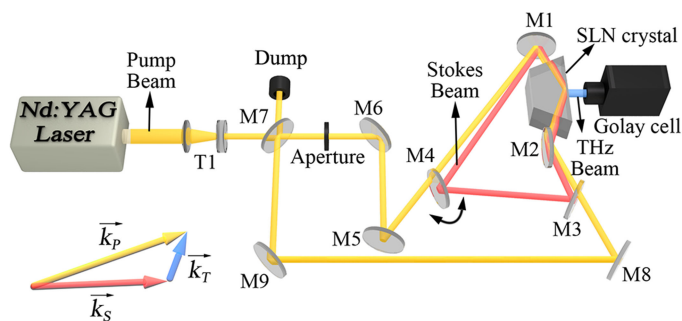


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Abstract: The energy scaling of THz wave in wide range based on surface-emitted (SE) ring-cavity terahertz parametric oscillator (TPO) with pump recycling technique is demonstrated in this paper. The continuously tunable range from 1.24 to 3.77 THz was achieved with full pump beam recycling, whereas the tunable range was 1.24–2.72 THz without pump beam recycling. The terahertz output energy and conversion efficiency increased more than seven times throughout the whole tuning range. The maximum enhancement ratio of the terahertz output energy was 12.8 times at 2.72 THz. Additionally, under the same experimental conditions, THz conversion efficiency was increased from 7.77×10^{-6} to 7.85×10^{-5} and the threshold energy was significantly reduced by 41% at 1.63 THz. Moreover, through the theoretical analysis, the enhancement mechanism of TPO by pump recycling was verified to originate from the threshold reduction of the stimulated polariton scattering (SPS) process and the interaction improvement between the pump and Stokes waves in the difference frequency generation (DFG) process.

Index Terms: Terahertz radiation, nonlinear optics, Raman scattering, optical resonators.

1. Introduction

The terahertz wave (THz wave), due to the special properties, has received much attention in several related areas of atmospheric research, materials science, biosensing, biomedical and food safety inspection. With the development of THz technologies and its application in practical research areas, the widely tunable, high-efficiency and high energy THz source is still in urgent demand.

Terahertz parametric oscillator (TPO) has been demonstrated as an efficient method to generate the widely tunable, high energy output THz wave, which is based on stimulated polariton scattering from the A1-symmetry soft mode in nonlinear crystals such as MgO-doped congruent LiNbO₃ [1], MgO-doped near-stoichiometric LiNbO₃ [2], KTiOPO₄ [3], KTiOAsO₄ [4] and RbTiOPO₄ [5] crystals. Over the past two decades, most of the studies on TPO have focused on improving the THz output characteristics and extending the tuning range through the efforts of etched grating coupler [6], Si prism arrays coupler [7], surface-emitting (SE) cavity design [8], [9] and various fundamental wavelength pumping [10], [11]. However, it should be noted that the pump pulse will be dumped after a single pass through the nonlinear crystal in the traditional terahertz parametric techniques. Thus, only a small part of pump energy (<50%) can be utilized for the THz generation and the energy conversion efficiency is low. To solve this problem, a single recycled pump beam in linear-cavity TPO has been reported to improve the terahertz output energy. Terahertz output energy was increased about four times at 1.53 THz with the decrease in threshold. However, in the configuration mentioned above, the recycled pump beam and the fresh pump beam had a small angle in the crystal, instead of being completely collinear. Thus, multiple recycling could not be realized, and in the single recycling, only about 47%–50% volume of the tilted recycled beam interacts with the fresh pump beam in the crystal [12]. Meanwhile, the Stokes wave excited by the recycled pump beam can not resonate in the Stokes cavity because of the phase-matching condition, which was not beneficial to the efficient use of the recycled pump beam. Besides, during the frequency tuning process, the path of residual pump beam would change due to the crystal rotating. This makes the pump recycling was only achieved in a fixed frequency point, instead of whole tuning range, which is not beneficial to practical application. Moreover, it is difficult to implement the fast frequency tuning by this kind of TPO with single recycled pump configuration.

In this paper, the widely tunable, high-efficiency and high energy surface-emitted (SE) ring-cavity TPO has been demonstrated based on the pump recycling technique. The continuously tunable THz frequency range of 1.24 THz to 3.77 THz was achieved with full pump beam recycling. The maximum enhancement ratio of the terahertz output energy was 12.8 times at 2.72 THz. In addition, the threshold energy was significantly reduced by 41% at 1.63 THz. Moreover, the enhancement mechanism of TPO with pump recycling technique has been analyzed.

2. Experimental Details

Fig. 1(a) shows the experimental setup of the SE ring-cavity TPO with pump recycling technique. Except for the optical recycle design, this optical design is similar to the surface-emitted ring-cavity TPO that we proposed [13]. A multimode Q-switched 1064 nm nanosecond laser was used as the pump source. The repetition rate and pulse-width were 10 Hz and 10 ns, respectively. The pump beam was shaped and collimated by the telescope lens T1 to reduce the spot size for increasing the power density. The beam diameter was reduced to 5 mm through an aperture. Then mirrors M5 and M6 having HR coating for the infrared range were used to adjust the appropriate incident angle and height of the pump beam. The ring-cavity consisted of the plane-parallel mirrors M1, M2, M3 and M4 to be used as a Stokes resonator. Among them, mirrors M1 and M2 coated HR in the range of 1063–1078 nm. Moreover, mirrors M3 and M4 were both coated with high transmission more than 98% in 1063–1064.7 nm wavelength range and high reflection in the range of 1067 nm–1078 nm ($R > 70\%$ @ 1067–1070 nm, $R > 90\%$ @ 1070–1078 nm) for the s-polarization at the incident angle of 30°. These specially coating mirrors were fabricated in the OCJ/Optical Coatings Japan. In order to rapidly tune the THz frequency, mirror M4 was fixed on a high-speed optical scanner (Cambridge Technology Inc., 6230H) with a tuning time of 600 μ s/THz frequency, whose performance is described in [13]. The pump recycling technique was achieved by mirrors M7, M8 and M9. Mirror M7 was coated with the transmission of 40% in the infrared range at the incident angle of 45°, which can not only realize the recycled residual pump beam and overlapped with the fresh beam here, but also acts as an attenuator to achieve high conversion efficiency by controlling the fresh pump energy with good beam quality [14]. The pump energy after the aperture is used reasonably for calculating the conversion efficiency, called as the fresh pump energy in the following

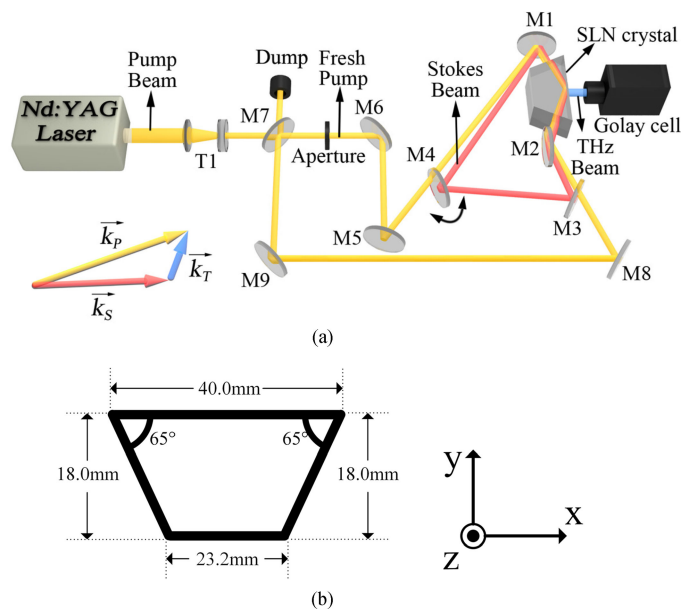


Fig. 1. (a) The experimental setup of the SE ring-cavity TPO with pump recycling technique. (b) The crystal geometry.

context. Mirrors M8 and M9 were coated HR in the infrared range. Through mirrors M7, M8 and M9, the recycled pump beam was aligned with the fresh pump beam to achieve the three-dimensional collinearity. Thus, the pump beam can be completely circulated in such cavity. The optical recycled design (M7-M6-M5-M1-M8-M9-M7) had an optical path length of about 600 mm, which means the recycled pump beam had a time delay of approximately 2 ns for the fresh pump beam. The time delay was determined by the temporal relationship among the fresh pump, residual pump and Stokes pulses, which will be discussed later. Moreover, a 1 mol.% Magnesium oxide doped near-stoichiometric lithium niobate (MgO:SLN) crystal, in which the composition ratio between Li and Nb was 49.6:50.4 (mol. %), was used as the nonlinear gain medium. Due to its larger Raman scattering cross section and smaller absorption coefficient, the SLN crystal is hopeful to realize widely tunable and high-energy THz output [2]. The MgO:SLN crystal was cut as an isosceles trapezoid configuration, and the crystal surface was optically polished without the coating, as shown in Fig. 1(b). This configuration ensures the pump and Stokes waves with total reflection at the crystal surface and makes the generated THz wave emitting approximately perpendicular from the crystal surface without any coupler. The THz wave output signal was measured by the Golay cell detector (TYDEX, GC-1P), whose typically energy conversion parameters are calibrated by the CW or CW-like THz source. Considering the THz repetition rate of 10Hz in our experiment, the Golay cell is used to roughly estimate the magnitude of the THz output energies. In order to avoid the inaccuracy of the THz energy measurement, all THz signals in the manuscript were expressed in millivolts without any conversion. A 1mm thickness black polyethylene sheet covered on the detector was used as a filter to block the injection of the scattered infrared lights. The Stokes wavelength was measured by an infrared spectrometer (Agilent, 86142B).

3. Results and Discussion

Firstly, the THz output properties of the SE ring-cavity TPO with pump recycling technique will be revealed. After that, the enhancement mechanism of pump recycling technique will be analyzed. As shown in Fig. 2, the Stokes wavelength and the generated THz frequency in the SE ring-cavity TPO with pump recycling technique were measured at a certain fresh pump energy of 120 mJ. Varying the angle of the scanner by controlling the voltage signal, the continuously tunable Stokes

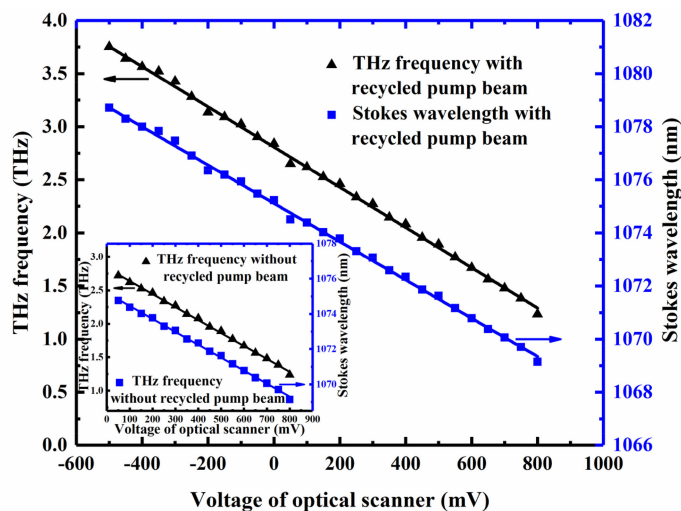


Fig. 2. Measured tuning characteristics of the Stokes wavelength and the THz frequency with pump recycling technique. Inset: the tuning characteristics of the Stokes wavelength and the THz frequency without pump recycling technique.

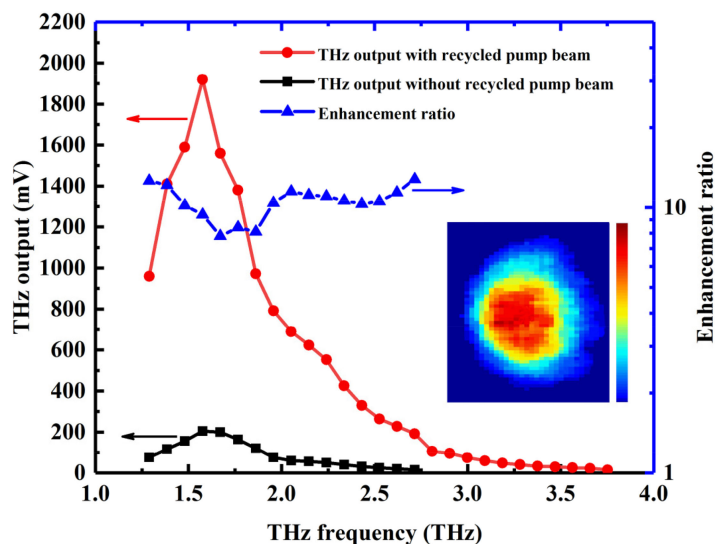


Fig. 3. The THz output amplitudes with and without pump recycling technique. Inset: the THz energy distribution of the SE ring-cavity TPO with pump recycling technique at 7 cm from output surface.

wave in the range of 1069.1 nm to 1078.7 nm was obtained, corresponding to the tunable THz frequency range of 1.24 THz to 3.77 THz. As a contrast, the tuning THz frequency range without pump recycling technique was measured under the same experiment situation. The tuning range from 1.24 THz to 2.72 THz was obtained, as shown in the inset. It is obvious that the tuning THz frequency range with pump recycling technique was up to higher frequency, which can be attributed to the threshold reduction. Moreover, the linewidth of THz output has been measured by a scanning F-P etalon at 1.63 THz, and estimated as 30 GHz by the free spectral range (FSR) of F-P etalon.

The THz output amplitudes from two kinds of TPO systems were measured to reveal the THz output performance improvement resulting from pump recycling technique, and shown in Fig. 3. Under a certain fresh pump energy of 120 mJ, the maximum THz output amplitudes with and without pump recycling technique were 1920 mV and 204 mV at 1.63 THz, respectively. The THz

output enhancement ratios of two kinds of TPO systems were calculated. Obviously, the significant enhancement of THz wave output was achieved throughout the whole tuning range. Among them, the maximum and minimum enhancement ratios were calculated to be 12.8 times at 2.72 THz and 7.8 times at 1.72 THz, respectively. Moreover, the average enhancement ratio in the whole tuning range (1.24 THz–2.72 THz) was 10.53 times. Furthermore, in order to evaluate the beam quality of the SE ring-cavity TPO with pump recycling technique, the THz energy distribution at the 7 cm far from the output surface was measured by using two-dimensional scanning with a 400 μm diameter pinhole and shown in the inset. The measured THz beam profile had a Gaussian distribution both in the vertical and horizontal directions, which is because that the pump beam from the Nd:YAG laser has a Gaussian distribution in the spatial domain. The half maximum (FWHM) beam diameters of 6.2 mm and 9.2 mm and the divergence of 3.05° and 4.57° were measured in the vertical and horizontal directions, respectively.

Here, two troughs were found on the curve of enhancement ratio in the range of 1.6 THz to 1.9 THz and 2.3 THz to 2.5 THz. It is deduced that this phenomenon is related to the resonance peaks of 1.9 THz and 2.5 THz for the A1 phonon-polariton in LiNbO₃ crystal, which could result in the dramatic increase of the refractive index and absorption coefficient around 1.9 THz and 2.5 THz [15], [16]. According to the Eqs. (5) and (6) in [17], the THz wave gain g_T can be simplified as follows:

$$g_T = \left(\frac{\alpha_T^2}{4} + A \frac{I_P}{n_T} \right)^{\frac{1}{2}} - \frac{\alpha_T}{2}, \quad (1)$$

where I_P , n_T and α_T are the pump wave intensity, refractive index and absorption coefficient in THz region, respectively. A is $2\pi\omega\omega_s\chi_p^2\cos\theta/c^3n_s n_p$, which is related to the pump frequency, THz frequency, refractive index of pump and Stokes waves, phase-matching angle, second- and third-order nonlinear processes. In our situation, A could be consider as a constant at around a certain THz frequency. The first derivative of g_T with respect of I_P can be described as

$$\frac{\partial g_T}{\partial I_P} \propto \frac{1}{\sqrt{n_T \alpha_T}}, \quad (2)$$

which will decrease with increasing of n_T and α_T . The dramatic increasing of n_T and α_T was occurred around the resonance peaks of 1.9 THz and 2.5 THz. It is known that the adverse effect of the resonance peak on the enhancement ratio is a frequency band rather than a frequency point. Thus, we deduce that the decrease of enhancement for the frequency range of 1.6 THz to 1.9 THz and 2.3 THz to 2.5 THz are attributed to the resonance peak at 1.9 THz and 2.5 THz, respectively.

Fig. 4 shows the THz output amplitudes with and without recycled pump beam under different pump energies at 1.63 THz. It is seen that the threshold energies with and without pump recycling technique were 49 mJ and 83 mJ, respectively. This indicates that the threshold energy of the SE ring-cavity TPO with pump recycling technique can be efficiently reduced by 41%. Meanwhile, the enhancement ratio tended to be stable with the increase of pump energy. When the fresh pump energy was 130 mJ/pulse, the THz output amplitude with recycled pump beam was about 10.2 times larger than that obtained without recycled pump beam. The corresponded THz conversion efficiency was increased from 7.77×10^{-6} to 7.85×10^{-5} . Intuitively, for single recycling, the residual pump energy after first round was about 60% of the fresh pump energy, and then though M7, less than 36% of the fresh pump energy entered the crystal again, which improved the total pump energy for SE ring-cavity TPO system. But the enhancement ratio of THz output was about 10.2 times larger than that obtained without recycled pump beam, which was much higher than the improvement ratio of the pump energy. As contrast, if the pump energy of 36% in SE ring-cavity TPO was increased without recycled pump beam, the THz output increases just about 3~4 times. This phenomenon can be clearly indicated in Fig. 4.

In order to explore the mechanism of THz output enhancement with pump recycling technique, the principle of TPO is explained in a different way here. As we know, the THz generation in TPO involves the second-order and third-order nonlinear processes [17]. On one hand, the Stokes and THz waves are initially generated from the stimulated polariton scattering (SPS) under the condition

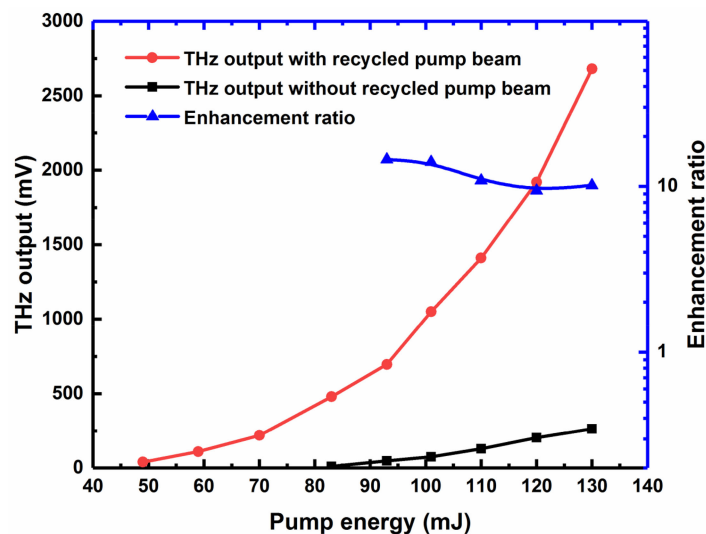


Fig. 4. The THz output amplitudes with and without pump recycling technique under different pump energies at 1.63 THz.

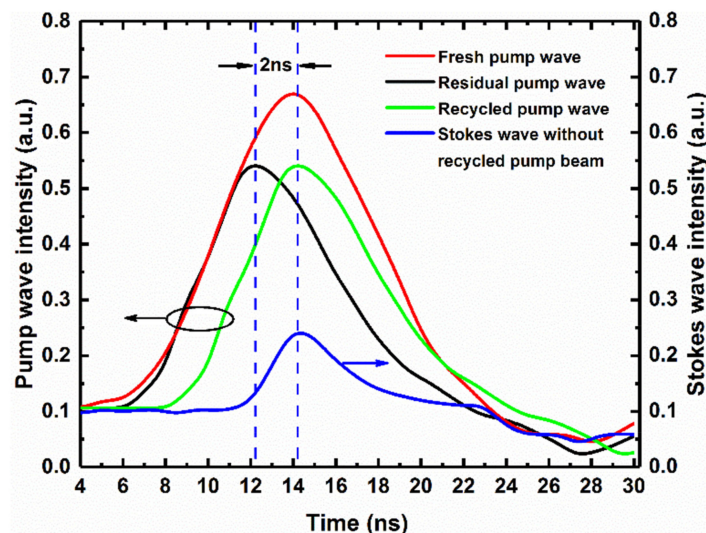


Fig. 5. The temporal relationships among the pump, residual pump, recycled residual pump and Stokes waves.

of noncollinear phase-matching, which occurs above the threshold. Meanwhile, the Stokes wave is enhanced by optical resonator, interacting with pump wave to generate the THz wave by difference frequency generation (DFG), which is a non-threshold process. Therefore, the generated THz wave is derived from two parts, namely the SPS and DFG processes, even though the two parts cannot be well distinguished in the time domain. Thus, there is no doubt that the THz output and conversion efficiency could be enhanced by reducing the threshold of SPS or increasing the interaction between the pump and Stokes waves.

Fig. 5 shows the temporal relationships among the fresh pump, residual pump (depleted pump), recycled pump (a single recycling) and Stokes waves. They were measured by a fast response InGaAs detector (Thorlabs DET08C) under the fresh pump energy of 120 mJ at 1.63 THz. It is clear that the fresh pump pulse is significantly depleted with the Stokes pulse building and there is a time delay of about 5 ns between the front edges of fresh pump pulse and Stokes pulse due to the

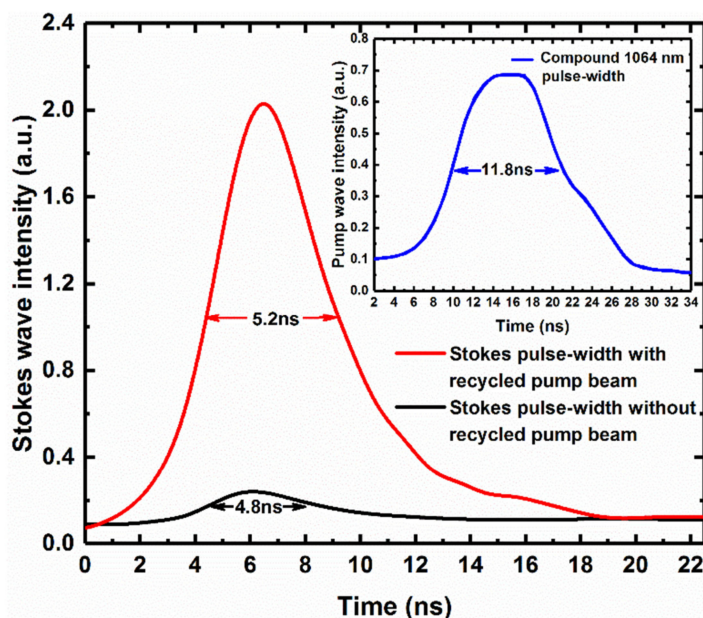


Fig. 6. The Stokes pulse-widths of the SE ring-cavity TPO with and without pump recycling technique at 1.63 THz. Inset: the compound 1064 nm pulse-width.

energy accumulation over the threshold. Based on the relative amplitudes and temporal shapes of the fresh pump and depleted pump pulses, most part of the fresh pump energy is left (around 61% in our experiment) after the single interaction in crystal, which can be reutilized in order to improve the conversion efficiency. When the residual pump beam is incident into the ring-cavity TPO, the THz wave can be generated again based on the principle of TPO with the processes of SPS and DFG. On one hand, the residual pump beam can overlap with the fresh pump beam, the total pump intensity will be improved, which is beneficial for the THz gain according to Eq. (1) and can reduce the pump threshold of SPS. On the other hand, because the Stokes wave has been oscillated in the cavity, we infer that the DFG process between the recycled pump pulse and Stokes pulse plays an important role in the enhancement of THz generation. Considering the conversion efficiency of DFG increases monotonously with the product of the pump wave intensity and the Stokes wave intensity, the relative position and the degree of overlap between the recycled pump pulse and Stokes pulse are key factors. Thus, the recycled optical path length should be optimized in order to fully utilize the recycled pump pulse. During the experiment, the recycled optical path length was firstly designed to be 600 mm, corresponding to the 2 ns time delay between the fresh pump and the recycled pump. This brings the peaks of the recycled pump and Stokes pulses almost overlapped in time domain, which could maximize the conversion process of the second-order DFG. From the above discussion, the enhancement mechanism of TPO with pump recycling technique was verified to originate from the threshold decreasing of the SPS process and the interaction increasing between the pump wave and Stokes wave in the DFG process.

Fig. 6 shows the full-width half maximum (FWHM) pulse-widths of Stokes wave with and without pump recycling technique. The Stokes pulse-widths with and without pump recycling technique were 5.2 ns and 4.8 ns under the fresh pump energy of 120 mJ/pulse at 1.63 THz. Hence, it can be indicated that the pulse width of the generated THz wave is still as narrow as several nanoseconds despite the Stokes pulse-width being slightly broaden by 0.4 ns. When the THz energy was significantly enhanced by pump recycling technique, the peak power of THz wave was increased as the same order accordingly. Besides, the enhancement ratio of the Stokes wave intensity with and without recycled pump beam was about 8.79 times. It is almost the same with the enhancement ratio of THz output at 1.63 THz, which coincides with the reports in [12]. Additionally,

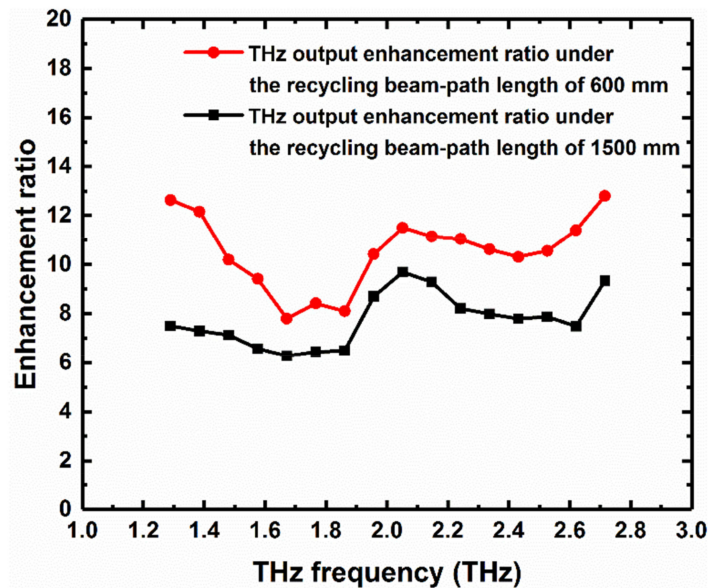


Fig. 7. The THz output enhancement ratios under the recycled optical path length of 600 mm and 1500 mm, respectively.

it is found that the Stokes pulse with recycled pump beam was established earlier than that without recycled pump beam. It verifies that the threshold of TPO can be efficiently reduced with the pump recycling, as the discussed above. Furthermore, a mirror with 10% reflectance in the infrared range was inserted between the M8 and M9 at the angle of 45°, the pulse-width of the 1064 nm laser inside the SE ring-cavity was measured as pump beam was completely circulated, which is called as the compound 1064 nm pulse here. As shown in the inset of Fig. 6, the pulse-width of the compound 1064 nm laser inside the cavity was 11.8 ns, which was broadened by 1.8 ns compared with the original incident 1064 nm pulse. This verifies the overlap between the fresh pump beam and recycled pump beam. Moreover, the temporal waveform was similar to the flattened Gaussian beam, which can guarantee the compound 1064 nm pulse has good temporal overlap and longer interaction duration with the Stokes pulse. In other words, the peaks of the compound 1064 nm pulse and Stokes pulse can fully interact with each other, and the THz output energy can be enhanced.

Furthermore, the enhancement ratios of THz output under different recycled optical path lengths were investigated with the same fresh pump energy and tunable range and shown in Fig. 7. To make a compelling comparison, the longer time delay of 5 ns between the fresh pump and recycled pump beams was chosen, which can be caused by the recycled optical path length of 1500 mm. It is seen that the variation tendencies of enhancement ratio under different recycled optical path lengths were similar during the whole frequency range. When the optical path length was adjusted to 1500 mm, the maximum and the minimum enhancement ratios were measured to be 9.69 times at 2.72 THz and 6.27 times at 1.72 THz, respectively, which are much smaller than those under the recycled optical path length of 600 mm. In addition, the average enhancement ratio was 7.75 times. It was reduced by 35.9% compared with that under the recycled optical path length of 600 mm. Consequently, the substantial enhancement of the THz output energy can be achieved through pump recycling technique.

4. Conclusion

In conclusion, the dramatic enhancement of THz wave output energy based on SE ring-cavity TPO with pump recycling technique has been demonstrated. The THz frequency has been achieved from 1.24 THz to 3.77 THz with full recycling of the pump beam. In the entire tuning range, more than

seven times improvement of the terahertz output energy and conversion efficiency were achieved. Additionally, THz conversion efficiency was increased about ten times and the threshold energy was significantly reduced. The enhancement mechanism was verified to originate from the threshold decreasing of the SPS process and the interaction increasing between the pump wave and Stokes wave in the DFG process. It is expected that such high-efficient TPO systems with wide tunability can promote its practical applications.

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