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Recent Progress on Compact and Portable Terahertz Source Based on Parametric Conversion from Dual-Frequency Solid-State Laser Pulses

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Abstract

We review the progress made on power scaling and compact and portable THz sources. By reversely stacking GaP plates, we improved photon conversion efficiency from 25% to 40%, which is the maximum value. As the number of the plates was increased from 4 to 5, the output power was decreased due to back conversion. We also investigated THz generation by mixing two frequencies generated by a single Nd:YLF solid-state laser. The average output power reached 1 μ W. By introducing two Nd:YLF crystals, we significantly improved the output power to 4.5 μ W. Such a configuration allowed us to generate different output frequencies. We have also reviewed our effort of making the THz source further compact by exploiting passively Q-switched laser pulses.

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I. Introduction

The terahertz (THz) region of the electromagnetic spectrum, having its frequencies being defined between 0.1 THz and 10 THz, many several potential applications. First, the transition frequencies between rotational states in gases are typically in this region [1]. Therefore, one can develop a rather compact spectrometer to fingerprint molecules. Since the linewidth for the rotational transitions can be 50 MHz (0.0017 cm⁻¹) or narrower for low-pressure gases, such a spectrometer must have a sufficiently high resolution to resolve each rotational transition. In comparison, in the domains from visible to mid-infrared, most molecules exhibit the congested and unresolved ro-vibrational spectra [2]. Therefore, it may not be feasible for anyone to truly fingerprint molecules in these domains. Second, identification and detection of biological agents based on THz spectroscopy have become increasingly important [3]. Third, due to the presence of the atmospheric windows, THz waves could be used for probing as well as imaging [4]. Based on our discussion made above, it is important for us to develop an active THz system that is capable of taking the multi-spectral images of targets and simultaneously analyzing the targets based on the THz spectra. One of the major components of the system is a THz source. A tunable and monochromatic THz source has its advantages over a broadband THz source, especially for revolving weak and congested absorption features.

Broadband THz pulses were generated based on optical rectifications [5], photoconduction [6], and Cherenkov radiation [7]. These schemes all require ultrafast laser pulses. In order to sense chemicals, time-domain spectroscopy was developed [8]. These broadband pulses were used to measure rotational spectra of gases [9], to identify explosives [10], and to take THz images [11].

Besides optical rectifications, other parametric processes in nonlinear crystals can be quite efficient for converting optical pulses to the THz waves. For example, based on THz parametric oscillation in a LiNbO₃ crystal [12], the output power on the level of ~ mW was generated in the range of 140-290 μ m (1.03-2.14 THz). By mixing two coherent infrared laser beams in nonlinear-optical crystals such as GaSe [13-15], ZnGeP₂ [16-18], and GaP [19,20], we efficiently generated the THz waves with extremely-wide tuning ranges and very high peak powers as well as high conversion efficiencies. The widest tuning range achieved by us so far covers the wavelengths anywhere from 66.5 μ m to 5.66 mm (from 4.51 THz down to 53 GHz) based on the type-*oe-e* phase-matched difference frequency generation (DFG). On the high-frequency side of

the reststrahlen band, we were able to generate the radiation tunable in the range of $2.7-28.7 \,\mu\text{m}$ using a different phase-matching configuration [21]. One can see that the tuning range achieved by us almost covers the entire THz region (0.1-10 THz) and part of the millimeter-wave or microwave domain, i.e. 1 mm to 5.66 mm (53-300 GHz). The highest output peak power measured by us is 389 W at 203 μ m (1.48 THz) and 4.7 W at the wavelength of 1 mm (300 GHz), corresponding to the conversion efficiencies of 0.098% and 0.0012% in terms of the pulse energies, respectively. The highest photon conversion efficiency was about 19% [15].

We have explored some important applications by using our widely-tunable THz sources for. Indeed, by measuring the absorption spectra of the commonly-used chemicals in the vapor phase, we demonstrated that THz absorption spectrometer developed by us could be used to fingerprint a class of molecular species [22,23]. We also measured the transmission spectra of the isotopic variants [23,24], biological species [14,25], and VX nerve agent [26].

Recently, by reversely stacking GaP plates, we were able to improve the photon conversion efficiency from 25% [27] to 40% [28]. We demonstrated that 40% corresponds to the maximum conversion efficiency [28]. As the number of the stacked GaP plates was increased from 4 to 5, the output power was decreased due to back conversion. In order to make our THz source truly compact and portable, we investigated THz generation by mixing two frequencies generated by a single Nd:YLF solid-state laser [29]. The average output power reached 1 μ W at 1.64 THz [29]. Such a THz source can have a dimension of $12'' \times 12'' \times 6''$. By introducing two Nd:YLF crystals in the laser cavity, we significantly improved the output power to 4.5 μ W [30]. Such a configuration allowed us to generate different THz frequencies by combining two different laser crystals [30]. Through our further optimizations and utilizing novel configurations, we will reduce the dimension of the THz source down to first $12'' \times 6'' \times 4''$ and eventually to the size of a laser pointer by exploiting passively Q-switched laser pulses [31].

In this article, we give a review over our effort on the power scaling of the THz waves generated by mixing two near-infrared coherent radiation beams in stacked GaP plates. We also summarize our recent results on the development of compact and portable THz sources by mixing a pair of frequencies generated by dual-frequency actively and passively Q-switched solid-state lasers.

II. Power scaling by stacking GaP plates

In this section, we summarize our results on power scaling of THz radiation based on DFG in high-resistivity and low-dislocation-density GaP plates [27]. We stacked GaP plates such that three nonzero elements of second-order nonlinear susceptibility have the same sign or opposite signs from one plate to the next. These two stacking configurations yielded completely different results in terms of the highest output peaks and corresponding wavelengths. The highest output peak power measured by us is about 2.36 kW, which is two orders of magnitude higher than our previous result [19,20].

Each of the semi-insulating undoped (110) GaP plate used in our experiment has a resistivity of > 1.0 MΩ-cm and a dislocation density of 2.25×10^5 cm⁻². It has a diameter of 48.5 mm and a thickness of 663 µm. Both of the (110) facets are polished. In order to generate monochromatic THz pulses, two coherent radiation beams were a Nd:YAG laser at 1.064 µm with pulse duration of 10 ns and pulse energy of 20.7 mJ (the highest used in our experiment) and the idler output from a β -BaB₂O₄-based optical parametric oscillator (OPO) pumped by the third-harmonic output of the Nd:YAG laser. The highest pulse energy for the idler beam is 18.7 mJ whereas its pulse duration is 5 ns. These two beams were collimated, combined, and then illuminated the GaP plates. The beam radius for both of the mixing beams at the GaP plates was measured to be 1.0 mm. Therefore, the highest peak intensity for the idler beam was 120 MW/cm², which is a factor of 5.4 below the highest intensity of 650 MW/cm² used without causing any damage [32]. They collinearly propagated in a direction perpendicular to the two large facets of the GaP plates with their polarizations parallel to the [1,-1,0] and [0,0,1] directions, respectively. Under such a configuration, the effective second-order nonlinear coefficient is much higher than that from a (100) GaP crystal [19,20]. Germanium and polyethylene windows were used to block the two mixing beams. The THz radiation generated by DFG was attenuated, collimated by a parabolic mirror, and then focused onto a power meter by using another parabolic mirror. The THz output wavelengths were calibrated by an etalon.

By measuring the transmission and reflection spectra of each GaP plate using Fourier transform infrared spectroscopy, we deduced the absorption spectrum. According to Fig. 1, the absorption coefficient is 2.2 cm^{-1} at $120 \mu \text{m}$, which is 7 times lower than that from our previous sample. This implies that we could stack up to seven plates to effectively increase the output power. Based on the dispersion relation available [33], the coherence length for the THz generation is linearly increased with the wavelength from 581 μm at 76.5 μm . At 304 μm , e.g.

the coherence length becomes 7.37 mm. We measured the THz output powers at different output wavelengths. According to Fig. 2, the THz output radiation was tuned continuously from 3.91 THz (76.7 μ m) to 454.9 GHz (659.5 μ m). Such a tuning range was achieved by slightly adjusting the frequency (wavelength) of the idler beam generated by OPO. Within such a range, the plate thickness is either comparable to or much shorter than the coherence length calculated by us. Therefore, the THz generation is phase-matched. At 2.775 THz (108.1 μ m), the output power reached the highest value, i.e. 722 W. Using the pulse width of ~5 ns, the highest average power of the generated THz radiation is 36.1 μ W at the average input power of 207 mW at 1.0642 μ m and 187 mW at 1.0748 μ m, respectively. Such an output power is a factor of 46 larger than the highest peak power achieved on GaP so far [19,20].

By using a (110) GaP wafer, we can access a much higher value of the effective secondorder nonlinear coefficient [20]. Second, the two parallel facets of the GaP crystal can induce Fabry-Perot effect, which can also contribute to the enhancement of the THz output power. In fact, from Fig. 2 one may notice that the THz output power changes periodically. On the lowfrequency side, i.e. for the frequencies below 2.5 THz, the frequencies at most of the peaks and valleys can match those calculated based on Fabry-Perot effect. If we only assume such an effect for the THz wave, the ratio of a local maximum power and its adjacent minimum value should be about 3.3. For the frequencies higher than 2.5 THz, the frequencies at the peaks and valleys are no longer consistent with those predicted by Fabry-Perot effect. In addition, the modulation depths are much larger. We believe these modulations are primarily caused by the water-vapor absorption. Third, the absorption of the THz pulses generated by DFG is much less than that for the previous crystal [19,20]. As a result, the frequency at the highest output power from our crystal (2.775 THz) is much higher than that measured previously, i.e. 1.73 THz [19,20]. There was the subsequent absorption of the THz pulses by a rather thick crystal at relatively high frequencies [19,20]. Such an assumption is further supported by the fact that the high-frequency limit measured on our crystal is significantly higher than the previous value. Fourth, since the dislocation density of our GaP crystal is remarkably low, the damage threshold is significantly increased. As a result, we were able to use much higher peak powers for the two mixing beams.

We measured the dependence of the THz output power on the input power for one of the mixing beams. Based on the linear-least-square fit to the power dependences, we have determined the internal conversion efficiency to be 0.067% (the photon conversion efficiency of

6.8%). Without taking into consideration absorption losses, considering the Fabry-Perot effect for the THz wave, and assuming that the three nonzero elements of the second-order nonlinear optical susceptibility for GaP are identical, the maximum conversion efficiency for the THz generation in terms of peak powers is given by [34]:

$$\eta_{\max} = \frac{P_{THz} T_{THz}^{-1}}{P_1 T_1} = \frac{16d_{eff}^2 L^2}{\varepsilon_0 c n_1 n_2 n_{THz} \lambda_{THz}^2} \frac{P_2}{\pi w_0^2} \frac{T_2}{T_{THz}^2}$$
(1)

where max is used to designate the local maximum values due to Fabry-Perot effect, d_{eff} is the effective nonlinear coefficient, P_{THz} is the THz output peak power, P_1 and P_2 are the input peak powers at 1.064 µm and at the idler wavelength, respectively, λ_{THz} is the output wavelength, *L* is the total thickness of the GaP wafer, w_0 is the beam radius for the pump beams, n_i are the refractive indices of the GaP crystal at the respective wavelengths of the three interacting waves, and T_i is the Fresnel transmission coefficients at single surface. Assuming $d_{\text{eff}} \approx 47$ pm/V, we have obtained $\eta_{\text{max}} \approx 0.015\%$ from Eq. (1). This value is about a factor of 4.5 lower than our experimental value. We believe the difference between two may be caused by the uncertainty for the value of d_{eff} used in Eq. (1).

In addition to one GaP plate, we have attempted to stack two and three GaP plates. We have stacked them according to two different configurations, i.e. three elements of second-order nonlinear susceptibility for GaP have (A) same sign and (B) opposite signs between the adjacent GaP plates. Fig. 3 illustrates our result obtained for the stacking configuration A. One can see that the wavelength corresponding to the highest output power has been significantly red-shifted from 108.1 μ m to 204.8 μ m to 303.9 μ m. The corresponding power is reduced from 722 W to 698 W to 611 W. 611 W corresponds to a photon conversion efficiency of 16% inside the stacked plates. These results indicate that there is an optimal interaction length for each specific output wavelength. We believe that the red-shift of the optimal output wavelength is primarily caused by the combination of the linear wavelength dependence of the coherence length and wavelength-dependent output power reflected by Eq. (1). Therefore, at the wavelengths of 108.1 μ m, 204.8 μ m, and 303.9 μ m, the coherence lengths should be roughly close to 663 μ m, 1.326 mm, and 1.989 mm, respectively.

Under the stacking configuration B our result is completely different. One can see from Fig. 4 that the wavelength corresponding to the highest output power is kept at 120.3 μ m for the two and three plates. Therefore, quasi-phase matching is achieved at this wavelength. This implies

that the coherence length at 120.3 μ m is about 663 μ m. On the other hand, using the dispersion relation [33], we have calculated coherence length to be 1.65 mm. We believe that the factor of 2.5 between the two is probably caused by the inaccuracy in the THz index of refraction calculated by using the dispersion relation [33]. Using 663 μ m as the coherence length, we have determined the index of refraction at 120.3 μ m to be 3.217. This value is just 3.8% lower than that calculated by using Ref. [33]. Such an estimate illustrates the fact that the coherence length is quite sensitive to the index of refraction at the output wavelength. At such a wavelength, the output peak power was significantly increased from 433 W to 1.36 kW and 2.36 kW, corresponding to two and three plates, respectively. The peak power of 2.36 kW corresponds to an average power of 118 μ W. Compared with our previous result [19,20], we increased the peak power by a factor of 151, i.e. two orders of magnitude. The internal conversion efficiency deduced from our experiment is 0.22%, corresponding to the photon conversion efficiency of 25%.

III. Saturation of conversion efficiency and back conversion

In this section, we summarize our results following our previous investigation of power scaling of the THz generation based on DFG by stacking GaP plates [28]. In particular, we achieved the maximum photon conversion efficiency of 40%. The THz peak power generated by us approaches 4 kW. As the number of the GaP plates is increased from four to five, we observed the evidence on back conversion.

Similar to Section II above, we mixed two coherent input beams in stacked GaP plates. One beam has the highest input peak power of 1.5 MW at 1.064 μ m (10 ns; 10 Hz). The second one generates an output wavelength tunable in 1.079 - 1.066 μ m (5 ns; 10 Hz) with the highest peak power of 4.1 MW. The two collimated beams were directed to the GaP plates without focusing (the beam radii of 1 mm). The total intensity of the two beams at the GaP plates was 178 MW/cm². Each GaP plate had a diameter of 48.5 mm and thickness of 663 μ m. They were cut and polished along the (110) plane and stacked between a pair of aluminum plates. The two input beams propagated along the [110] direction with their polarizations being parallel to [1-10] and [001], respectively. The output THz radiation was attenuated by filters and was then measured using a power meter.

In order to achieve quasi-phase-matching (QPM) for the THz generation, we rotated every the other plate by 180°. Such a configuration is named as alternatively-rotated stacked GaP plates (i.e. reversed plates). In such a case, the second-order nonlinear coefficient changes its sign between any adjacent ones. In order to find out the enhancement factor caused by QPM, we have compared with the second stacking configuration in which all the GaP plates are perfectly aligned such that their respective crystalline axes are along in the same directions (i.e. aligned plates). Therefore, in such a case the second-order nonlinear coefficients of all the plates have the same sign.

We measured the spectrum of the THz output generated by four reversed plates (the total thickness of 2.65 mm). As illustrated by Fig. 5, the generated THz radiation is continuously tuned from 78 μ m to 300 μ m. The highest output peak power was measured to be 2.73 kW (the corresponding power of 3.77 kW generated inside the plates) under the input powers of 1.5 MW and 4.1 MW at 1.064 μ m and 1.074 μ m, respectively. At such powers, the output wavelength was 120.3 μ m. It was chosen as a compromise between the THz absorption dictating the effective interaction length and the dependence of the optimal power on the THz wavelength, see Eq. (3) below. The corresponding power conversion efficiency, defined as P_T/P_1 where P_T and P_1 are the THz peak power and input peak power at 1.064 μ m, is determined to be 0.182%. Taking into the consideration the Fresnel reflections for the input beam at the entrance facet and THz output radiation at the exit facet, we have obtained the internal photon conversion efficiency of 39.6%. The optimal THz output wavelength of 120.3 μ m corresponds to the 1st order QPM condition in the GaP plates having a period of 1.326 mm for the spatially modulated second-order nonlinear coefficients. The modulations appearing in Fig. 5 including those in 120-160 μ m are caused by the water-vapor absorption and Fabry-Pèrot effect in the GaP parallel plate.

We measured the spectrum of the THz output radiation generated by four aligned plates. As one can see from Fig. 6 the generated THz radiation covers the range of 135-500 μ m. Below 135 μ m, the output powers were quite low, due to nearly zero values for the sine function, see Eq. (2) below. The highest output power reaches 510 W at the wavelength of 236 μ m under the input power of 4.1 MW at 1.069 μ m. The power conversion efficiency is determined to be 0.034%. The corresponding internal photon conversion efficiency is 14.6%, which is significantly lower than that for the reversed GaP plates. Around such a value, the optimal THz wavelength is primarily determined by the combination of the absorption and inverse-wavelength dependence

of the THz power, see Eq. (1) below. Even though the aligned plates can be used to achieve phase-matching at sufficiently long THz wavelengths, the inverse dependence and increased optimal power, see Eq. (3) below, make the reversed plates significantly outperform the aligned plates.

Fig. 7 shows the dependence of the internal photon conversion efficiency on the average input power. For the four reversed plates, the conversion efficiency increases linearly below the average power of 50 mW for the input beam at 1.074 µm. However, as the average power is increased further, the saturation of the conversion efficiency becomes obvious. When the input power approaches 200 mW, the conversion efficiency reaches a maximum value. The highest conversion efficiency achieved in our experiment is 39.6% at the input power of 204 mW. Considering the repetition rate of 10 Hz, pulse width 5 ns and beam size of 1 mm, this high conversion efficiency was achieved at pump intensity of 130 MW/cm² at 1.074 µm. Assuming $P_2 >> P_1$, where P_2 is the peak power at 1.074 µm, the photon conversion efficiency for the THz generation inside the GaP plates can be obtained as follows [35]:

$$\eta_p = \eta_{\max} \sin^2 \left[(\pi/2) \sqrt{P_{ave} / P_{opt}} \right]$$
⁽²⁾

where $\eta_{\text{max}} = a_{\text{T}}/a_1$ is the maximum conversion efficiency with a_{T} and a_1 being the areas of the THz radiation and input beam at 1.064 µm, P_{ave} is the average input power at 1.074 µm. In Eq. (1), P_{opt} is an optimal power:

$$P_{opt} = \frac{c\varepsilon_0 n_1 n_2 n_T \lambda_1 \lambda_T a_2 \tau_2 R}{16d_{eff}^2 L^2 T_2}$$
(3)

where d_{eff} is the effective second-order nonlinear coefficient, a_2 , τ_2 , and T_2 are the beam area, pulse duration, and transmittance at 1.074 µm, respectively, and *R* is the repetition rate. Following the nonlinear least-square-fit to the data points in Fig. 7 by using Eq. (2) above, we have obtained $\eta_{\text{max}} \approx 40.0\%$ and $P_{\text{opt}} \approx 209$ mW. Therefore, we have indeed achieved the maximum photon conversion efficiency in our experiment. To the best of our knowledge, this is the first report on the complete saturation of the conversion efficiency for the THz pulses generated by DFG. Based on $\eta_{\text{max}} \approx 40.0\%$, we have obtained $a_{\text{T}} \approx 0.400 a_1$. Let us assume that the two input beams have Gaussian spatial distributions having exactly the same beam sizes, i.e. $a_1 = a_2$. After neglecting the effect caused by the saturation of the conversion efficiency, we have obtained $a_{\text{T}} \approx 0.5 a_1$. The deviation of our result from the theory is attributed to the fact that the smaller effective area for the input beam at 1.074 µm. In addition, the weak reflections between the adjacent GaP plates could contribute to the deviation. In comparison, for the aligned plates no obvious saturation was observed, see Fig. 7. The highest photon conversion efficiency is 14.6%. If this value is significantly increased, we will need to introduce a more complicated expression by taking into the consideration the phase-mismatch factor [35] to fit the saturation behavior.

With increasing the number of the GaP plates from four to five, the THz output peak power is significantly decreased from 2.73 kW for the four plates to 2 kW for the five plates, see Fig. 8. This is due to back parametric conversion. Since the optimal power is inversely proportional to the square of the length, i.e. L^{-2} , see Eq. (3), we have used Eq. (2) to achieve a good fit of the data points in Fig. 8. Using thicker GaP plates we could increase the QPM wavelength such that a larger number of the plates may be stacked to reach the zero output power beyond the optimal point shown in Fig. 8 and predicted by Eq. (2).

IV. Compact and portable THz source

In this section, we summary our previous result following our demonstration of a compact and portable THz source which has been implemented by us based on DFG in a GaSe crystal [29]. An average THz output power can be as high as 1 μ W at 1.64 THz (182 μ m) in a linewidth of 65 GHz, corresponding to the peak power of 20 mW.

In our experiment, we have chosen a Nd:YLF crystal as the lasing medium since two different frequencies of 286.5 THz and 284.9 THz (wavelengths of 1047 nm and 1053 nm) can be generated from a single Nd:YLF laser. Indeed, since a Nd:YLF crystal is birefringent, it emits two transitions at 1047 nm and 1053 nm with the polarizations being orthogonal to each other, designated by π and σ , respectively. Since the two transitions have the stimulated emission cross sections of 1.8×10^{-19} cm⁻² and 1.2×10^{-19} cm⁻² at 1047 nm and 1053 nm, respectively, it is feasible for us to tailor the output powers at these two wavelengths from a single laser crystal to be close to each other. In such a case, the corresponding THz power reaches an optimal value under the same sum of the two input powers. Moreover, the THz output frequency generated by mixing these two laser beams is 1.643 THz (i.e. the wavelength of 182.42 µm), which is sufficiently close to 200 µm, and therefore, the THz output power is close to the maximum value from a GaSe crystal [13-15]. Furthermore, a Nd:YLF crystal is known for its low thermal effect and long upper-state lifetime. In the past, a single Nd:YLF crystal was used to simultaneously

generate the output beams at 1047 nm and 1053 nm without introducing any optical element inside a single laser resonator [36]. However, the ratio of the powers generated at these two wavelengths could not be controlled at all. In addition, the laser output powers are also too low for the efficient generation of a THz output based on DFG. Although a Q-switched Nd:YLF laser was implemented at 1053 nm [37], the simultaneous lasing of the two wavelengths has not been achieved. Besides the Nd:YLF crystal, other combinations of the active ions and host materials can emit two different frequencies, see e.g. Ref. [38]. As demonstrated by us below, Q-switched solid-state lasers may be one of the optimal choices for implementing a compact THz source based on DFG.

Our experimental setup is shown in Fig. 9. The section bordered by a close-loop dashed line is a schematic for a dual-frequency Nd:YLF laser. In principle, one can simultaneously generate the coherent beams at 1047 nm and 1053 nm from a single Nd:YLF laser. However, under such a case balancing the output powers at the two wavelengths is out of question due to the strong competition between the gains for the two transitions within the same laser medium. In our experiment, we introduced an intracavity polarizer to separate the two perpendicularlypolarized beams into two laser cavities both of which shared the same lasing medium for alleviating the gain competition. In our design, input mirror (IM), output couplers (OC1), and polarizer (P) make up the cavity for lasing at 1047 nm (π , o-wave), whereas IM, P, and couplers OC2 make up the cavity for lasing at 1053 nm (o, e-wave). An a-cut 1.0%-Nd-doped YLF $(4 \times 4 \times 10 \text{ mm}^3)$ crystal was shared arm by the two laser cavities. A plano input mirror (IM) has a coating for producing the high transmittance of T > 95% at 808 nm and simultaneously a high reflectivities of R > 99.5% at 1047 nm and 1053 nm. The polarizer (P) has a coating in such a way that when being placed at a Brewster's angle relative to the input beam, the π -polarized beam has a high transmittance of T \approx 98% whereas the σ -polarized beam has the high reflectivity of R > 99.9. The Nd:YLF crystal was pumped by a diode laser via a fiber pigtail after the laser beam was collimated by a lens assembly. Such a diode laser has the dimension of 1.75"×1.25", which is portable. Considering the fact that the stimulated emission cross section for the 1047nm transition is 50% larger, one of the most effective approaches for balancing the output powers of the two transitions is to introduce an additional amount of the loss in the cavity for the 1047-nm beam. First, we have specifically chosen the different reflectivities for the two output coupler to be 75% and 80% at 1047 nm (OC1) and 1053 nm, respectively. As a result, the lasing

thresholds for the two transitions become closer to each other. Second, the output coupler (OC1) was slightly tilted to introduce an additional amount of the loss at 1047 nm. In order to generate sufficiently high peak powers, an acoustic-optic Q-switch was placed next to the Nd:YLF crystal in such a way that it was shared by the two beams oscillating in the two cavities. This represents an important method for us to synchronize the output pulses generated from the two laser cavities. The typical dimension of the entire THz source illustrated by Fig. 9 is about $12'' \times 12'' \times 6''$. Through further optimization, it can be reduced to $12'' \times 6'' \times 4''$. In addition, such a compact THz source currently consumes the electrical power of ≤ 20 W. Therefore, it can be readily packaged into a portable system for field applications.

Fig. 10 shows the dependence of Q-switched output powers at 1047 nm and 1053 nm and the sum of the output powers on the output powers from the laser diode. The lasing thresholds were measured to be 1.17 W and 1.64 W at 1047 nm and 1053 nm, respectively. From Fig. 10, we can see that below the pump power of 5 W, both of the output powers increased linearly with the pump power. In addition, the powers at 1047 nm are significantly higher. Above 5 W, however, the output power at 1047 nm became slightly saturated whereas the output power at 1047 nm was increased at a higher rate. This is due to the fact that two transitions accessed the same population of the electrons in the upper lasing level. When the gain for one transition was saturated, the gain for the second one was increased. Therefore, the dependence for the sum of the output powers is still linear, which agrees well with the theory for the solid-state laser [39]. By measuring the output powers as a function the repetition rate, we found an optimal repetition rate of 3.9 kHz for the Q-switch in terms of the THz output powers. At the pump power of 9.69 W, we generated the output powers of 1.196 W and 0.608 W at 1047 nm and 1053 nm, respectively, corresponding to the net conversion efficiency of 18.6%. The ratio of the output powers at the two wavelengths is measured to be around 2:1. By further optimizing the design of the optical cavities, we can reduce the ratio to nearly 1:1. M² factors were measured to be 5.18 and 3.07 for 1047 nm and 1053 nm, respectively, at the pump power of 5.9 W. The poorer beam quality at 1047 nm is attributed to the tilting of its cavity mirror, which was used by us to reduce the ratio of the output powers for the two transitions. The pulse shapes of the two output beams are shown in Fig. 11. The two pulse trains were measured simultaneously by using two photodiodes. According to Fig. 11, the two pulses were synchronized. At the pump power of 9.69 W, the pulse widths were measured to be 15.47 ns and 18.59 ns at 1047 nm and 1053 nm, respectively.

Using two convex lenses with the same focal length, the two beams were collimated and then combined by a polarization cube, as shown in Fig. 9. It was adjusted in such a way that the optical paths at the two wavelengths were exactly the same. As a result, the two input beams were overlapped in both time and space and focused on the 15-cm-long GaSe crystal by using a convex lens (L3). After DFG, the residual pump beams were blocked by using a white polyethylene filter and the reflected pump beams were collected by a beam dump. The generated THz beam was collected by two off-axis parabolic mirrors and then focused onto a power meter. The input beams at 1047 nm and 1053 nm were ordinary and extraordinary waves inside the crystal whereas the THz wave was an extraordinary wave. The external phase matching angle was measured to be 11.5°, which is close to the theoretical value of 11° [34]. The azimuthal angle was optimized in our experiment in order to reach the optimal value of the effective nonlinear coefficient [34]. The output wavelength was measured by scanning a Si-based etalon, shown in inset to Fig. 12. The output wavelength was measured to be 183.3 µm, which agrees well with 182.4 µm, calculated from the two input wavelengths of 1.04666 µm and 1.0527 µm [39]. We plotted the dependence of the average and peak output powers on the sum of the output powers generated by the Nd:YLF laser as the input powers for DFG, see Fig. 12. The solid curve corresponds to the quadratic fit to the data points, which is consistent with the characteristics of DFG. Our experimental result indicated that the average output reached optimal values at the repetition rate of 3.9 kHz. When the sum of the input powers was 1.8 W, the average output power from the GaSe crystal was measured to be 0.948 µW. The pulse width for the THz beam was deduced to be 12.36 ns by measuring the temporal profile of the sum-frequency signal in a KTP crystal. Based on such a pulse width, the highest THz peak power was determined to be 19.7 mW. The linewidth of the THz wave was estimated to be 65 GHz by measuring the linewidth of the sum-frequency signal (i.e. 0.06 nm). A DLATGS pyroelectric detector operating at room temperature can have a noise equivalent power of as low as 230 pW. Therefore, the output power of 0.948 µW corresponds to a dynamic range of 4120, which is sufficiently high for realizing the key applications mentioned above. We can further improve the output powers to at least 100 µW by placing a nonlinear crystal inside the optimized Nd:YLF laser cavities.

V. Power scalability and frequency agility

In this section, we summary our results of introducing two laser crystals to generate two different output frequencies [30]. Our experimental result illustrates that the output powers of the solid-state lasers based on two crystals are significantly improved. Consequently, we have improved the THz output power by mixing the two laser frequencies in a nonlinear crystal. After the second laser crystal is introduced, the THz source still maintains its compactness. Moreover, such a configuration can be used to extend to the combinations of different laser crystals for generating different output frequencies.

The experimental setup for the new configuration of laser cavities is shown in Fig.13. The input mirror 1, polarizer, output coupler make up the first cavity for lasing at 1053 nm while the input mirror 2, polarizer, output coupler make up the second cavity for lasing at 1047 nm. This cavity configuration is completely different that in Section IV above [29]. Indeed, in the previous section [29], the Nd:YLF crystal was placed at the arm sharing the 1047 nm and 1053 nm cavities. As a result, the two radiation beams at 1047 nm and 1053 nm transition accessed the same population inversion within a single Nd:YLF crystal pumped by the same diode laser. Since the 1047 nm and 1053 nm beams have perpendicular polarization directions, we introduced an intracavity polarizer to separate the two beams into the two cavities. This approach alleviated the gain competition between the 1047 nm and 1053 nm radiation beams inside the Nd:YLF crystal. However, due to the competition, we observed the unbalanced powers between the two frequencies, see Section IV above [29]. In our current configuration, we introduced two Nd:YLF crystals (*a*-cut, Nd doped at 1.0%, and $4 \times 4 \times 10$ mm³), labeled as laser crystals 1 and 2 in Fig. 13. The two laser crystals were placed at the two divided arms decoupled by the polarizer. As a result, the 1047 nm and 1053 nm transition beams now access the population inversions from two separate gain media, representing an ultimate solution to the gain competition. An acoustic-optic Q-switch was placed at the shared arm of the two laser cavities. Thus, the dualfrequency pulses are synchronized by simultaneously modulating the losses of the two cavities. The other improvement of the cavities is an output coupler shared by the two cavities, i.e. a concave mirror (curvature = 15 cm) having a reflectivity of R=75% at both 1047 nm and 1053 nm. Since the dual-frequency beams are emitted from the same output coupler, see Fig. 13, the two beams are collinearly propagating. In order to generate a THz output based on frequencymixing, we can simply place a nonlinear crystal right after the output coupler. Therefore, the THz source is truly compact.

The two diode pump beams at 808 nm are collimated and focused onto the Nd:YLF crystals through a couple of convex lenses, respectively. The diode pump power can be tuned separately. This feature is quite critical for synchronizing the dual-frequency pulses when the lasers are operated at the pulsed mode. In such a case, the pulse build-up times for the two lasers must be exactly the same. After the acoustic-optic become highly transparent, the laser pulse is not generated instantly. The pulse build-up time is the time it takes to generate the laser pulse when the Q-switch is instantly open. This value is determined by the stimulated emission cross section of the laser gain medium, loss of the cavity, output coupling coefficient, and pumping level above threshold. In fact, once the laser crystals are chosen and the cavity is well aligned, the former three factors cannot be changed. As a result, the only way for changing the pulse build-up time is the pump power level. In our configuration, we can separately change the pump power levels of the two lasers simply by varying the driving currents of the lasers.

When the repetition rate of the Q-switch is set at 5 kHz, we have measured the dependence of the Q-switched output powers on the pump powers at both 1047 nm and 1053 nm, see Fig. 14. Both of the output powers at 1047 nm and 1053 nm have increased linearly. As a result, we have completely solved the issue of the gain competition in our previous section [29]. At the pump power of 10.55 W and 9.55 W, we have obtained the output powers of 2.8 W and 1.918 W at 1047 nm and 1053 nm, corresponding to net conversion efficiencies of 26.5% and 20.1%, respectively. The corresponding slope efficiencies are 29.8% and 22.5%, respectively. Compared with the results in the previous section [29], the total output power has been improved by more than twice. In addition, the ratio of slope efficiencies between 1047 nm and 1053 nm is 1.32. This value is close to the ratio of the corresponding stimulated cross sections (i.e. 1.5) [29].

We have used a 15-mm-long GaSe crystal to measure the THz output by mixing the two laser beams, see Fig. 13. The GaSe crystal is placed directly after the output coupler. This setup is different from our previous one where a beam splitter was used to combine the two dual-frequency beams onto a nonlinear crystal. Obviously, our configuration shown by Fig. 13 is much more compact. We have measured the power dependence, see Fig. 15. At the highest incident power of 4.24 W, we have achieved an output power of 4.464 μ W at 1.643 THz (182.4 μ m). According to Fig. 15, the dependence is nearly quadratic, which is characteristic for DFG.

Compared with the previous result, we have increased the output power by 4.7. Such an enhancement is attributed to the improvement of the laser powers at 1047 nm and 1053 nm. The pulse widths at 1047 nm and 1053 nm are measured to be 17.7 ns and 11.71 ns, respectively. The laser linewidths are measured to be 77.5 GHz and 76.5 GHz, respectively. Assuming the pulse shape is Gaussian, the pulse width of THz radiation is estimated to be 9.766 ns. We have also measured the polarization of the THz radiation as a function of the azimuthal angle of a THz polarizer, shown by inset to Fig. 15. Based on the sinusoidal oscillation, the THz polarization can be determined.

One of the advantages for our configuration lies in the fact that we can generate the THz output frequency simply by choosing two different laser crystals. Since more than 100 Nd-doped laser materials or ceramics have been studied [40] with their wavelengths covering from 1.03 µm to 1.1 µm, it is conceivable for us to generate any THz output wavelength. To demonstrate the frequency agility, we have replaced the Nd:YLF crystal with a 10-mm-long and 1.0%-Nd-doped YAG crystal. All the rest of the optical components remain the same. At the diode pump powers of 9.55 W and 8.653 W, we have generated the output powers of 1.981 W and 1.706 W at 1053 nm and 1064 nm, respectively. At the repetition rate of 5 kHz, the pulse widths are measured to be 9.92 ns and 12.48 ns, respectively. The laser linewidths are measured to be 76.5 GHz and 75.0 GHz, respectively. By mixing the two laser beams on the GaSe crystal, we have generated the THz radiation at 2.983 THz (100.5 µm). The external phase-matching angle for the type II DFG is measured to be 18.3°, which is consistent to the theoretical value [34]. The highest output power is 2.09 µW. The lower output power at such a frequency, compared with that at 1.64 THz, is due to the increased absorption of the THz radiation by the GaSe crystal. The estimated THz pulse width is 7.766 ns. We have measured the THz output wavelength by scanning a Si-based etalon, see Fig. 16. According to Fig. 16, the output wavelength is to be 98 µm, which is close to 100.5 µm, calculated from the two pump wavelengths. The THz linewidth is deduced to be 42 GHz from Fig. 16. This value is close to 53.5 GHz, estimated from the laser linewidths. Using surface-emitting geometry [41,42], it is feasible for us to extend the output wavelengths to the far-infrared region.

VI. THz generation by passively Q-switched laser pulses

In this Section, we summarize our results on THz generation by mixing the passively Qswitched laser pulses in a nonlinear crystal. When two Nd:YLF crystals share a Cr:YAG crystal functioning as a single passive Q-switch, the timing jitter between each pair of dual-frequency pulses generated by the two crystals has been reduced by a factor of 20. Such a reduction in the timing jitter allows us to generate THz pulses by focusing such a passively Q-switched laser beam onto a nonlinear crystal. Such a result represents the first step for us to eventually implement a compact THz source based on ultra-compact microchip lasers. Our success in the THz generation is primarily attributed to our novel design of the cavity for the passively Qswitched dual-frequency Nd:YLF laser, see Fig. 17.

Passively Q-switched lasers [43] have completely revolutionized solid-state laser industry. However, the pulses generated by these lasers suffer from large timing jitters. Such a disadvantage severely limits their applications in differential optical absorption spectroscopy [44] and THz generation [29]. In the past, several approaches for reducing the timing jitter were investigated [45-49]. In order to synchronize each pair of the pulses generated by two passively Q-switched lasers, specific cavities were exploited [49-51].

The laser consists of two cavities being jointed together by a polarizer (P). The two cavities are used to generate two beams at 1047 nm and 1053 nm with the orthogonal polarizations (Π and σ). The polarizer being placed at the Brewster's angle has a high transmittance of T \approx 98% for the Π -polarized beam and a high reflectivity of R > 99.9% for the σ -polarized beam. This configuration is different from our previous work [30]. In this cavity, a passive Q-switch made of a 1-mm-thick Cr4+:YAG crystal with an initial transmittance of 90% (i.e. under a sufficiently low input intensity) is shared by the two cavities. Without introducing any complicated circuit and electronic driver, such a passive Q-switch can be fully integrated with the laser crystal [43]. Thus, the dimension of the entire laser can be reduced to 1-2".

Before the dual cavities simultaneously reached the lasing conditions, we tested the timing jitter of the laser pulses at 1047 nm. While the pulse width of the laser was measured to be about 80 ns, the timing jitter was higher than 400 ns. Since the corresponding pulses at the two wavelengths are no longer synchronized, such a dual-frequency laser system is not suitable to THz generation based on difference-frequency generation (DFG) or sum frequency generation (SFG). Due to the unique design of the cavities, we effectively reduced the timing jitter between the laser pulses at the two frequencies. First, the dual-frequency laser pulses are self-triggered

through the occurrence of multiple pulsing [39]. The opening time of the Q-switch is shorter than the pulse build-up time. When the first pulse is generated, the loss of the Q-switch has not reached the minimum. However, the population inversion has not been depleted completely. As the loss the Q-switch is reduced further, an additional pulse is generated shortly after the first one. Via our unique design, multiple pulsing is used to our advantage. As the first pulse is generated in the 1047-nm laser, the passive Q-switch becomes saturated and changes the cavity loss for both of the 1047-nm and 1053-nm laser oscillators. The pulse at 1053 nm is generated shortly after the first pulse at 1047 nm. In the meantime, due to the multiple-pulsing effect, the second pulse at 1047 nm is also generated. As a result, the second pulse at 1047 nm and pulse at 1053 nm can be synchronized, and therefore, they can be used as the input mixing signals for DFG and SFG.

Besides, we were able to manipulate the instance when each pulse is generated. This was accomplished by controlling the pumping current for each output frequency, and therefore, we synchronized the pulses at the two different frequencies. In our experiment, we varied the pump current for the laser at 1053 nm while keeping the pump current for the laser at 1047 nm at a fixed value. The temporal profiles are shown in Fig. 18; all of the measurements were triggered by the laser pulses at 1047 nm.

When the current was low, the laser pulse at 1053 nm exhibited a random temporal profile, and therefore, the timing jitter between the dual-frequency pulses is expected to be large. As the pump current for the laser at 1053 nm was increased, the laser pulse at 1053 nm moved slowly toward the laser pulse at 1047 nm. At some current, the laser pulse at 1053 nm became stable, and therefore, the timing jitter was reduced to about 40 ns, see inset (a) (zone II). This represents the reduction of the timing jitter by a factor of 20. It is attributed to the presence of the passive Q-switch. Indeed, the laser pulse at 1047 nm was generated ahead of the laser pulse at 1053 nm due to a higher pumping level for the laser cavity at 1047 nm. The laser pulse at 1047 nm saturated the passive Q-switch, and therefore, reduced the loss for the Q-switch from 10% to a negligible amount. Since the laser cavity at 1053 nm shared the same Q-switch, the laser at 1053 nm started to emit pulses shortly after the laser pulses at 1047 nm.

Based on our experiment, such a mechanism can be used to significantly reduce the timing jitter for the laser pulses at 1053 nm relative to the laser pulses at 1047 nm. However, as the pump current was further increased, the laser cavity at 1053 nm can reach the lasing threshold

without the loss modulation by the 1047 nm pulse. As a result, it was not possible for us to precisely control the instance when the laser pulses were generated at 1053 nm, see inset (b) (zone III). Besides, it was hard for us to determine the wavelength at which the first pulse was generated. But once the first pulse was generated, the loss in the Q-switch was saturated and the pulse at the other wavelength was correspondingly triggered. When the pump current reached a certain value, the laser pulses at 1053 nm were generated first. Since the loss in the Q-switch was reduced, the laser pulses at 1047 nm were subsequently generated. As a result, the temporal profile became relatively stable again, see inset (c) (zone IV). When the pump current became even higher, the temporal profile became relatively unstable, see inset (d) (zone V), similar to the profiles in zone I.

Due to the significant reduction of the timing jitter between each pair of the dual-frequency pulses, these pulses can be used for achieving THz generation based on frequency mixing. Indeed, we have successfully generated THz pulses from a 15-mm GaSe crystal. We believe that this is the first demonstration of THz generation from passively Q-switched dual-frequency laser pulses. While keeping the pump current for the laser cavity at 1047 nm as a constant, we have varied the pump current for the laser cavity at 1053 nm. According to Fig. 19, the behavior of the THz output power vs. the pump current is peculiar. Through our analysis, we have concluded that such a behavior is strongly correlated with the timing jitter between each pair of the dualfrequency laser pulses. Indeed, the first THz peak from the low-current side in Fig. 19 corresponds to the region between zones I and II in Fig. 18 where the second pulse at 1047 nm synchronizes well with the pulse at 1053 nm. Then, as the pump current increases, the pulse at 1053 nm moves to the center part between the two pulses at 1047 nm whereas none of the pulses at these two wavelength could be synchronized. This corresponds to the first dip in the THz power around the pump current 1.2 A. As the pump current is further increased, either 1053 nm or 1047 nm pulse may be generated first, shown in Fig. 18(b) (zone III). However, once being triggered by the first pulse at one wavelength, the second pulse can still be synchronized with the pulse at the other wavelength. As a result, the THz radiation can still be efficiently generated. This region corresponds to the second peak in the THz power in Fig. 19. Then, after the current reaches around 1.4 A, the pulse at 1053 nm is generated first and becomes dominant as the triggering pulse. Similarly, as the pulse at 1047 nm moves around the center point between the two 1053 nm pulses, few pulses at the two wavelength will be synchronized. Accordingly, the THz power has the second dip. The last peak in terahertz power is attributed to the mixing between the 1047-nm pulse and second pulse at 1053 nm.

Besides the THz generation, we have also demonstrated SFG by using this dual-frequency passively Q-switched laser as the two mixing beams. In order to understand the synchronization between the SF pulses and second-harmonic pulses, a dual-frequency laser beam was divided into two beams using a beamsplitter. Each SF pulse was generated by a KTP crystal by one of the split beams. The other split beam, used for SHG, was sent through a half-wave plate and a cubic polarizer. Due to the perpendicular polarizations of the beams at 1047 nm and 1053 nm, either the 1047-nm beam or 1053-nm beam can be blocked completely by rotating the half-wave plate. Another KTP crystal was used for achieving second-harmonic generation (SHG). In Fig. 20(a), the first pulse of the SH beam at 1047 nm was generated by the first pulse at 1047 nm that triggered the following pulses at 1047 nm and 1053 nm. Fig. 20(b) shows the pulse profiles of the SF and SH beams. Our measurement yielded a timing jitter of 5 ns, which is much smaller than the value for the timing jitter between each pair of the pulses at 1047 nm and 1053 nm. In this approach, we synchronized the pulses at two different wavelengths based on a passively Q-switched laser.

VI. Concluding remark

We have reviewed our recent results on THz generation based on difference frequency generation. In particular, we demonstrated that just using four reversely stacked GaP plates, we achieved the maximum conversion efficiency of 40% in terms of photon numbers. By adding the fifth GaP plate to the stack, the THz output power was significantly reduced from that for the four stacked plates. This is the evidence on back conversion. On the other hand, to make our THz source truly compact and portable, we took a completely different approach to the THz source. Indeed, by mixing two frequencies generated by a dual-frequency Nd:YLF laser we implemented a compact and portable THz source with an average output power reaching 1 μ W. By introducing two Nd:YLF crystals instead of one inside the laser cavity, we significantly scaled up the output power to 4.5 μ W. Through further optimizations and utilizing novel configurations, we will be able to improve the THz output power to the mW level. The current dimension of our THz source is about 12"×12"×6", which can be readily reduced to 12"×6"×4" through the further optimizations. Our future goal is to develop a THz source as compact as a laser pointer,

operating at room temperature. Such a dimension can be achieved by exploiting the passively-Q-switched laser pulses.

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Fig. 1. Absorption coefficient is plotted vs. wavelength following the measurement of transmission and reflection spectra on a single GaP plate.



Fig. 2. THz output power was measured vs. output wavelength for a single GaP plate. As discussed in the text, the modulations were caused by convolution between Fabry-Perot effect for THz wave and water-vapor absorption.



Fig. 3. Output peak power vs. wavelength: open circles – single GaP plate; squares – stacked two GaP plates; dots – stacked three GaP plates. For both of the stacked two and three plates, second-order nonlinear coefficients have same sign between adjacent plates.



Fig. 4. Output peak power vs. wavelength: squares – stacked two GaP plates; open circles – stacked three GaP plates. For both of the stacked two and three plates, second-order nonlinear coefficients have the opposite signs between adjacent plates.



Fig. 5. Spectrum of THz output generated by four reversed GaP plates, covering range of 78-300 μm.



Fig. 6. Spectrum of THz output generated by aligned four GaP plates, covering tuning range of 135-500 μm.



Fig. 7. Conversion efficiency for THz generation versus input power at 1.074 μm for four reversed GaP plates (red dots) and 1.069 μm for four aligned GaP plates (blue circles). Blue dashed curve (bottom) represents linear fit, whereas the red curve (top) is fitted using Eq. (2).



Fig. 8. Highest THz output power generated by reversed GaP plates versus the number of plates. Blue curve represents nonlinear least squares fit to data points using Eq. (2).



Fig. 9. Experimental setup for THz DFG based on compact and portable dual-frequency Nd:YLF laser having dual cavities: IM, laser input mirror; P, polarizer; OC1-OC2, laser output couplers; L1-L3, convex lenses; M1-M3, high reflection mirrors; WP1-WP2, half-wave plates; PE, high density white polyethene filter; and PM1-PM2, parabolic mirrors.



Fig. 10. Dependence of powers from dual-frequency Q-switched Nd:YLF laser on pump power from laser diode.



Fig. 11. Temporal profiles of dual-frequency output pulses generated by from Q-switched Nd:YLF laser. Ch1: 1053 nm and Ch2, 1047 nm.



Fig. 12. Dependences of average and peak output powers on sum of the output powers generated by the single Nd:YLF laser. Solid curve corresponds to quadratic fit to data points. Inset: plot of power being transmitted through a Si etalon vs. distance between two Si plates in the etalon.





Fig. 13. Experimental layout for a configuration of the dual-frequency solid-state laser and compact THz source. The dual-frequency laser cavities are marked by the dashed border line.



Fig. 14. Q-switched output power vs. pump power at 1047 nm and 1053 nm. Dots and squares correspond to data points; straight lines correspond to linear fits to data points.



Fig. 15. THz average and peak power dependence on the combined power at 1047 nm and 1053 nm; straight lines correspond to quadratic fits to data points. Inset is the intensity of the THz radiation as a function of the azimuthal angle of a THz polarizer.



Fig. 16. THz power at 100.5 μm, generated by mixing dual frequencies from Nd:YLF and Nd:YAG lasers, being transmitted through a Si etalon as a function of the displacement between two Si wafers.



Fig. 17. Experimental setup of the dual-frequency passively Q-switched Nd: YLF solid-state laser. IM 1 and 2, input mirror; P, polarizer; OC, output coupler.



Fig. 18. Temporal profiles of dual-frequency laser pulses.



Fig. 19. THz intensity vs. pump current of 1053 nm laser.



Fig. 20. a) Synchronized pulses of SFG and SHG of 1047 nm; (b) Synchronized pulses of SFG and SHG of 1053 nm.