

# Efficient and widely step-tunable terahertz generation with a dual-wavelength CO<sub>2</sub> laser

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**Abstract** Coherent terahertz radiation in a widely step-tunable range of 72.3–2706  $\mu\text{m}$  (0.11–4.15 THz) has been generated in GaAs crystal by difference-frequency generation using one CO<sub>2</sub> laser with dual-wavelength output. The peak power of THz pulse reaches 35 W at the wavelength of 236.3  $\mu\text{m}$ , which corresponds to a pulse energy of 2.1  $\mu\text{J}$ . An average power of 10  $\mu\text{W}$  has been achieved when working repetitively. This efficient terahertz radiation source is more compact and widely tunable than other THz sources pumped by CO<sub>2</sub> laser.

## 1 Introduction

A tunable and coherent terahertz (THz) source has attracted great attention for its potential applications in chemical identifications, biomedical diagnostics and THz spectroscopy [1–3]. Among most of the THz sources, difference-frequency generation (DFG) in nonlinear crystals is one of the effective ways to generate high-power, tunable and narrowband coherent terahertz radiation.

Recently, THz generations based on DFG have been demonstrated in many nonlinear optical crystals, such as GaSe [4, 5], ZnGeP<sub>2</sub> [6], GaP [7, 8], LiNbO<sub>3</sub> [9] and GaAs [10–14], but most of them were pumped by near-infrared laser with the wavelength around 1  $\mu\text{m}$ . According to the Manley–Rowe relation, the maximum conversion efficiency can be improved by one order of magnitude when using CO<sub>2</sub> laser with longer wavelength running at 10  $\mu\text{m}$ . Although the

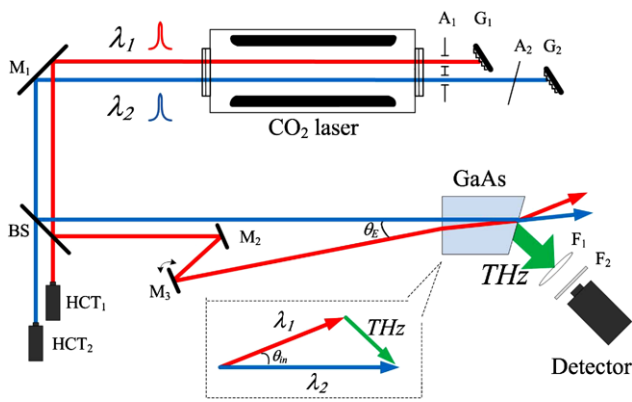
DFG terahertz sources pumped by CO<sub>2</sub> lasers were also reported before [10–14], they were established with two lasers or two independent discharge sections. These made the systems bulk and complex. Furthermore, among the variety of nonlinear crystals, GaSe and GaAs meet the requirements for having large nonlinear coefficient and high transparency in both 10  $\mu\text{m}$  band and terahertz region. But GaSe crystal suffers from large Fresnel reflection losses since it is difficult to be cut or coated. Considering the maturity of CO<sub>2</sub> laser technology and large-aperture GaAs crystals are commercially available, it will be an effective way to generate THz radiation based on DFG in GaAs crystals with CO<sub>2</sub> laser.

In this paper, an efficient and widely step-tunable THz source based on DFG in GaAs crystal with a dual-wavelength CO<sub>2</sub> laser is demonstrated. Compared with previous results, this THz source is pumped by a single CO<sub>2</sub> laser, which is more compact and easy to be established. This system can also generate THz radiation efficiently in a more widely tuning range of 72.3–2706  $\mu\text{m}$  (0.11–4.15 THz) in GaAs crystal at room temperature.

## 2 Experimental setup

The experimental setup is shown in Fig. 1. A self-made short pulsed TEA CO<sub>2</sub> laser was used as the fundamental pump source. The laser had a discharge volume of  $2 \times 3 \times 100 \text{ cm}^3$  with UV preionization and working at a maximum repetition rate of 10 Hz [15]. Independent controls of multiline emission from a TEA CO<sub>2</sub> laser have been successfully used before [16, 17]. For our dual-wavelength laser system, two intra-cavity gratings were placed before and after and two laser lines oscillated independently in a shared discharging section. The two beams propagated in parallel, both of which could be tuned from 9.2  $\mu\text{m}$  to 10.7  $\mu\text{m}$  by the two

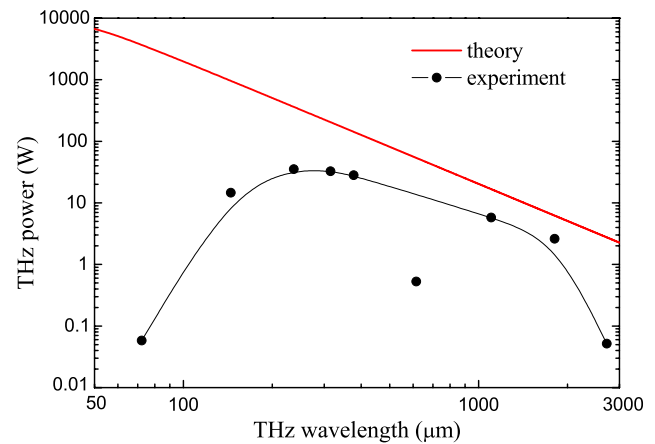
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**Fig. 1** Experimental setup for THz generation based on DFG in a GaAs crystal.  $G_1$ ,  $G_2$ , gratings;  $A_1$ , iris;  $A_2$ , attenuator;  $M_1$ – $M_3$ , mirrors;  $BS$ , beam splitter;  $F_1$ ,  $F_2$ , tsurupica lens ( $f = 18$  cm) and quartz filter, respectively.  $HCT_1$ – $HCT_2$ , HgCdTe detectors; Detector, both of Golay cell and pyroelectric detector were used for THz energy detection

plane blazed gratings (150 lines/mm). To synchronize the two optical pulses, an intra-cavity attenuator  $A_2$  was introduced to compensate the gain differences between two laser lines. The pulse energy of each line was measured to be about 120 mJ with a pulse duration of 60 ns. The HgCdTe detectors  $HCT_1$ – $HCT_2$  were used for the inspection of the time synchronization between the two lines.

The GaAs crystals used in this experiment had a high resistivity ( $>10^7 \Omega \text{ cm}$ ). The crystals were cut along [111] direction and the input face had an  $(\bar{1}10)$  orientation with an area of  $20 \times 20 \text{ mm}^2$ . The entrance surface was coated with anti-reflection film at  $10 \mu\text{m}$ . In order to avoid the total reflection of THz radiation at the output face, we cut the crystals at an angle of  $\sim 10^\circ$  as shown in Fig. 1. For the isotropic GaAs crystal, the coherence length for the collinearly phase-matching configuration by mixing with CO<sub>2</sub> laser beams is of the order of the generated THz wavelength. Consequently, it is impossible to obtain high THz power output in such GaAs plates whose thickness is of the order of millimeter or less. However, the phase matching for the DFG in GaAs crystal can be achieved by a simple noncollinear geometry of Fig. 1. Two unfocussed infrared beams possessed the same polarization parallel to the [111] direction. They were ensured by a noncollinear geometry to satisfy the phase-matching conditions in GaAs crystal. The mirror  $M_3$  could be rotated to modify the phase-matching angles. The THz wave generated from GaAs crystal was focused into detectors with a tsurupica convex lens, and a crystalline quartz plate was used to block scattering pump radiation. The transmittance of these filters was about 70% at the terahertz region. Indeed, we used both of Golay cell (TYDEX, GC-1P) and a sensitive pyroelectric detector (Spectrum, SPJ-A-8-OB) to the measure THz energy, and the Golay cell with a known spectral response was calibrated by using this pyroelectric detector.



**Fig. 2** THz peak power at different generated THz wavelengths, continuous curve represents theoretical THz power without considering absorption and dots correspond to the experimental data

### 3 Results and discussions

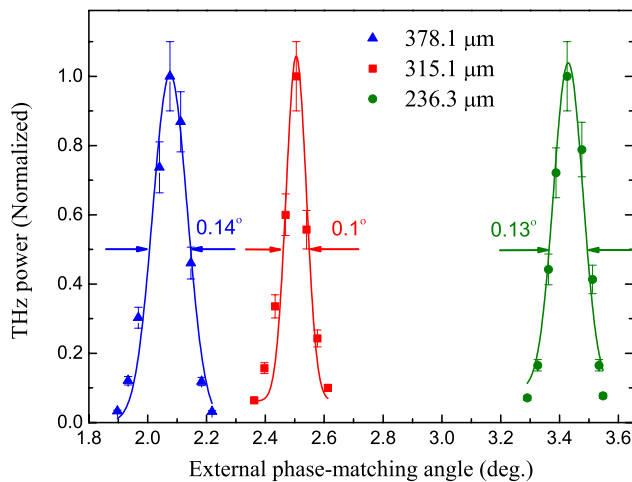
Efficient generation of coherent terahertz radiation was achieved in GaAs crystal with tunability in a wide range of  $72.3$ – $2706 \mu\text{m}$  ( $0.11$ – $4.15$  THz). We represent the spectra dependence of THz peak power in Fig. 2. Compared with previous results [11, 12], we have achieved a more wider tuning range of THz generation in GaAs crystal at room temperature. The maximum power peaked at the wavelength of  $236.3 \mu\text{m}$ , which was generated in a 23-mm-long crystal and pumped by the 10R(22) line ( $10.23 \mu\text{m}$ ) and 10P(30) line ( $10.69 \mu\text{m}$ ) with a power intensity  $3 \text{ MW/cm}^2$  of each beam. We detected about  $2.1 \mu\text{J}$  of pulse energy with corresponding peak power of 35 W assuming a 60 ns THz pulse. It led to an external power conversion efficiency of  $1.8 \times 10^{-5}$ . One can estimate the THz output power without absorption by using the expression [18] in the plane-wave fixed-field approximation:

$$P_T = \frac{8\pi^2 d_{\text{eff}}^2 L_{\text{eff}}^2}{\varepsilon_0 c n_T n_1 n_2 \lambda_T^2} \frac{P_1 P_2}{A} T_T T_2 T_3$$

with the expression of  $L_{\text{eff}}$  [11]

$$L_{\text{eff}} = \frac{D}{\sin \phi} \left( \frac{L}{D} \sin \phi - \frac{1 + \sin^2 \phi}{3 \cos \phi} \right)^{1/2}$$

where  $\lambda_T$  is the output terahertz wavelength,  $n_i$  and  $T_i$  ( $i = T, 1, 2$ ) denote the refractive indices and Fresnel transmittance of three participating waves, respectively. Here  $d_{\text{eff}} = 50 \text{ pm/V}$  and  $L$  is the crystal length 23 mm.  $P_1$ ,  $P_2$  are the pump powers and the area of pump beams  $A$  is  $0.7 \text{ cm}^2$ . The diameter of the input beams  $D = 9 \text{ mm}$  and the output angle  $\phi \simeq 21^\circ$  for the experimental conditions. For our 23-mm-long GaAs crystal, the calculated equivalent interaction length  $L_{\text{eff}} = 18 \text{ mm}$ . For this value of  $L_{\text{eff}}$ , the theo-

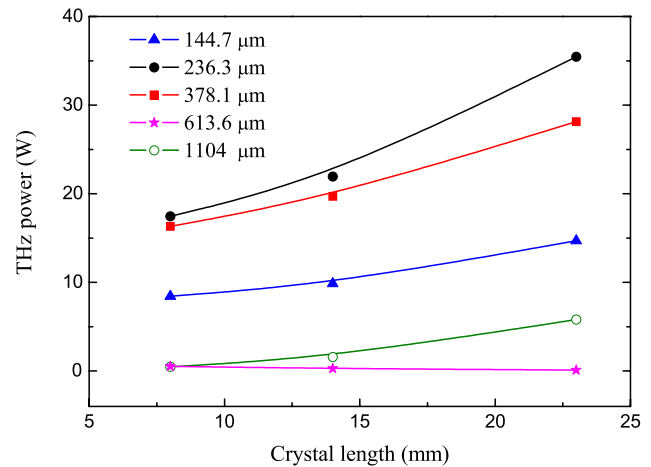


**Fig. 3** External phase-matching tuning curves at 377.4 μm (triangles), 315.1 μm (squares) and 236.3 μm (dots) with 14-mm-long crystal

retical THz powers without considering absorption are represented in Fig. 2 with continuous curve. The differences between experimental data and theoretical predictions were mainly due to THz absorptions in the GaAs crystal. Additionally, the limited area of the output surface was not large enough to let out all the THz radiation inside the GaAs crystal. This may in part be responsible for the modest conversion efficiency.

Note that the power decreased when the THz wavelength was below 200 μm (above 1.5 THz), this was owing to the increasing photon absorption in GaAs crystal at room temperature [19]. In order to enhance the output power in this region, cooling of GaAs crystal may be required. The shortest wavelength we obtained was 72.3 μm, which was generated by mixing the 9R(12) line (9.32 μm) and 10P(30) line (10.69 μm). It is close to the limit of the short-wavelength end that transparent in GaAs at room temperature. Moreover, it should be mentioned that a sudden dip appears at 613.6 μm. We believe that it was due to an absorption peak of GaAs crystal at this wavelength, since we found that the THz output power decreased with a longer crystal length. Compared with the report before [12], we have also extended the long-wavelength end of tuning range from 600 μm to 2706 μm, corresponding to the pump wavelengths of 10P(22) and 10P(26) lines. In addition, due to the multi-mode structure of our pulsed CO<sub>2</sub> laser, the bandwidths of the pump waves were 250 MHz (full width at half maximum) approximately. Therefore, the linewidths of the THz waves should be narrower than those of the pump waves, which could provide a narrow bandwidth output.

We measured the external angles when noncollinear DFG processes were phase matched inside a 14-mm-long GaAs crystal, which were shown in Fig. 3. For the 236.3 μm, the maximum output power centered at the external angle  $\theta_E = 3.4^\circ$  (corresponding to an internal angle of  $1^\circ$ ).

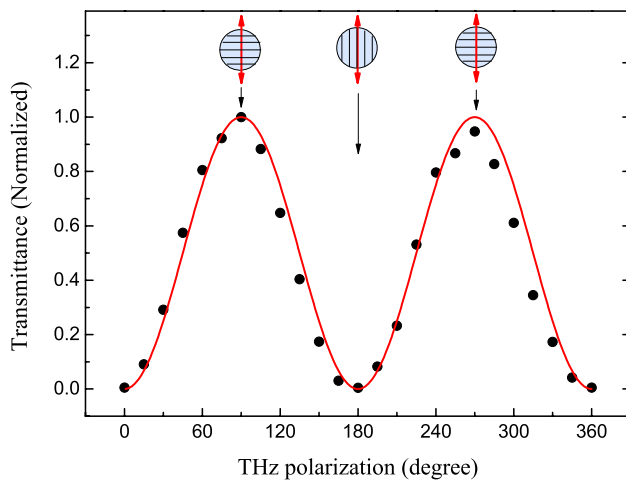


**Fig. 4** THz peak power versus GaAs crystal length

It was in a good agreement of the theoretical calculation  $3.38^\circ$  by using the Sellmeier equation given by Skauli et al. [20]. When the THz source generated at 315.1 μm and 377.4 μm, the experimental external angles were measured to be  $2.5^\circ$  and  $2.076^\circ$ , which were also in perfect agreements with the prediction values  $2.5^\circ$  and  $2.08^\circ$ , respectively. The external phase-matching angle decreased when the generated THz wavelength increased. Meanwhile, tuning external angles showed the phase mismatch acceptance at the three THz wavelengths. The measured full width of the phase-matching curve  $\Delta\theta_E$  was  $0.1\text{--}0.14^\circ$ , and the theoretical values were about  $0.1^\circ$ , which were calculated by using an expression in [10]  $(\Delta\theta_E)_{1/2} \simeq (1/L)(n\lambda_1\lambda_2/2\Delta n)^{1/2}$ , where  $n = n_1 = n_2$ ,  $\Delta n = n_T - n$  and  $L$  is the crystal length.

Three GaAs crystals were utilized to generate THz radiation in this experiment, which were 8, 14 and 23 mm long, respectively. As shown in Fig. 4 that the 23-mm-long crystal resulted in a higher output power than the other two crystals with the same pump intensity. One can see from the figure that the THz power at 613.6 μm decreased with longer crystal length, it confirmed the assumption mentioned above that there was an absorption peak at this wavelength in GaAs crystal. Furthermore, we modified the repetition rate of CO<sub>2</sub> laser to generate higher average output power. When the pump laser was operated at 10 Hz, an average power of 10 μW was achieved at 236.3 μm. However, the average power of THz radiation not increasing linearly with the repetition rate is mainly due to the pump power did not increase linearly and/or the synchronization between two pulses changed when worked at higher repetition rate.

We further measured the terahertz polarization at 315.1 μm by using a wire grid polarizer (MICROTECH, G25), which is shown in Fig. 5. One can see that the terahertz radiation polarized linearly and the maximum transmission appeared when the THz polarization was perpendicular to the orientation of wire grid. Our experimental



**Fig. 5** Transmitted THz power after propagating through the wire grid vs. polarization angle. Dots, experimental results; solid line,  $\sin^2(\theta)$

data agreed well with the theoretical calculations. The measured polarization direction (parallel to the [111] direction of GaAs crystal) was the same as those of the pump beams.

#### 4 Conclusions

In conclusion, efficient, coherent and widely step-tunable terahertz radiation has been generated by noncollinear difference-frequency generation in GaAs crystals with a single CO<sub>2</sub> laser. This source system can generate THz pulses in the range of 72.3–2706  $\mu\text{m}$  (0.11–4.15 THz) with maximum pulse energy of 2.1  $\mu\text{J}$  (corresponding to a peak power of 35 W) and yield a average power of 10  $\mu\text{W}$  at 236.3  $\mu\text{m}$ . Compared with previous results, we have achieved the widest tuning range of THz generation in GaAs crystal at room temperature and this THz source is more compact than others pumped by two CO<sub>2</sub> lasers before.

Recently, THz-wave generation was demonstrated by difference-frequency mixing with two CO<sub>2</sub> laser beams in stacked GaAs wafers, resulting in the average output power of as high as 30  $\mu\text{W}$  [21]. The THz power was scaled up due to the quasi-phase-matched (QPM) structure of GaAs and the focussed pump beams in a collinear geometry. However,

it would be inconvenient that the period of this QPM structure should be changed every time in order to generate tunable terahertz waves. Considering the advantages of using a dual-wavelength CO<sub>2</sub> laser and large-aperture GaAs crystals are commercially available, it confirms an effective way to get high-power and widely step-tunable terahertz generation by frequency mixing of CO<sub>2</sub> laser beams in the GaAs crystal.

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