

# High Power 4.65 $\mu\text{m}$ Single-wavelength Laser by Second-harmonic Generation of Pulsed TEA $\text{CO}_2$ Laser in $\text{AgGaSe}_2$ and $\text{ZnGeP}_2$ <sup>1</sup>

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**Abstract**—A high power 4.65  $\mu\text{m}$  single-wavelength laser by second-harmonic generation (SHG) of TEA  $\text{CO}_2$  laser pulses in silver gallium selenide ( $\text{AgGaSe}_2$ ) and zinc germanium phosphide ( $\text{ZnGeP}_2$ ) crystals is reported. Experimental results show that the average output power of SHG laser is not only restricted by the damage threshold of the nonlinear crystals, but also limited by the irradiated power of fundamental-wave laser depending on the operating repetition-rate. It is found that  $\text{ZnGeP}_2$  can withstand higher 9.3  $\mu\text{m}$  laser irradiation intensity than  $\text{AgGaSe}_2$ . As a result, using a parallel array stacked by seven  $\text{ZnGeP}_2$  crystals, an average power of 20.3 W 4.65  $\mu\text{m}$  laser is obtained at 250 Hz. To the best of our knowledge, it is the highest output power for SHG of  $\text{CO}_2$  laser by far.

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## 1. INTRODUCTION

A  $\text{CO}_2$  laser is generally operated in the wavelength band of 9–11  $\mu\text{m}$ , and it can be transferred into the mid-infrared band of 4.5–5.5  $\mu\text{m}$  by means of second-harmonic generation (SHG), that is called  $\text{CO}_2$  laser frequency-doubling. Both bands are of great interest for many applications especially in the long-distance situation because they are in the atmospheric transmission window, such as environmental measurements, laser radar and so on [1–13]. For a continuous-wave (CW) operation, the optical conversion efficiency for SHG of  $\text{CO}_2$  laser is in the range of  $10^{-4}$ – $10^{-3}$ , which is too low to use practically [14, 15]. Whereas, this efficiency can be increased about two orders of magnitude in pulsed operation and the highest efficiency of ~60% is reported [16–18]. Usually, there are two types of pulsed  $\text{CO}_2$  laser used as the fundamental-laser for SHG. One is a multi-mode TEA  $\text{CO}_2$  laser and the other is single-mode or low-order-mode eclectic-optically (E–O) Q-switched  $\text{CO}_2$  laser. Using a 100 ns single-pulsed TEA  $\text{CO}_2$  laser and a  $18 \times 18 \times 52 \text{ mm}^3$   $\text{AgGaSe}_2$  crystal, Badikov obtained SHG laser with the highest output energy of 350 mJ [19]. Kato reported the highest power of 8.1 W 5.2955  $\mu\text{m}$  laser at 100 kHz by using a 10 ns E–O Q-switched  $\text{CO}_2$  laser and a 25 mm-length  $\text{AgGa}_{1-x}\text{In}_x\text{Se}_2$  crystal [20].

Kinds of nonlinear crystals had been applied in the SHG of  $\text{CO}_2$  laser, such as  $\text{ZnGeP}_2$ ,  $\text{AgGaSe}_2$ ,  $\text{AgGa}_{1-x}\text{In}_x\text{Se}_2$ ,  $\text{AgGaS}_2$ ,  $\text{GaSe}$ ,  $\text{AgGaGeS}_4$ ,  $\text{AgGaGe}_5\text{Se}_{12}$  and so on [21–26]. Among these materials,  $\text{AgGaSe}_2$  is used popularly for its small absorp-

tion coefficient in the band of 3–11  $\mu\text{m}$  [2, 20, 27].  $\text{ZnGeP}_2$  has a bigger optical absorption beyond the band of 3–8  $\mu\text{m}$ , but it has a potential for SHG of  $\text{CO}_2$  laser because of its comprehensive merits, including larger nonlinear coefficient ( $d_{36} \approx 75 \text{ pm/V}$ ), higher thermal conductivity and damage threshold [18, 28, 29].

In this paper, a high power SHG of TEA  $\text{CO}_2$  laser is demonstrated. Fundamental-laser is improved by removing tail pulse and selecting 9.3  $\mu\text{m}$  single-wavelength output. Frequency-doubling system is designed and optimized by using  $\text{AgGaSe}_2$  or  $\text{ZnGeP}_2$  crystals. Employed a parallel array of seven  $\text{ZnGeP}_2$  crystals, maximum average power of 20.3 W 4.65  $\mu\text{m}$  laser is obtained at 250 Hz.

## 2. EXPERIMENTAL SETUP

As shown in Fig. 1, the experimental setup is mainly consisted of a TEA  $\text{CO}_2$  laser system and a frequency-doubling system. The resonator contains a gold coating mirror (M1) and a ZnSe output coupler with special coating films (M2). Brewster plate in the resonator makes the output beam be horizontal polarization. In the frequency-doubling system,  $\text{AgGaSe}_2$  in serial or parallel fashion and  $\text{ZnGeP}_2$  in parallel array are designed and used, as shown in Figs. 1a, 1b, and 1c, respectively. Parallel array is consisted by seven 12 mm-length  $\text{ZnGeP}_2$  crystals. Total area of the array is 5.76  $\text{cm}^2$  (24 mm  $\times$  24 mm), which is quite different from serial and parallel fashions for the available beam size of fundamental-laser is enlarged. The 9.3  $\mu\text{m}$  fundamental-laser beam with suitable size and intensity is

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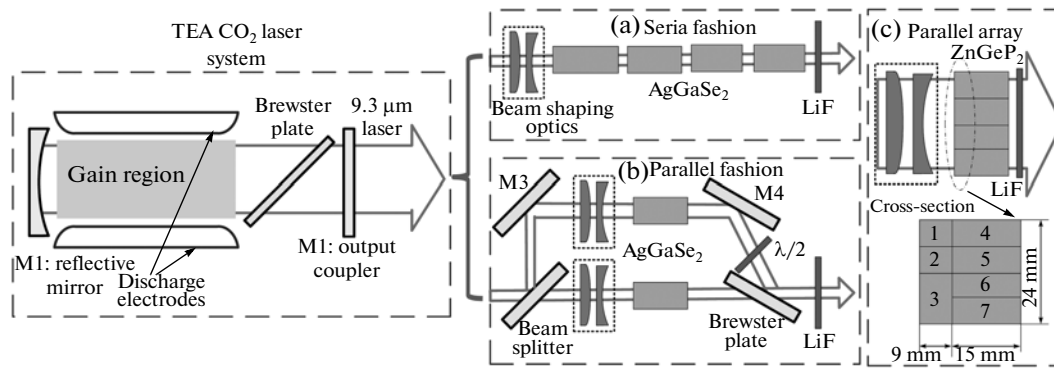


Fig. 1. Experimental setup for high power SHG of TEA CO<sub>2</sub> laser.

changed by passing through beam shaping optics before irradiating the crystals. LiF cut-off filters are used for passing the 4.65  $\mu\text{m}$  frequency-doubling laser and removing the 9.3  $\mu\text{m}$  fundamental-laser thoroughly.

### 3. EXPERIMENTAL RESULTS AND DISCUSSION

A free-running TEA CO<sub>2</sub> laser generally radiates a pulsed beam with multi-wavelengths, which has a spectral distribution centered at 10.5910  $\mu\text{m}$  between 9–11  $\mu\text{m}$ . The output pulse is composed by a spike pulse and a tail pulse with multi-microsecond duration [30–33], as shown in Fig. 2a. The tail pulse contains more than 50% pulse energy, resulting in a lower peak power and restricting the efficient transfer to SHG laser. Experimental results show that the property of the tail pulse correlates with the parameters of TEA CO<sub>2</sub> laser closely. When the parameters are chose properly, the tail pulse can be eliminated completely, as shown in Fig. 2b [34]. According to [35], by coating special films on the optical surface of the output coupler, the TEA CO<sub>2</sub> laser realizes single-wavelength

output at 9.3054  $\mu\text{m}$ . As a result, improved TEA CO<sub>2</sub> laser is obtained with single pulse energy of 10J and tailless pulse width of 80–100 ns. The laser can be operated at single-pulsed or repetition-rate mode, and the repetition-rate can be changed from 1 to 300 Hz. The output power is raised linearly with the increase of repetition-rate because the pulse energy can be kept invariable. The output laser is high-order transverse mode, and the beam area is  $\sim 30 \times 30 \text{ mm}^2$  with a homogeneous intensity distribution, as shown in Fig. 2b. In addition, the output laser is horizontal polarization.

Restricted by the grown technology, nonlinear crystals for SHG of CO<sub>2</sub> laser with large size are difficult to obtain. The size of AgGaSe<sub>2</sub> and ZnGeP<sub>2</sub> crystals used in our experiments are in  $\varnothing 10 \text{ mm} \times (10\text{--}20) \text{ mm}$  approximately. The crystals are cut for a Type I critical-phase-matching of 9.3  $\mu\text{m}$  fundamental-laser irradiating vertically, and both surfaces are coated by anti-reflection (AR) at 4.65 and 9.30  $\mu\text{m}$ . In our prior work, SHG of TEA CO<sub>2</sub> laser by using single AgGaSe<sub>2</sub> crystal had been studied, and the highest power of 0.94 W 4.65  $\mu\text{m}$  laser was obtained [34]. To further increase the output power, AgGaSe<sub>2</sub> crystals in serial and parallel fashions are investigated.

In serial fashion using single-pulsed CO<sub>2</sub> laser, four AgGaSe<sub>2</sub> crystals with the length of 8, 10, 15, and 20 mm are used as described in Fig. 1a. By analyzing and comparing of the experimental results through changing the optical axis orientation of the crystals and exchanging the location and sequence of each crystal, it is found that the highest output energy of SHG laser is 2–2.5 times than that using single crystal. The sum length of crystals is optimized to be 30–40 mm. Exceeding the optimal length, the output energy cannot be further enhanced. Experimentally, the theory has never been validated that the output energy is proportional to the square of the crystal

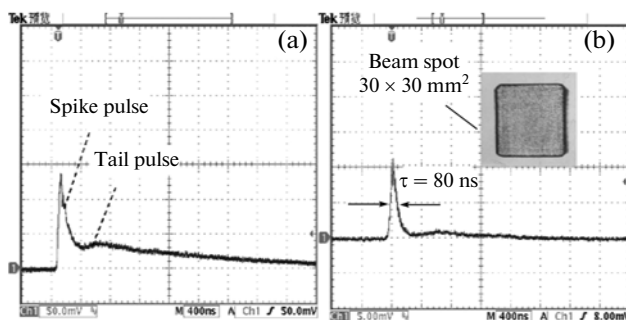


Fig. 2. TEA CO<sub>2</sub> laser pulse shape (a) free-running laser (b) improved laser.

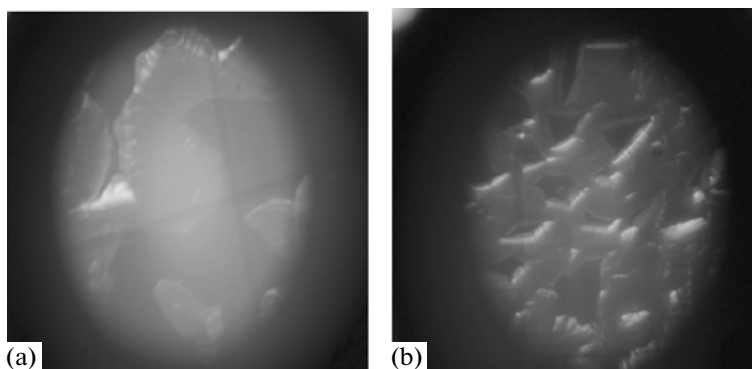


Fig. 3. Micrograph of AR films damage for  $\text{ZnGeP}_2$ .

length. More crystals are used in series, lower conversion efficiency per unit length is induced.

In parallel fashion using single-pulsed  $\text{CO}_2$  laser, two ways of  $\text{AgGaSe}_2$  are designed as shown in Fig. 1b. The total output energy is 1.1–1.5 times than that using a single crystal. Only the output of the first way maintains the original state, while the SHG conversion efficiency in second way is much lower. This can be attributed to the polarization of the fundamental beam is changed in the complex parallel fashion. When the laser beam is divided, transmission, reflection and refraction processes exist, resulting in laser performance degradation inevitably. Furthermore, additional optical loss is induced when the  $\lambda/2$  plate and Brewster plate are inserted for the synthesis of the output beam, reducing the overall SHG efficiency.

In order to increase the SHG output power, it is the key issue that the nonlinear crystal can withstand higher repetition-rate  $\text{CO}_2$  laser radiation below damage threshold. Irradiated by the  $\text{CO}_2$  laser pulses with an energy density of  $0.7 \text{ J/cm}^2$ ,  $\text{AgGaSe}_2$  sample is cracked at 100 Hz and its power damage density is estimated to be  $\sim 70 \text{ W/cm}^2$ . Experimental results indicate that the body damage will be induced if the temperature change was over  $5^\circ\text{C/min}$ . Obviously, the cause of this phenomenon is that poor thermal properties of  $\text{AgGaSe}_2$ , for example, thermal expansion coefficient is opposite at two directions ( $19.8 \times 10^{-6} / \text{K}$  ( $\perp c$ -axis) and  $-8.1 \times 10^{-6} / \text{K}$  ( $\parallel c$ -axis)) and the thermal conductivity ( $0.011 \text{ W/(cm K)}$ ) is relative lower. For  $\text{ZnGeP}_2$  sample, the damage threshold is measured to be  $1.3 \text{ J/cm}^2$  in single-pulsed operation. Irradiated by the  $\text{CO}_2$  laser pulses with an energy density of  $1.0 \text{ J/cm}^2$ , only AR film damage occurs at 300 Hz, but no crystal surface and body damage are observed, as shown in micrographs in Fig. 3. Power damage density of  $\text{ZnGeP}_2$  is  $\sim 300 \text{ W/cm}^2$ , which is 3–4 times higher than that of  $\text{AgGaSe}_2$ . Therefore, comparative with  $\text{AgGaSe}_2$ ,  $\text{ZnGeP}_2$  crystal is more suitable for high power SHG operation.

Accordingly, the output power of SHG laser cannot be enhanced significantly by the serial or parallel fashion. For high power SHG output, experiments are carried out by using a parallel array of  $\text{ZnGeP}_2$  crystals, as shown in Fig. 1c. Using the parallel crystal array, not only the irradiating power of  $\text{CO}_2$  laser can be enhanced, but also available size of crystals is enlarged. Under the irradiation of  $\text{CO}_2$  laser with the energy density of  $1.0 \text{ J/cm}^2$ , the output energy of SHG laser versus the area of nonlinear crystal array is shown in Fig. 4. With the increase of nonlinear crystals, the output energy is increased linearly. The maximum SHG output energy of 197 mJ is obtained, with an energy conversion efficiency of 3.4%. The output energy is in direct proportion with the area of crystal array, so SHG laser can be further enhanced using the parallel array with more crystals if enough fundamental-laser energy can be provided.

As shown in Fig. 5, high-repetition-rate operation is investigated using parallel  $\text{ZnGeP}_2$  array, and high

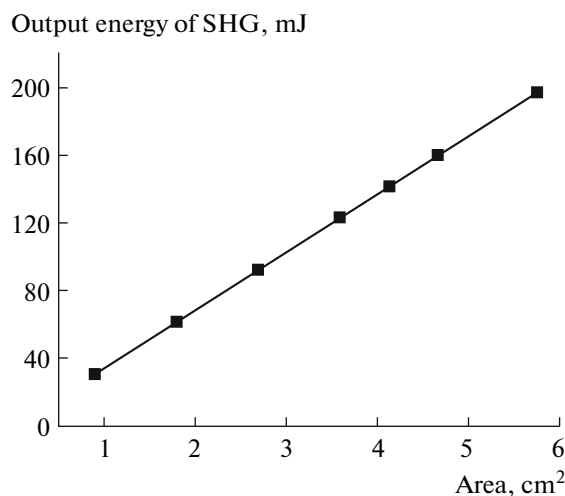


Fig. 4. Output energy of SHG laser versus the area of  $\text{ZnGeP}_2$  crystals.

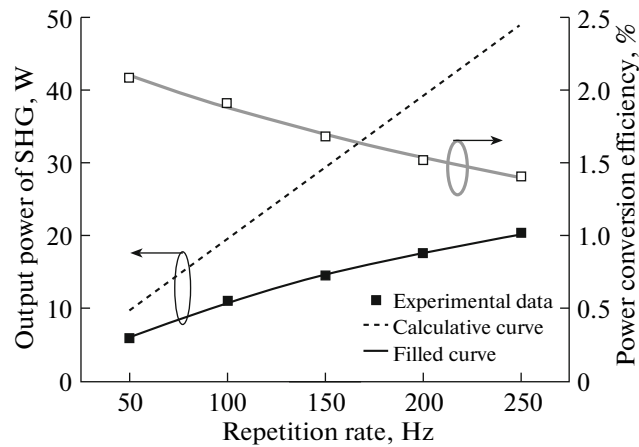


Fig. 5. Output power and power conversion efficiency of SHG laser versus repetition rate.

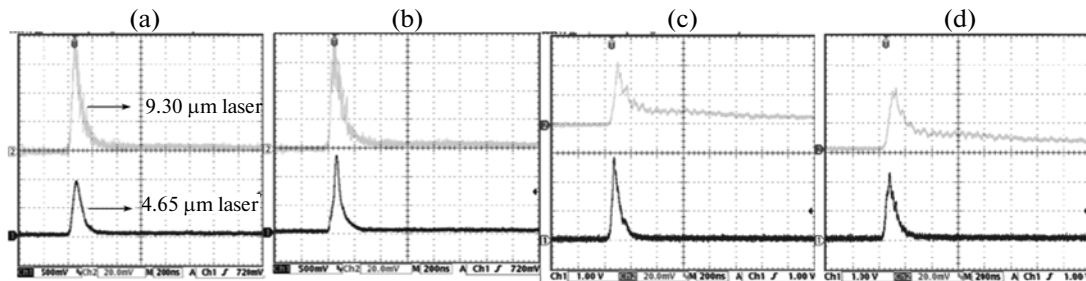


Fig. 6. Typical pulse shapes of CO<sub>2</sub> laser and the synchronous SHG laser.

power SHG laser is obtained. Kept the fundamental-laser pulse energy of 1.0 J and changed the repetition-rate, the maximum output power of 20.3 W SHG laser is obtained at 250 Hz, with a power conversion efficiency of 1.4%. According to the energy conversion efficiency of 3.4% in Fig. 4, the dependence of output power on repetition-rate can be calculated, as shown by the dashed in Fig. 5. However, the output power at every repetition-rate is lower than the calculative value. Furthermore, increased from 50 to 250 Hz, the power conversion efficiency is reduced from 2.1 to 1.4%, attributing to the increase of temperature gradient in ZnGeP<sub>2</sub> crystals. Nevertheless, no output saturation is observed at 250 Hz, and the output power can be further enhanced if temperature gradient was eliminated.

In addition, the SHG conversion is influenced by the irradiating intensity of fundamental-laser. Different laser intensity is produced at different time, resulting in different conversion efficiency. As shown in Fig. 6, typical pulse shapes of fundamental-laser and the synchronous SHG laser are recorded. It can be seen that the pulse width of SHG laser is near or narrower than that of fundamental-laser. In Fig. 6a, two pulse widths are almost same. In other three cases, the

pulse width of SHG laser is narrower than that of fundamental-laser. At the spike pulse, the conversion efficiency is much higher, but the tail pulse contributes less to SHG laser.

#### 4. CONCLUSIONS

High power SHG of TEA CO<sub>2</sub> laser is presented. Experiments approve that the SHG laser power cannot be enhanced evidently by using nonlinear crystals in serial or parallel fashion, but it can be realized by a parallel array. It is also found that ZnGeP<sub>2</sub> can withstand higher 9.3 μm laser intensity than AgGaSe<sub>2</sub>. By using a parallel array of ZnGeP<sub>2</sub> crystals, the maximum SHG output energy of 197 mJ is obtained in single-pulsed operation, with an energy conversion efficiency of 3.4%. In repetition-rate operation, the maximum power of 20.3 W 4.65 μm laser is obtained at 250 Hz, with a power conversion efficiency of 1.4%. To the best of our knowledge, it is the highest output power for SHG of CO<sub>2</sub> laser by far.

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