Pulsed CH$_3$OH terahertz laser radiation pumped by 9P(36) CO$_2$ lasers

Jiu Zhi-Xian $^1$, Zuo Du-Luo $^2$, Miao Liang $^2$, and Cheng Zu-Hai $^2$

1. Department of Mathematics and Physics, Wuhan Polytechnic University, Wuhan 430023 China

2. Wuhan National Laboratory for Optoelectronics, College of Optoelectronic Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074 China

E-mail: jiuzhixian@163.com

Abstract An efficient pulsed CH$_3$OH terahertz (THz) laser pumped by a TEA CO$_2$ laser was investigated experimentally. A simple terahertz cavity and a TEA CO$_2$ laser for the optically pumped THz radiation were studied experimentally. To improve THz laser energy and photon conversion efficiency, two different TEA CO$_2$ lasers were developed to pump CH$_3$OH. When CH$_3$OH was pumped by the 9P(36) line with different powers of the CO$_2$ laser, the generation of terahertz radiation with energy as high as 0.307mJ and 23.75mJ were obtained, respectively. The corresponding photon conversion efficiencies were 0.29% and 2.4%. The photon conversion efficiency increases by a factor of about 8. Meanwhile, higher peak power of pump laser effectively improves the photon conversion efficiency. And the optimum THz laser pressure increases with narrower pulse width of pump laser because of increasing absorptive gases molecules of CH$_3$OH with higher peak power of pump laser.

1. Introduction

With the development of the applications of terahertz (THz) (30 μm ~ 3 mm) laser, the THz laser source technology is becoming increasingly important. Since T. Y. Chang et. al. obtained THz laser by using a CO$_2$ laser to pump CH$_3$OH and other molecules in 1970 [1], methanol has become the THz laser-active molecule with the richest spectral lines. Just at that time, TEA CO$_2$ lasers have been employed as the optical pumping sources generally. The most powerful and efficient THz laser line was the 118.8 μm of CH$_3$OH pumped by the 9P(36) CO$_2$ laser line [2]. Most research has been dedicated on the experimental apparatus of THz laser pumped by CW CO$_2$ lasers or chopped CO$_2$ lasers with 9P(36) CO$_2$ laser line [3-10]. For example, THz power with the addition of buffer gases has been measured to be 1.25 W with 125 W of CO$_2$
pump power. The corresponding photon conversion efficiency has been 24.51% [6]. However, relatively less attention has been concentrated on energy of pulsed optically pumped CH$_3$OH THz lasers.

In this paper, a simple optically pumped THz laser system was designed. Two tunable TEA CO$_2$ lasers were developed to pump CH$_3$OH active medium, respectively. The corresponding generation of terahertz radiation with energy as high as 0.307mJ and 23.75mJ was obtained, with the photon conversion efficiency of 0.29% and 2.4%. All of these prove our experimental system is efficient.

2. Experimental system and results

The schematic of the experimental apparatus was consists of three main parts: the TEA CO$_2$ laser tuned by a 150 gmm$^{-1}$ grating, the THz cavity, and the detection system.

Both home-made TEA CO$_2$ laser served as the THz laser pump source, respectively. The first TEA CO$_2$ laser was that the discharge chamber was terminated with an antireflection-coated GaAs window and a semitransparent-coated GaAs window on both sides spacing 1.4 m apart. Dispersive element was a metallic reflective grating (150 gmm$^{-1}$) mounted on a rotating platform and blazed at 10.6 µm. Two flat gold-plated Si mirrors and a GaAs lens with focal length 2 m were used to transmit the output CO$_2$ laser radiation and compress its beam size. The second TEA CO$_2$ laser was that the discharge chamber was terminated with an antireflection-coated GaAs window and a ZnSe output coupler with only one surface antireflection-coated on both sides spacing 2.64 m apart. The dispersive element in the CO$_2$ laser cavity was a metallic reflective grating (150 gmm$^{-1}$) blazed at 10.6 µm and mounted on a rotating platform. The grating was placed in the position of 4.15 m away from the output coupler. Two flat gold-plated Si mirrors were used to transmit the output CO$_2$ laser radiation.

The THz cavity was composed of a 2 m long and 50 mm i. d. quartz tube, an input window and an output window. The input window of the THz laser was a 6 mm thick GaAs plate coated antireflection film, which fully transmits CO$_2$ laser. The output window was a 2 mm thick crystal quartz. It is opaque to CO$_2$ laser, and transparent THz radiation, which ensures detector only receives the latter. A tsurupica lens of 180 mm focal length (Microtech Instruments) is used for focusing the THz emission. It’s highly transparent to THz emission. The THz pulse energy was detected by a high responsive pyroelectric detector (Spectrum Detector SPJ-A-8-OB) which can detect an energy range from 60 nJ to 600 µJ with a wavelength range from 0.1 to 1000 µm. Finally, the signal was recorded by a digital oscillograph (Agilent DSO7034A) with a 350 MHz bandwidth.

The photon conversion efficiency is one of the conversion efficiency of the THz laser. The photon conversion efficiency is $\eta = (E_T/E_p)(2\lambda_T/\lambda_p)$, where $E_T$ ($E_p$) is THz output energy (CO$_2$ laser pump energy) and $\lambda_T$ ($\lambda_p$) is THz wavelength (CO$_2$ laser pump wavelength). $\lambda_p/(2\lambda_T)$ is the energy conversion efficiency in quantum limit [9, 11].

When the first TEA CO$_2$ laser served as the THz laser pump source, the pressure ratio of the gas mixture in the cavity of the TEA CO$_2$ laser was CO$_2$: N$_2$: He = 3: 1: 6 in the total pressure of about 50 kPa. The corresponding CO$_2$ energy was 2.6 J. The generation of terahertz radiation with energy of 307 µJ was obtained [12]. The photon conversion efficiency at 118.8 µm was 0.29%. The THz energy versus the CH$_3$OH pressure is shown in figure 1. An optimum THz laser pressure of about 260 Pa was observed. For low gas pressures, the
pumping pulses are sufficiently intense to saturate the pumping transition so that the output is limited only by the density of the gas. On the other hand, at higher pressures the laser transition becomes collision broadened, the upper laser level is no longer saturated so that the THz output eventually decreases to zero [13].

![Figure 1. The THz energy vs. the CH₃OH pressure pumped by the first TEA CO₂ laser.](image)

When the second TEA CO₂ laser served as the THz laser pump source, the pressure ratio of the gas mixture in the cavity of the TEA CO₂ laser was CO₂: N₂: He = 3: 1: 12.7 in the total pressure about 100 kPa. Shown in figure 2 are the laser pulse waveforms detected by a photo drag detector (Hamamatsu B749). The corresponding pump intensities and the full width at half-maximum (FWHM) were about 15.75MW/cm² and 42 ns, indicating that the pump intensity is greatly improved. The CO₂ energy was 24.2 J. The generation of terahertz radiation with energy of 23.75 mJ was obtained. The photon conversion efficiency was 2.4%. In figure 3, the optimum THz laser pressure of about 820 Pa increases by three-fold, compared with the former of 260 Pa.

In the results, the photon conversion efficiency increases by a factor of about 8. Meanwhile, The THz laser energy increases with the pump intensity, because the CH₃OH transition gain is dependent on the pump intensity [14]. Higher peak power of pump laser effectively improves the photon conversion efficiency. And the optimum THz laser pressure increases with narrower pulse width of pump laser because of increasing absorptive gases molecules of CH₃OH with higher peak power of pump laser [15].

3. Conclusions
The efficient pulsed CH₃OH terahertz (THz) laser pumped by two different TEA CO₂ lasers was studied experimentally. When CH₃OH was pumped by the 9P(36) line with different powers of the CO₂ laser, the generation of terahertz radiation with energy as high as 0.307mJ and 23.75mJ were obtained, respectively. The corresponding photon conversion efficiencies were 0.29% and 2.4%. The photon conversion efficiency increases by a factor of about 8. The
results prove that higher peak power of pump laser effectively improves THz laser energy and photon conversion efficiency. And the optimum THz laser pressure increases with narrower pulse width of pump laser because of increasing absorptive gases molecules of CH$_3$OH with higher peak power of pump laser.

![Figure 2](image)

**Figure 2.** The TEA CO$_2$ laser pulse temporal waveform.

![Figure 3](image)

**Figure 3.** The THz energy vs. the CH$_3$OH pressure pumped by the second TEA CO$_2$ laser.

**References**

[1] Chang T Y, Bridges T J and Buihardt E G 1970 *Appl. Phys. Lett.* 17 249
[2] Moruzzi G, Silos Moraes J C and Strumia F 1992
[3] Devoy D and Walker B 1984 *J. Phys. E: Sci. Instrum.* 17 1132
[4] Boscolo L 1988 *Appl. Opt.* 27 3325
[5] Mansfield D K, Horlbeck E, Bennett C L and Chouinard R 1985 *Int. J. IR & MM Waves.* 6 867
[6] Farhoomand J and Pickett H M 1987 *Int. J. IR & MM Waves.* **8** 441
[7] Pellemans H P M, Burghoorn J, Klaassen T O and Wenckebach W Th 1996 *Infra. Phys. & Tech.* **37** 635
[8] Heppner J, Weiss C O, Hubner U and Schinn G 1980 *IEEE J. Quantum Electron.* **16** 392
[9] Hodges D T, Foote F B and Reel R D 1976 *Appl. Phys. Lett.* **29** 662
[10] Bakos J S, Mandula K and Sorlei Z 1991 *IEEE J. Quantum Electron.* **27** 1094
[11] Marchetti S, Martinelli M, Simili R, Fantoni R and Giorgi M 2000 *Infra. Phys. & Tech.* **41** 197
[12] Jiu Z X, Zuo D L, Miao L, Cheng Z H and Qi C C 2010 *J. Infra. Milli. Terahz. Waves.* **31** 885
[13] Jiu Z X, Zuo D L, Miao L, Cheng Z H and Qi C C 2010 *Chin. Phys. Lett.* **27** 024211
[14] Heppnet J, Weiss C O, Hubner U and Schinn G 1980 *IEEE J. Quantum Electron.* **16** 392
[15] Bae J, Nozokido T and Shirai H 1989 *IEEE J. Quantum Electron.* **25** 1591