Theoretical and Experimental studies on CH$_3$OH THz Laser Pumped by Pulse Carbon Dioxide Laser

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Abstract. In this paper, according to the molecular structure and vibration mode of micro-asymmetric gyroscope CH$_3$OH molecule, dynamic process of optically pumped Terahertz laser is analyzed theoretically. The rate equation models based on three level systems are given according to the theory of typical laser rate equation. The output THz pulsed laser waveform is obtained by solving the rate equation model. An all-metal Terahertz laser pumped by RF waveguide carbon dioxide laser is designed with CH$_3$OH as its working gas. The pulsed Terahertz laser output is obtained. The waveform and repetition frequency of the optically pumped laser are measured in the experiments. The Terahertz laser designed does not need water cooling system. It also has the advantages of simple structure and small size.

1. Introduction
High repetition frequency pulse optically pumped Terahertz lasers can be applied in high-speed Terahertz imaging, Terahertz laser radar, medical diagnosis and other fields. The optically pumped THz laser can be continuous or pulse operation [1~4]. Currently, most pulse optically pumped THz lasers adopt TEA$\text{CO}_2$ laser as their pump sources. The wave lengths range is from mm to 40$\mu$m. Although the peak power of the THz lasers with TEA$\text{CO}_2$ laser as their pump source are high, its repetition frequencies are low (usually 10 times per second), which greatly limits their application. To solve this problem, J.Bae et al. developed the Q-switched carbon dioxide laser with gain and mechanical switching. When the repetition frequency is 1 KHz, the peak power of the output laser is more than 1KW. When pumped with 500Hz repetition frequency by this laser, the CH$_3$F gas can generate a 496$\mu$m laser, whose peak power is 6.5W and the pulse width is 100ns. R.C.Viscovini et. al. reported the CD$_3$OD THz laser pumped by adjustable wideband waveguide carbon dioxide laser, which can worked either continuously or with long pulses, whose the peak power is 100W, the repetition frequency ranges from 800Hz to 1200 Hz and the pulse widths are 30$\mu$s to 150$\mu$s [5]. In recent years, international researchers have been constantly explored new THz pump laser light spectrum lines and have some new findings of optical pumped THz laser spectral lines [6~8].

In China, researchers at Sun Yat-sen University and Huazhong University of Science and Technology have carried out a series of theoretical and experimental researches in THz lasers pumped by TEA$\text{CO}_2$ laser. The focus of their researches is the NH$_3$ gas THz laser. They have performed a series of profound and systematic researches on the mechanisms and the spectrum characteristics of the laser, as well as improving the working parameters and miniaturizing the size of the laser [9]. He
Zhihong et al. in Tianjin University studied the TEACO$_2$ laser pumped heavy water gas theoretically and experimentally and obtained the pulsed THz laser output of 385μm.

The Pump source of most THz lasers are the water cooling glass DC-exciting carbon dioxide laser or TEACO$_2$ laser, which are bulky and complicated [10, 11]. The highest pulse repetition frequency is about 1000Hz. Based on the result of the previous studies, we designed an all-metal RF waveguide carbon dioxide laser pumping THz laser, whose pulse repetition frequency can reach 5KHz. The pump source is the all-metal RF waveguide carbon dioxide laser. The THz generator is also of all-metal structure. Therefore, the laser can work with air cooling system instead of water cooling. Under the low pulsing frequencies, it can work without cooling. The size of the THz laser system can thus be designed smaller and the structure less complicated. The laser working gas is CH$_3$OH. THz laser wave form and the features of the output THz lasers under different repetition frequencies of the pumping laser are measured. These researches might serve as a reference to other relevant researches and as the foundation of later researches.

2. Dynamic Analysis for Pumping Progress

Up to now, the rate equation (R.E) models have been one of the main quantitative methods used to evaluate the power efficiency of pulse optically pumped Terahertz lasers [12~14].

Figure 1 shows the energy level transfer process of CH$_3$OH when pumped by CO$_2$ 9P(36). The THz laser generated by stimulated absorption transition (0.16,0.1.8) → (1.16,0.1.8) and stimulated emission transition (1.16,0.1.8) → (1.15,0.2.7) which are main parts of the energy transition is chosen as the main research object [15]. The three-energy-level theoretical model consists of (0.16,0.1.8),(1.16,0.1.8),(1.15,0.2.7) is shown in Figure 2.

![Fig.1 The relevant energy transition of CH$_3$OH pumped by CO$_2$ 9P(36)](image_url)

![Fig.2 The Simplified CH$_3$OH three-energy-level system model](image_url)

$E_1$ shows the ground state of the energy level. Under the action of pump light CO$_2$ 9P(36), particles in the $E_1$ ground state are pumped to $E_3$, the upper laser level, which will lead to the particles in $E_1$ exploded in number. Before the population inversion, particles in $E_3$ transit to $E_2$ and $E_1$, mainly by means of spontaneous transition and radiationless transition. However this transition is incredibly rare, so particles in $E_3$ has a longer residence time (a longer life span). If the pump speed
of particles to $E_3$ is high enough, there will easily form a distribution for population inversion between $E_2$ and $E_1$. Thus the stimulated radiation between $E_3$ and $E_2$ will take absolutely advantage, and further generate THz laser radiation with its wavelength of 118.8μm.

Through analysis of above experiment, we can establish the rate equation model of optically pumped CH$_3$OH molecular laser as follows:

$$\frac{dn_1}{dt} = n_1 \left( \frac{g_3}{g_1} \right) B_3 \rho_p(t) + \frac{1}{\hbar \nu} \left[ \left( \frac{g_3}{g_2} \right) n_2 - n_3 \right] \sigma_{32} (\nu, \nu_0) I_T - n_3 \omega_{31}$$

$$\frac{dn_2}{dt} = \frac{1}{\hbar \nu} \left[ n_3 - \left( \frac{g_3}{g_2} \right) n_2 \right] \sigma_{32} (\nu, \nu_0) I_T - n_2 \omega_{21}$$

$$\frac{dN_i}{dt} = \frac{1}{\hbar \nu} \left[ n_3 - \left( \frac{g_3}{g_2} \right) n_2 \right] \sigma_{32} (\nu, \nu_0) I_T - \frac{N_i}{\tau}$$

$$n_i + n_2 + n_3 = n$$

The $\rho_p(t)$ stands for energy density of the carbon dioxide laser pump light; $I_T$ stands for intensity of THz optical wave output; $n$ is the total number of per unit volume particles in the three-energy-level while $n_1$, $n_2$ and $n_3$ represent the particles per unit volume of the ground state $E_1$, the lower laser level $E_2$ and the upper laser level $E_3$. The $\sigma_{32}(\nu, \nu_0)$ represents the emission cross section of THz laser. $g_i = 2J_i + 1$ represents the statistical weight of each energy level and $N_i$ represents the density of photon in laser resonant.

3. Calculation Results

For convenience, pump pulse is some what similar to the Gaussian pulse on the basis of the experimental results, which can be expressed as:

$$\rho_p(t) = \rho_{p0} \exp \left[ -\left( t - t_0 \right)^2 / \sigma^2 \right] \quad 0 < t < 1.5 \mu s$$

Where $\rho_{p0}$ represents the photon density of the pump pulse peak and $\sigma$ is the width of pulse laser.

The parameters for the rate equations’ calculation are as follows:

| Physical Parameter | Sign | Value     | Unit  |
|--------------------|------|-----------|-------|
| Electric dipole moment | $\mu_3$ | $2.23 \times 10^{-30}$ | C·m   |
|                     | $\mu_1$ | $1.79 \times 10^{-30}$ | C·m   |
| THz spectrum line homogeneous broadening line width | $\Delta v_H$ | $2.8 \times 10^6$ | Hz |
| THz spectrum line Doppler broadening line width | $\Delta v_D$ | $7.9 \times 10^6$ | Hz |
| Upper energy level particle number before pumping | $n_3(0)$ | $0.00183n_0$ | m$^{-3}$ |
| Lower energy level particle number before pumping | $n_2(0)$ | $0.00264n_0$ | m$^{-3}$ |
| Each statistical weight level ratio | $g_3 / g_2$ | 1.065 | —     |
|                     | $g_3 / g_1$ | 1 | —     |
By using Matlab program, we have numerically solved the rate equations. Fig. 3 shows that the particle distribution is in accordance with Boltzmann distribution in thermal equilibrium. The particle number of lower energy level is more than the particles in upper energy level in the beginning. The particles of both the lower and upper energy level begin to increase slowly under the action of pump light. However, there is a rapid increase after 0.8µs. The THz laser output is obtained as a result of particles in upper energy level increase rapidly under the action of pump pulse and form the population inversion. Due to the stimulation, the particles in lower energy level also increase and reach to the peak after 1µs. Then the particles of the lower and upper energy level decrease gradually with pump light decreasing until it resorts to the particles distribution in the thermal equilibrium. From Fig. 4, we can get a conclusion that the output THz wave reaches its peak around 1.22µs and the decline stage after the peak is relevantly slow which is due to the relaxation from lower energy level to the ground state. In the pumping process, the relaxation of particles transiting from upper energy level to the lower level and from the lower level to the ground state makes some particles still gather in the lower energy level which will influence the population inversion and further reduce the speed of photon generation, thus makes the pulse wave relatively slow. From Fig. 5, we can get a conclusion that the output THz pulse wave has some time delay compared with the input pumping pulse wave. Its peak power is lower and pulse width is shorter.

Fig. 3 Time dependence curve of upper and lower laser level population

Fig. 4 The output THz pulse waveform
4. Experimental results

The THz laser consists of two parts: One is the pump source [16]; the other is the THz laser generator. The pump source is grate tuning all-metal RF waveguide carbon dioxide laser [17]. The CH$_3$OH gas is inflated into the THz laser through the air supply system. According to the references, the laser can output 118.8μm THz laser with the 9P (36) line pump laser [18]. In the beginning, the pump laser was operated in low repetition frequency (90Hz). The voltage on the piezoelectric ceramics was then adjusted, so as to tune the frequency of the pump laser and maximize the intensity of the THz output. The waveform and intensity of the generated THz were observed via oscilloscope. As was shown in Fig. 6, the THz laser resembled the triangular wave, the reason of which is that the band width of our detector is 300KHz, which is only enough to detect the laser signal and is not capable of showing the real waveform of the pulsed THz laser. Theoretical analyses above show that the output waveform of the THz laser should be similar to the pump laser, with only a lower intensity.

![Fig. 6 Measured THz laser output (5ms/div.)](image)

The repetition frequency of the pump laser can be as high as 5KHz. Theoretically, the frequency of the THz output should also be 5 KHz. Thus, we measured the waveform of the THz output while increasing the repetition frequency of the pump laser. In the experiment, the oscilloscope shows that the intensity of THz signal decreased as the repetition frequency increased. The obtained signal is shown in Fig.7 at the pump laser repetition frequency 200Hz. If further improving the repetition frequency of the pump laser to 300Hz, the signal would scarcely be observable. We believe the phenomenon is due to the band width of the detector being narrow. If a detector with high band width being adopted, the true waveform of THz output could be obtained.
5. Conclusion

In this paper, the dynamic process of CH\textsubscript{3}OH THz laser pumped by carbon dioxide laser is studied. Through the energy-level analysis of CH\textsubscript{3}OH, a three-level system model of energy level transition of CH\textsubscript{3}OH is set up. Based on the typical theory of laser rate equation, the rate equation of the three-level system model is also given. The dynamic process of pulse THz laser is analyzed theoretically. Through the numerical calculation, we have got the transitions of particles on the different energy levels.

On the experimental side, we have studied the THz laser pumped by all-metal Radio-frequency waveguide carbon dioxide laser with grating line selected. The whole laser including pump source and THz generator is all-metal structure. It has the advantages of a simple structure and small volume without water cooling. The laser can work either on continuous or pulse modes. In this experiment, we measured the parameters for the THz pulse laser.

On further experiment, the following points need to be modified or improved: (1) The CH\textsubscript{3}OH used in this experiment is common in the market with a purity of only 99.5%. The laser should have high efficiency by using spectroscopically pure CH\textsubscript{3}OH. (2) Presently, we use manually control the voltage on the piezoelectric ceramic to change the pump laser frequency to get the strongest output of THz laser, which result in a poor stability of the output of laser. On further experiment, we consider employing an automatic control system to improve the output stability. (3) Limited by the detector bandwidth, the true THz pulse laser wave form couldn’t be obtained, and it is also not able to measure THz laser at high-repetition rate. So on further study we will consider purchasing a broadband detector or cooperate with qualified organizations.

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