A NEW FIR LINE OF CH_3OH *

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Abstract Using the CH_3OH molecular energy levels data base management program, it was deduced that when a CH_3OH laser is pumped by CO_2-9P(16) line, there should be a FIR laser line of 918μm wavelength which has not been reported before, corresponding to a transition in a ground-state reversed three-level energy system. The spectrum of this new line was calculated by solving the density matrix equations. Experimentally, the CH_3OH FIR laser line of 918μm wavelength was obtained by pumping with a TEA-CO_2 laser. The experimental results were in good agreement with the theoretical calculations.

Key words CH_3OH FIR laser, new FIR line.

INTRODUCTION

CH_3OH is known as the most prolific active medium for FIR laser. The high yield of lasing transitions of CH_3OH is due to that 1) the strong absorption of CO-streching band of CH_3OH molecule is in coincidence with CO_2 laser lines of 9-10μm R and P branches; 2) the concentration of its rotational spectrum is high; 3) the presence of a fairly large permanent electric dipole moment makes the selection rules much less restrictive than for symmetric top molecules[1].

CH_3OH FIR output could be obtained by different methods. Most of them were found by using CW optically pumped lasers, and most FIR sample tubes are longer than 1m. The mini-OPFIRL (Optically Pumped Far-infrared Laser) is a new technology developed in recent year, and CH_3OH FIR laser lines have been obtained by pumping a 10cm or 20cm long miniature CH_3OH OPFIRL in our lab[2].

The gain coefficient of ground-state transition in reversed three-level energy system could be derived by solving semi-classical density matrix[3]. Based on this method, calculations on CH_3OH molecular spectra were done by computer.

The calculation showed that there should be a FIR laser line of 918μm wavelength when pumped by CO_2-9P(16) line, with $-0.00224cm^{-1}$ frequency offset, which is corresponding to a transition in a ground-state reversed three-level energy system. The spectra

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showed double peaks and the output power was concerned with operating gas pressure.

Experimentally, a TEA-CO_2 laser was used for the pumping source, the $CO_2$9P(16) line was selected to pump a 20cm or 10cm long miniature cavity CH_3OH OPFIRL, and the FIR line of 918μm wavelength was obtained successfully, and double peaks were observed. The experimental result was in good agreement with the theoretical calculations.

I. THEORETICAL CONSIDERATION

1.1 The Gain coefficient (G)

An OPFIRL could be looked upon as: when pumped by the IR laser source, the medium molecules in sample tube formed an active molecule system and the feedback of the optical resonant cavity amplified the FIR signal of local background noise and made FIR laser output. In the three-level system approximation, the FIR signal gain coefficient G_s and IR pumping signal absorption coefficient G_p were expressed as follows:

$$G_s = \frac{4\pi N_v \mu_s^2 \omega_s T_a}{\varepsilon_0 \eta C B h} \mathrm{Im}(P_{12}) - \alpha_s \tag{1}$$

$$G_p = \frac{4\pi N_v \mu_p^2 \omega_p T_b}{\varepsilon_0 \eta C B_p h} \mathrm{Im}(P_{13}) - \alpha_p \tag{2}$$

where N_v is the population density of molecules joining in the laser action, $\eta = \sqrt{\varepsilon/\varepsilon_0}$ is the refractive index of the laser medium, C is the light speed, h is the Planck's constant.

1.2 Energy-level structure

A data base management program on the basis of CH_3OH molecular energy level table[2] was compiled. Given the selection rules of the vibrational absorption transition of CO_2-9P(16) line and corresponding FIR pure rotational transition, and the frequency offset was limited to $\pm 0.0030 cm^{-1}$ range, the computer gave the result of a FIR laser transition with 918μm wavelength (corresponding wavenumber is $10.89323 cm^{-1}$)[4]:

$$A, gr, 0, 0; 10 \rightarrow co, 0, 0; 11$$
$$gr, 0, 1; 10 \rightarrow gr, 0, 0; 10$$

with $-0.00224 cm^{-1}$ frequency offset, shown as Fig. 1. Figure 1 shows that this transition belongs to the transition of reversed ground-state three-level energy system, which is more difficult to occur and less efficient.

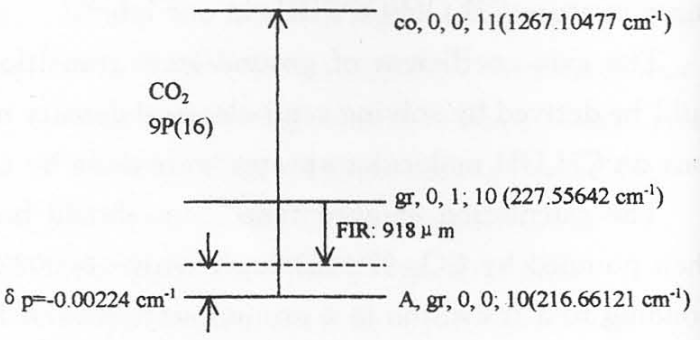

Fig. 1 The transition of 918μm wavelength
图1 918μm能级跃迁图

1.3 Operating parameters

(1) Complex transitional electric dipole moment (μ)

According to the selection rules, the vibrational absorption transition of CO_2-9P(16) belongs to *a*-type, *R*-branch transition, and the pure rotational transition of 918μm line belongs to *b*-type, *Q*-branch transition. The complex transitional electric dipole moment could be calculated[5]

(2) Popular density of molecules joining in the laser action (N_v)

The popular density of molecules joining in the laser action can be expressed as:

$$N_v = \frac{N}{V} \cdot f \quad . \tag{3}$$

N/V is the popular density of medium molecules, and

$$f = \sum_{j=1}^{3} \exp(-E_j/KT) / \sum_{j=1}^{\infty} \exp(-E_j/KT) \tag{4}$$

In Eq. (4), E_j is the energy of energy level. According to the CH_3OH data base program, we have:

$$\sum_{j=1}^{\infty} \exp(-E_j/KT) = 192.35461030$$

(3) Relaxation time T_a, T_b and initial FIR intensity I_{s0}

The relaxation time for three-level energy system mode of CH_3OH molecules at room temperature (300K) was given by[6]:

$$T_a = 2.098 \times 10^{-6}/TP$$

$$T_b = 1.2983 \times 10^{-5}/TP$$

where T_a is the transverse relaxation time, T_b is the longitudinal relaxation time, T is the temperature, and P is the gas pressure.

The initial FIR signal power intensity was the FIR part of Gaussian noise, which could be calculated from the blackbody radiation equation. For the sake of simplicity, the initial FIR power intensity was taken as $I_{s0}=1.0\times10^{-13}\mathrm{W}\cdot\mathrm{cm}^{-2}$

1.4 Calculated results

The whole calculation could be done by computer when all operating parameters were given. In order to calculate the spectra of FIR laser, the central frequency of FIR signals was taken as zero point, and the output power intensity was calculated one point by one point within a certain frequency offset range (−2GHz～+2GHz, for example). Figure 2 shows the spectrum of 918μm line in respect to 10cm sample tube, pumped by CO_2-9P(16) IR laser line. The pumping power intensity is $1.0\times10^6\mathrm{W/cm}^2$, and the operating gas pressure is 23 Torr.

(1) AC-Stark effect:

Figure 2 shows double peaks of AC-Stark splitting. It was caused by the strong pumping electric field, which caused the CH_3OH molecular energy levels to split and trans-

mitted into two lines with very closed frequency.

(2) Raman process interaction

Figure 3 shows the spectra of CH_3OH 918μm line with 20cm long sample tube in respect to 3.0, 13.0, 25.0, 42.0 Torr operating gas pressure. The pumping source is CO_2-9P(16) IR line, and the pumping power intensity is $3.0\times10^6 W/cm^2$. Figure 3 shows that, when the gas pressure is lower(P=3.0Torr), the two AC-Stark peaks are parted, there not exists the Raman process interaction, the FIR output power is low (curve A); The spectra of each single peak become broad with the rising of gas pressure(P=13.0Torr), the two peaks of AC-Stark become closer and overlapped (curve B); When the gas pressure is raised higher(P=25.0Torr), the Raman interaction becomes stronger and the FIR output higher(curve C); But if the gas pressure is raised further(P=42.0Torr), the competition of Raman process becomes stronger, resulting in the weaker peak totally absorbed by the stronger one, and only one peak could be seen and the FIR output power becomes lower(curve D).

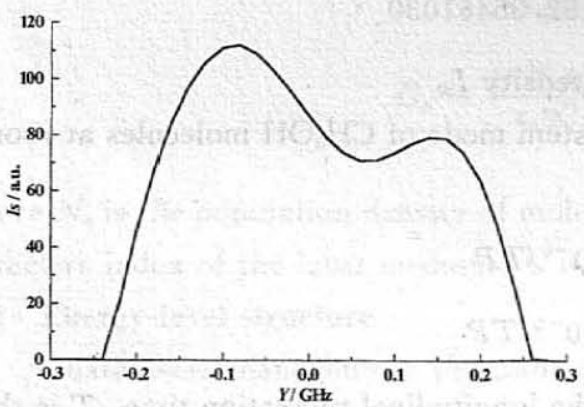

Fig. 2 The spectrum of 918μm line
图 2 918μm 谱线频谱特性图

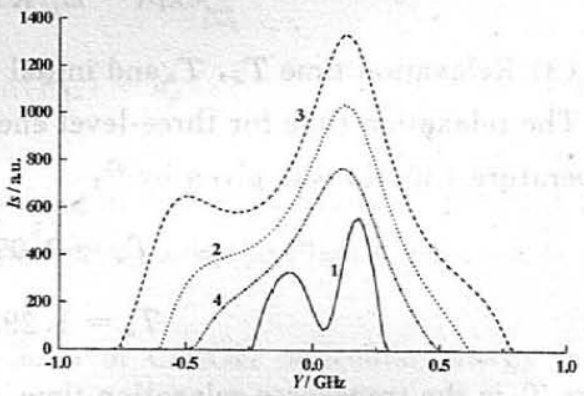

Fig. 3 Raman process interaction of 918μm line
图 3 918μm 谱线喇曼过程的相互作用

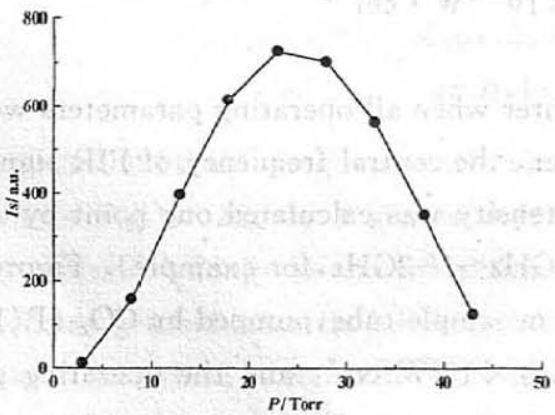

Fig. 4 Relationship of FIR output intensity *vs.* gas pressure
图 4 工作气压与远红外激光输出强度关系曲线

(3) The optimum operating gas pressure

FIR cavity laser output intensity was affected by many factors: the operating gas pressure, the reflected coefficient of input/output grid, the length of the sample tube, the environment temperature, etc., and the effect of the operating gas temperature was the most obvious one, and easy to observe in an experiment.

Figure 4 shows the relationship curves of FIR output intensity versus gas pres-

sure, where the pumping source is CO_2-9P(16) IR laser, the pumping power intensity is $3.0\times10^6 W/cm^2$, the FIR output is CH_3OH 918μm line, the FIR laser is 20cm long. Figure 4 seems that there existed an optimum operating gas pressure with which the FIR output power reached the maximum. When the FIR sample tube is 20cm long, the optimum gas pressure with respect to 918μm FIR laser is about 23 Torr, which is higher than that of another FIR laser line of 570μm wavelength pumped by CO_2-9P(16) line with the same sample tube length[7]. It is because the former belongs to the ground-state reversed three-level energy system, whose transmission is less efficient than that of the latter, reversed excited three-level energy system, and the operating gas pressure must be raised to increase the popular density of molecules joining in the laser action N_v.

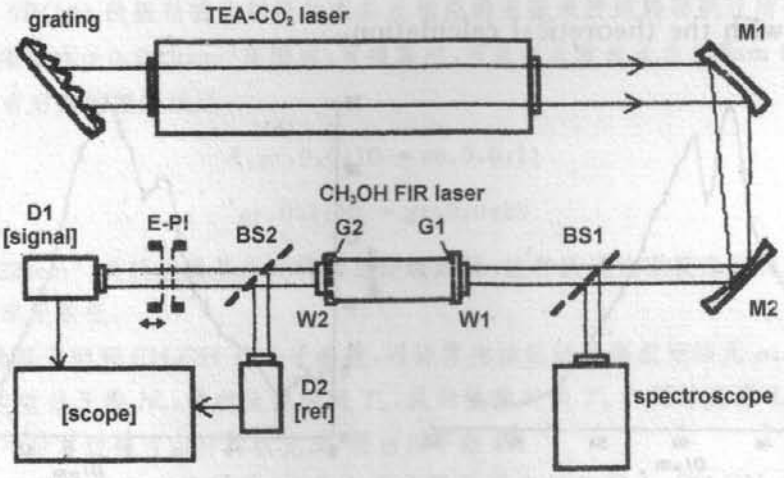

Fig. 5 Experimental setup
图 5　实验装置

Ⅱ. Experimental Results

The experimental setup consisted of a TEA-CO_2 laser as a pump laser, a FIR cavity laser, and measurement apparatus, shown as Fig. 5.

The TEA-CO_2 laser was tuned by a reflection grating, the mixed working gas was in a rate of $CO_2:N_2:He=2:5:2$, with total gas pressure 24KPa, concerning to 9P(16) line, the laser pulse width was about 150ns, the pulse energy was about 570mJ. In order to avoid the interference from high voltage discharge, the TEA-CO_2 laser and its power supply were shielded in a metallic mesh shelter. The infrared laser pump beam went out and was reflected by mirrors M1 and M2 into the FIR laser. The FIR laser was composed of a Pyres sample tube, which is 10cm or 20cm long, with 32cm inner diameter. The input window was a ZnSe plate, which is transparent to infrared radiation, and the output window was a thick teflon plate, which is transparent to FIR but opaque to IR radiation.

The FIR laser beam went out through a F-P interferometer, and the interferogram signal was picked up by a pyroelectric detector D1, and a reference signal from detector D2

was used to compensate the fluctuation of FIR intensity. When pumped by CO_2-9P(16) line, the FIR emissions were obtained, then measured by a F-P interferometer. By readjusting the experimental parameters appropriately, the FIR line of 918μm wavelength was observed, and the output power of a single pulse was about 2μJ, measured by a power meter.

Figure 6 shows the interferogram from 10cm and 20cm long CH_3OH lasers with wavelength 918μm. The figure shows double peaks in the interferogram, which is caused by AC-Stark effect. The strong pumping electric field makes the CH_3OH molecules energy level split into two levels, and two lines transmitted with very close frequency.

Comparing Fig. 6 with Fig. 2, we see that the interferogram is very similar to the theoretical spectra i. e. both of them have double peaks, and the experimental results are in good agreement with the theoretical calculation.

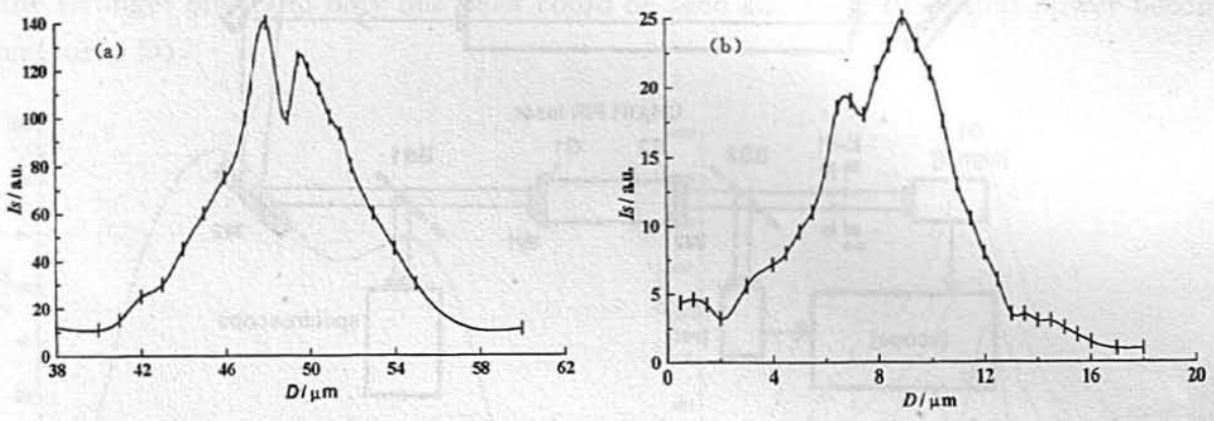

Fig. 6 F-P interferogram of 918μm line
(a) FIR laser length L=20cm (b) FIR laser length L=10cm
图 6 918μm 谱线的 F-P 干涉特性图
(a) 远红外激光管长 L=20cm (b) 远红外激光管长 L=10cm

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甲醇远红外激光的一条新谱线*

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摘要 甲醇(CH_3OH)分子是常用的FIR激活介质，具有很特别的光谱特征．而小型光泵脉冲亚毫米波激光器是近年才发展起来的新技术，作者已经成功地研制出管长为10～20cm的CH_3OH FIR激光器，并获得谱线．

本文利用CH_3OH分子能级表编制成数据库管理程序，对光泵CH_3OH亚毫米波激光进行计算．代入CO_2-9P(16)线振动吸收跃迁的条件及相应的亚毫米波纯转动跃迁所受的跃迁选择条件，将泵频偏限制在$\pm 0.0030cm^{-1}$范围内，可推算出，当亚毫米波波长为918μm（对应波数$10.89323cm^{-1}$）时，有对应的能级跃迁：

$$A,gr,0,0;10 \rightarrow co,0,0;11$$

$$gr,0,1;10 \rightarrow gr,0,0;10$$

泵频偏为$-0.00224cm^{-1}$，其跃迁属基态反转的三能级跃迁，这种跃迁比激发态反转的三能级跃迁更不易得到，效率比较低．

根据跃迁选择定则和CH_3OH的分子参数，可计算出该跃迁的偶极矩阵元μ，单位体积内参与激光过程的有效分子数N_0，横向弛豫时间T_a，纵向弛豫时间T_b和起始光强I_{s0}等参数．选定工作参数后，整个计算过程可由计算机完成．得出以下结果：

1．亚毫米波激射呈现双峰形状，这是由于在强泵浦电场作用下，CH_3OH分子能级出现AC-Stark分裂，从而发射出两支频率十分接近的谱线．

2．随着工作气压的逐渐升高，喇曼过程的相互作用逐渐增强，亚毫米波的输出功率逐渐增加至一最大值，但随着工作气压的进一步升高，喇曼过程的竞争加强，导致输出强度较低的小峰被输出强度较大的大峰完全吸收，输出功率反而下降，其结果是AC-Stark的双峰消失，只呈现单峰形状．

3．存在着一个最佳气压$P(opt)$，在该气压下亚毫米波激光器的输出功率达到最大．

实验中，当用CO_2 9P(16)线泵浦管长为20cm，10cm的小型脉冲CH_3OH分子亚毫米波激光器时，可以观察到亚毫米波谱线，当调节泵浦功率为$3.0\times10^6W/cm^2$，工作气压约为20Torr时，测量到波长为918μm的亚毫米波谱线．用功率计测量其单个脉冲能量约为2μJ．实验干涉图与理论计算的频谱特性图十分相似，呈明显的双峰结构，这是由于强泵浦电场作用下CH_3OH能级受到单色光强扰动而产生AC-Stark分裂的结果．结果表明理论计算与实验数据一致．

关键词 CH_3OH远红外激光，远红外新谱线．

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