

FREQUENCY MEASUREMENTS OF 3 TO 11 THZ LASER LINES OF CH₃OH[†]

S. C. Zerbetto,* L. R. Zink,^a K. M. Evenson, and E. C. C. Vasconcellos*

*National Institute of Standards and Technology
Time and Frequency Division
325 Broadway, Boulder, CO 80303-3328 USA*

*^aCooperative Institute for Research in Environmental Sciences
University of Colorado and NOAA
Boulder, CO 80303-3328 USA*

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* Permanent address:

Instituto de Física "Gleb Wataghin," Departamento de Eletrônica Quântica, UNICAMP, 13083-970 Campinas, SP, Brazil.

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ABSTRACT

We measured the frequencies of 12 far-infrared laser lines generated in a high-frequency Fabry-Perot laser cavity containing methanol, pumped by a CO₂ laser. The frequencies are in the range 2.8 to 11.4 THz (105.4 to 26.2 μm). Ten of the measured frequencies are higher than 7 THz, and help to fill the laser frequency gaps in this region. Five of the measured lines are new. The 11.4 THz line has the highest frequency of an optically pumped laser ever measured with the CO₂ laser heterodyne technique.

INTRODUCTION

Methanol (CH₃OH) is the most important far-infrared (FIR) active medium, generating about 600 laser lines at wavelengths from 25.4 to 1223.7 μm [1,2]. Approximately 31% of those lines have frequencies higher than 3 THz. Recently, an additional 16 high frequency laser lines were reported [3]. The frequencies of eight of those lines were measured, and tentative assignments are being made [4]. We report frequency measurements of 12 FIR laser lines from methanol, including two lines from reference 3 and five new lines. The frequencies are in the range 2.8 to 11.4 THz (105.4 to 26.2 μm) and 10 lines have frequencies higher than 7 THz.

EXPERIMENTAL

Our FIR laser has a low-loss Fabry-Perot cavity with variable coupling [3]. It has less than 0.5% diffraction loss at frequencies above 2 THz and uses a folded confocal geometry. The cavity consists of a 36 mm diameter, 2 m long Pyrex tube closed at one end by a fixed, gold-coated flat mirror, and at the other end by a 4 m radius-of-curvature, gold-coated concave mirror. The concave mirror, attached to a micrometer, is movable to tune the cavity. A 6 mm diameter, 45° copper mirror is situated on the side of the cavity and couples the FIR power out through a silicon window at Brewster's angle on the opposite side. We vary the output coupling by moving this mirror into the cavity mode. A parabolic mirror focuses the FIR radiation onto a metal-insulator-metal (MIM), point-contact diode which was used as both a detector and heterodyne mixer.

The CO₂ pump radiation is focused into the laser with a 5 m radius-of-curvature concave mirror, 2.5 m from the laser. The pump power enters the cavity through a 5 mm diameter hole 15 mm above the laser axis in the fixed flat mirror; thus the hole is outside the FIR mode, reducing

losses. The CO₂ radiation hits the center of the curved mirror at the opposite end of the laser and is reflected back to the flat mirror at an angle yielding V-type pumping [3]. We rotated the polarization of the CO₂ pump radiation to observe either perpendicular or parallel FIR laser lines.

We determined the wavelengths of the new lines by tuning the FIR laser cavity with the movable mirror and measuring the mirror displacement for 20 wavelengths of the laser line. This value is accurate within one part in 10³. This accuracy is very important when measuring frequencies of short-wavelength laser lines.

We measured the laser frequencies by mixing the radiations from the FIR laser, two frequency-stabilized CO₂ lasers, and a microwave source on the MIM diode [5]. A beat note is generated by the diode, and the unknown laser frequency ν_{FIR} is calculated by

$$\nu_{\text{FIR}} = |n_1\nu_{\text{L1}} - n_2\nu_{\text{L2}}| \pm m\nu_{\mu\text{w}} \pm \nu_{\text{beat}}, \quad (1)$$

where ν_{L1} and ν_{L2} are the CO₂ laser frequencies, $\nu_{\mu\text{w}}$ is the frequency of the microwave source, ν_{beat} is the beat frequency, and the integers n_1 , n_2 , and m are the harmonic orders. The CO₂ frequencies and harmonic orders are chosen to yield an RF beat frequency of less than 1.5 GHz. The beat note is amplified and displayed on a spectrum analyzer, using a peak-hold feature that records the signal maximum as the FIR laser is tuned over its gain curve. The center frequency of the recording is measured using a marker frequency from a synthesizer.

The intensity of the beat note decreases as the harmonic orders increase, so we usually choose $n_1=n_2=m=1$. However, when measuring frequencies higher than 6 THz, as in the case of most of the lines measured in this work, this was not possible. We used $n_1=n_2=3$ and $m=1$ to measure the 11.4 THz (26.2 μm) line.

We also measured the pump offsets for nine of the lines by first maximizing the FIR output by tuning the pump laser and then by mixing the pump laser radiation with the radiation from a frequency-stabilized reference laser in the MIM diode. The resultant beat frequency is then amplified, displayed on a spectrum analyzer, and measured with a marker frequency.

RESULTS AND CONCLUSIONS

Table I lists the frequency measurements. The offsets, relative intensities, relative polarizations to the pump laser, and operating pressures are also listed for most of the lines. All lines are pumped by regular lines of the 9R and 9P branches of the CO₂ laser. Three of the new lines are pumped by 9R(02), and two are pumped by 9P(34). The 11.4 and 10.9 THz lines are the highest reported frequencies of an optically pumped far-infrared laser measured with the CO₂ laser heterodyne technique [6]. These laser lines help fill the frequency gap in the 3 to 12 THz region and will be useful sources for atomic and molecular spectroscopy.

TABLE I: High-frequency far-infrared laser lines in optically pumped CH₃OH.

| CO ₂ Pump Line | λ^a μm | Frequency ^b MHz | Offset ^c MHz | Rel. Int. ^d | Rel. Pol. | Pressure Pa(mTorr) | Ref. |
|---------------------------|------------------------------|-------------------------------|----------------------------|------------------------|-----------|-----------------------|------|
| 9R(16) | 33.524 | 8 942 749.1 | | 0.5 | ⊥ | 33(250) | 7 |
| | 36.667 | 8 176 074.8 | | 1.6 | // | 27(200) | 7 |
| 9R(02)' | 35.058 | 8 551 428.9 | -13 | 0.3 | // | 20(150) | new |
| | 94.875 | 3 159 866.8 | -13 | 6.0 | // | 25(190) | 7 |
| 9R(02)'' | 36.392 | 8 237 945.6 | +25 | 0.8 | // | 25(190) | new |
| | 42.267 | 7 092 794.1 | +25 | 0.6 | ⊥ | 20(150) | new |
| | 105.438 | 2 843 293.3 | +25 | 5.0 | // | 24(180) | 7 |
| 9P(16) | 41.871 | 7 159 895.3 | | 1.0 | ⊥ | 21(160) | 7 |
| 9P(34) | 26.238 | 11 426 080.4 | -37 | 0.2 | // | 25(190) | 3 |
| | 27.386 | 10 946 757.0 | -37 | 0.1 | ⊥ | 25(190) | 3 |
| | 38.333 | 7 820 764.9 | -37 | 0.2 | // | 24(180) | new |
| | 40.802 | 7 347 410.8 | -37 | 0.1 | ⊥ | 25(190) | new |

a. Calculated from the measured frequency with $c=299\,792\,458$ m/s.

b. Estimated 1σ uncertainty in the reproducibility of the FIR laser frequency is $\Delta\nu/\nu = 2 \times 10^{-7}$.

c. Estimated uncertainty in the offset measurement is 2 MHz.

d. In this laser the 119 μm CH₃OH line has a relative intensity of 10. ' and '' denote different offsets.

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