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Frequencies of Optically Pumped Sub-Doppler Far-Infrared Laser Lines of Methanol

Edjar M. Telles,[1] Lyndon R. Zink, and Kenneth M. Evenson

Time and Frequency Division, National Institute of Standards and Technology-NIST, 325 Broadway, Boulder, Colorado 80303-3328

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Sub-Doppler signals from the gain curves of four lines of a transversely pumped far-infrared methanol laser were used to stabilize the frequencies of the laser. The frequencies of four of the strongest FIR laser lines (70, 119, 123, and 570 μm) were measured. Lasers locked to these lines have a frequency reproducibility of a few parts in 10^8 and an uncertainty of less than 30 kHz. © 1998 Academic Press

I. INTRODUCTION

We were able to improve the frequency reproducibility of four optically pumped far infrared (FIR) laser lines of methanol by locking to sub-Doppler Lamb dips at the center of each gain curve. The Lamb-dip signal was obtained by operating the laser at lower than usual pressure and transversely pumping the gain medium with CO_2 laser radiation.

Optically pumped FIR lasers are the most versatile, powerful, coherent sources in the FIR region. Due to good frequency reproducibility (few parts in 10^7) and stability (few parts in 10^9) they have been used in high-resolution spectroscopic studies in, for example, laser magnetic resonance (LMR) spectroscopy, active medium spectroscopy, tunable FIR spectroscopy, and plasma diagnostics. They are also used as local oscillators for heterodyne receivers in radio astronomy.

The frequency uncertainty is determined mainly by the resettability of the optically pumped FIR lasers to the center of their Doppler-broadened (1–10 MHZ) gain curve. In previous works, the frequency resettability was limited by the locking of the FIR laser line to the top of the Doppler-broadened gain curve (*2, 3*). Our recent measurements have improved the frequency accuracy by about a factor of 20 and make them much more accurate frequency and wavelength sources for spectroscopic applications.

II. EXPERIMENTAL APPARATUS

Figure 1 shows the experimental setup. The FIR laser uses a nearly confocal, 1.40-m-long Fabry-Perot cavity mechanically maintained by four Invar bars and four aluminum blocks. The laser cavity is divided into two regions (gain and absorption) by a polypropylene beam splitter 8 μm thick set at the Brewster angle. This beam splitter is also used as a partial reflector to couple out the FIR power. It can be rotated a few degrees near the Brewster angle to maximize the output power.

The gain region filled with gaseous CH_3OH is formed by an internally polished copper tube with a 2.5-cm inner diameter that is 80 cm long. However, for wavelengths longer than 200 μm, a second, larger copper tube of 5-cm inner diameter is used to minimize the diffraction loss and obtain lasing at longer wavelengths. The smaller tube has a better overlap between the short FIR wavelengths and several times more power is produced on the 63-μm line of $^{13}CH_3OH$ by using the smaller tube. The pump radiation is nearly perpendicular to the laser axis to prevent burning the beam splitter and to minimize transferred Doppler-pulling effects on the FIR laser line (*5*) (we will call this dispersion pulling). The CO_2 radiation enters the copper tube near one end through a ZnSe window at an angle of 75° with respect to the cavity axis and bounces back and forth down the pump tube. The CO_2 pump laser is of the same design as that recently developed in our laboratory (*4*) in which 275 lines can be grating selected from all 12 bands of the CO_2 laser.

The absorption region is contained in a Pyrex tube with a 7-cm inner diameter and has a length of 40 cm. However, no absorption gas was used in these experiments. Various gases can be introduced into this absorption cell to search for inverted Lamb dips on the laser output. The two regions are connected to avoid pressure damage to the beam splitter during pump-out. The cavity mirrors are gold coated; one is coupled to a micrometer for coarse tuning and the other to a PZT transducer for fine tuning. The FIR frequency of the laser is modulated by modulating the dc voltage applied to the PZT.

III. THE MEASUREMENTS

We studied four of the stronger FIR laser lines in methanol at wavelengths of 70, 119, 123, and 570 μm. These lines have been used in many other spectroscopy experiments. First, the

[1] Postdoctoral fellow from CNPq-Brasilia-Brasil.

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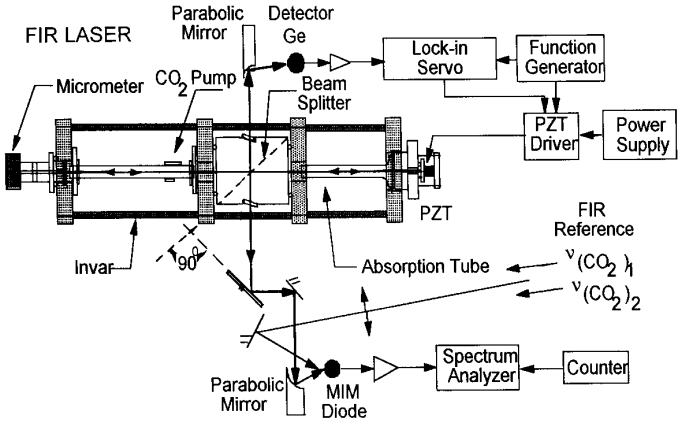

FIG. 1. Experimental setup to stabilize and frequency measure the FIR laser lines.

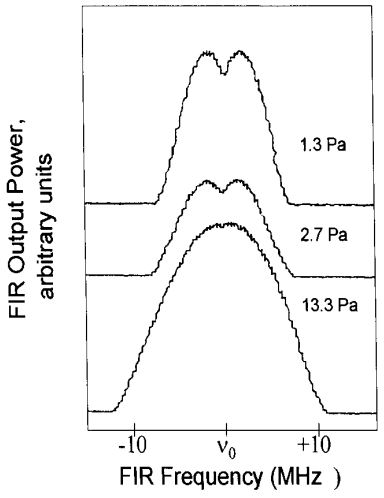

FIG. 2. Lamb dip observed on the gain curve of the 119-μm line.

laser power was maximized, then the methanol pressure was reduced to 1.3–6.7 Pa (10–50 mTorr) to produce a Lamb dips at the centers of the gain curves (see Fig. 2). The Lamb dip in the output power of the laser was detected with a liquid-helium-cooled germanium bolometer. The signal was amplified and sent to a lock-in amplifier for phase-sensitive detection at the modulation frequency. We modulated the FIR laser frequency at 600 Hz with a width of about 200 kHz. In some cases, detection at the third harmonic of the fundamental was used to eliminate baseline curvature. The demodulated signal was filtered and amplified and then sent to the PZT coupled to the cavity mirror in order to lock the frequency of the FIR laser to the center of the Lamb dip.

We measured the frequency of the FIR laser by mixing its radiation with the radiation from two CO_2 lasers (*6*) in a point contact metal-insulator-metal (MIM) diode. It has a tungsten whisker and a nickel base. The difference between the frequencies from the CO_2 lasers was used as the FIR reference frequency $[\nu_{CO2}(I) - \nu_{CO2}(II)]$. Each CO_2 laser was stabilized to the saturation dip in the 4.3-μm CO_2 fluorescence signal in an external reference cell (*6*). The FIR frequency measurements were performed at least 4 h after the lasers were turned on to ensure that all lasers had reached thermal equilibrium.

The beat-note signal from the MIM diode (ν_{beat}) was amplified and displayed on a spectrum analyzer. The center frequency was determined with the aid of a synthesizer-generated frequency marker. Each beat-note frequency was obtained from an average of at least 40 measurements. The FIR frequency (ν_{FIR}) is determined using the beat note frequency

$$\nu_{FIR} = |\nu_{ICO2} - \nu_{IICO2}| \pm \nu_{beat}. \quad [1]$$

The (+) or (−) sign was determined experimentally by tuning the FIR laser frequency before locking and looking at the beat note shift on the spectrum analyzer.

IV. RESULTS AND DISCUSSION

The Lamb dip on each gain curve shows a good signal-to-noise ratio and symmetry. Typically, the full-width at half maximum (FWHM) was less than 2 MHz. The frequency uncertainty was estimated from

$$\Delta\nu = [(2\sigma_{beat})^2 + \Delta\nu^2_{ICO2} + \Delta\nu^2_{IICO2} + \Delta\nu^2_{disper})]^{1/2}, \quad [2]$$

where σ_{beat} is the standard deviation of the beat note measurements, $\Delta\nu_{CO2}$ is the frequency uncertainty of the CO_2 lasers, and ν_{disper} is the frequency deviation due to dispersion pulling caused by the pump laser pulling the gain curve by Doppler pulling.

The standard deviation from 40 measurements of the beat note frequency measurement was below 10 kHz. Each measurement was an average from at least 50 scans of the spectrum analyzer with a scan width of 100 kHz, a sweep time of 2 s, and a resolution bandwidth of 10 kHz. The beat note frequencies

TABLE 1
Frequency Measurements of Sub-Doppler-Stabilized FIR Laser Lines from CH_3OH

FIR laser line		beat note		FIR Frequency		
λ	pump offset	3F	σ	Frequency	Δν[1]	Δν/ν
(μm)	(MHz)	(MHZ)	(kHz)	(MHz)	(kHz)	(x10^{-8})
70.5116	9P34 + 24	1009.995	5	4 251674.237	20	0.5
118.8341	9P36 + 24	1198.010	10	2 522 782.311	27	1.0
123.4515	9HP20+ 9	9.431	4	2 428 422.130	20	0.8
570.5684	9P16 + 64	1210.714[2]	6	525 427.746	21	4.0

[1] From expression (*2*).
[2] Locked at 1F.

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TABLE 2
Comparison of Frequency Measurements Reported by Different Methods

λ (μm)	This work (MHz)	FIR emission (MHz)	TuFIR (MHz)	mmw (MHz)
70.5116	4 251 674.237(20)	4 251 674.63 (15)[1]	4 251 674.56(10)[4]	
118.8341	2 522 782.311(27)	2 522 782.464(220)[2]	2 522 782.74(8)[4]	
			2 522 782.57(7)[2]	
123.4515	2 428 422.130(20)	2 428 422.60(50)[3]	2 428 421.90(5)[4]	
570.5684	525 427.746(21)	525 427.5(7)		525 427.902(50)[5]

Note. 1σ values in parentheses.
[1] Ref (*3*).
[2] Ref (*9*).
[3] Ref (*10*).
[4] Ref (*11*).
[5] Ref (*12*).

were within 10 kHz of each other for both 1F and 3F detection where 1F and 3F are first and third derivative locking to the centers of the Lamb dips.

The one-sigma frequency uncertainty of each CO_2 laser is about 10 kHz (*8*). The frequency deviation due to dispersion pulling (*5*) was measured by measuring the beat note shift when the frequency of the pump laser was changed. The beat note frequency shift was less than 70 kHz when the pump was detuned many times more than what would occur during normal operating conditions. The normal frequency error from dispersion pulling is estimated at 10 kHz. From time to time, the pump offset was reset to produce the maximum power in the FIR laser to minimize the effect of dispersion pulling on the FIR frequency.

The dispersion pulling for longitudinal pumping is

$$\Delta\nu_{\mathrm{FIR}} = \pm(\nu_{\mathrm{FIR}}/\nu_{\mathrm{PUMP}})\Delta\nu_{\mathrm{PUMP}}. \qquad [3]$$

At 2.5 THz, the FIR frequency would be shifted by 830 KHz for a 10-MHZ detuning of the pump laser, but with our transversely pumped laser this effect is only 10 KHz. The central Lamb dip is about 96% of that with longitudinal pumping at line center, and the pulled side-lobes are reduced by a factor of 4.

Table 1 summarizes the frequency measured using 1F detection for the line at 570 μm and 3F for the other lines. The total frequency uncertainty calculated from expression [*2*] is 27 kHz. Table 2 compares our more accurate results with frequencies using tunable radiation in the FIR region (TuFIR) for absorption measurements in the excited vibrational state of methanol and with laser radiation. The agreement is generally within the error bars of the less accurate measurements for about one-half of the frequencies. Presently, we do not understand this disagreement.

V. CONCLUSION

The reproducibility of a few parts in 10^8 of the measured frequencies is one order of magnitude better than the best previous results. The experiment can be extended to other strong FIR laser lines. The results will be useful for future FIR laser applications such as in secondary frequency standards and high-resolution spectroscopic applications.

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