Pressure effects on the frequency of continuous-wave optically pumped far-infrared lasers

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The frequency of the 170.6-μm cw CH_3OH optically pumped laser emission has been remeasured at different pressures without observing the pressure shift observed by Lawandy and Koepf [Opt. Lett. **5**, 383 (1980)]. The far-infrared frequency was synthesized with two stabilized CO_2 lasers. No measurable pressure shift over the operating pressure range of the laser was observed, and the frequency was confirmed to be 1 757 526.3 MHz. However, competing lasing lines were found to produce spurious effects on the frequency. These effects may explain the apparent shifts.

Optically pumped molecular lasers are the most widely used sources of radiation in the far-infrared (FIR) region of the electromagnetic spectrum. More than one thousand known lines span the wavelength spectrum from 30 μm to 1.2 mm, with power outputs from less than 0.1 to more than 100 mW, depending on the line and experimental conditions. The absence of a plasma in the cavity with its inherent fluctuations in the index of refraction is responsible at least in part for linewidths that are less than 1 kHz. Fractional frequency instabilities down to 2×10^{-12} have been demonstrated in apparatus especially designed for metrologic purposes.[1] Also, for conventional laboratory apparatus in free-running operation, the uncertainty in the frequency reproducibility is of the order of a few parts in 10^7. For instance, the measurements of the frequency of the 496-μm emission from CH_3F, performed in three different laboratories with three different experimental apparatuses, agree within 3 parts in 10^7.[2] As a result, optically pumped lasers have wide application in laser-frequency synthesis, laser-magnetic-resonance spectroscopy of atoms and molecules, and the spectroscopy of the lasing molecule itself.

Of fundamental importance for the frequency reproducibility of optically pumped lasers is the fact that pressure shifts on the molecular-laser active transitions are small, and it is generally assumed that they can be neglected in the range of operation from a few to about 100 Pa. However, an unexpectedly large change of frequency with the pressure was recently reported by Lawandy and Koepf.[3] They performed frequency measurements of the 170.6-μm emission line from a CH_3OH laser at two different pressures and interpreted the results as a pressure shift of 110 kHz/Pa, i.e., at least 1 order of magnitude larger than the one generally accepted. Because of the significant implications on the reliability of optically pumped lasers as reproducible sources, we decided to repeat the frequency measurements of this line at different pressures.

The FIR laser consisted of a 1-m Fabry–Perot FIR open resonator[4] pumped with a 1-m-long waveguide CO_2 laser. Pressure of the gas in the FIR resonator was measured with a thermocouple gauge, which was calibrated with a capacitance manometer. Frequency measurements were performed with the technique, described in Ref. 5, of synthesizing an appropriate local-oscillator signal from the difference frequency of two saturated-absorption fluorescence-stabilized CO_2 lasers. A W–NiO–Ni point-contact diode was used as mixer. The laser beams were focused on the conical tungsten antenna by parabolic mirrors.[6] The rf heterodyne output from the diode was fed to a spectrum analyzer for the measurement of the beat frequency. Simultaneously, the dc output was fed to an oscilloscope for observation of the power emitted by the laser. Many lines of methyl alcohol can be pumped with the same CO_2 line, and it is possible to record the modes of all the lines emitted at different wavelengths by scanning the length of the FIR cavity. The beat signal on the spectrum analyzer was obtained only from the line for which the proper frequency had been synthesized. The simultaneous observation of the mode pattern was important for an accurate frequency measurement. In fact, by scanning the FIR cavity we observed that the amplitude of the heterodyne signal on the spectrum analyzer was affected by the occurrence of a nearby mode of a competing line in the laser. The output was symmetric, i.e., the line could be observed along its whole frequency-tuning range, only for oscillation on single modes. However, the excitation of a competitive line at the low- or high-frequency side of the tuning range of a heterodyned signal caused a decrease in the output of the laser. Since the frequency of a FIR emission is stated as the maximum power point of the

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Table 1. Frequency Measurements of the 170.6-μm Laser Line at Different CH_3OH Pressures

CH_3OH Pressure [Pa (mTorr)]	Measured Frequency [(MHz) ± 2×10^{-7}]
31 (230)	1 757 526.054
27 (200)	1 757 526.084
17 (130)	1 757 526.054
6 (45)	1 757 526.594
5 (35)	1 757 526.454

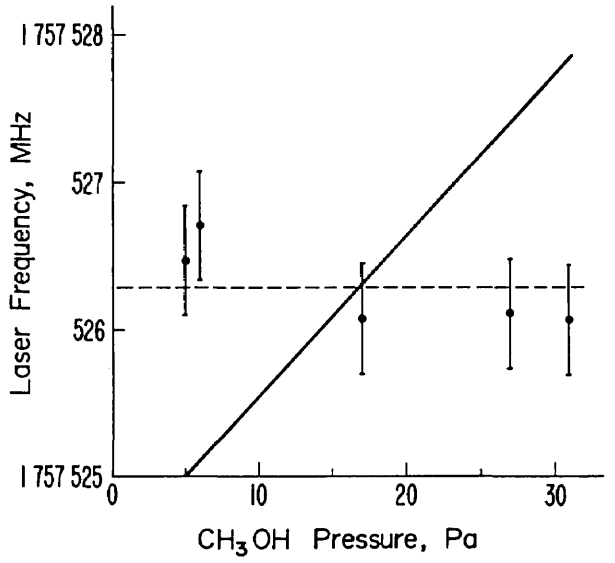

Fig. 1. Frequency of the 170.6-μm line as a function of CH_3OH pressure. Experimental points are taken from Table 1. The solid line refers to the pressure shift claimed in Ref. 3. The points represent the previously measured value.

tuning curve, the presence of a competitive line affecting the tuning curve itself can lead to erroneous determinations of the center frequency. For the CH_3OH emission at 170.6 μm [Pump $P_{II}(36)$], we synthesized a frequency of 1 756 866.114(23) MHz by using two stabilized CO_2 lasers oscillating on the $P_I(36)$ and $R_I(40)$ lines. We then measured the beat-note frequency of the FIR emission over a wide range of pressures, carefully selecting modes that were highly symmetric. The values obtained are summarized in Table 1. There is no evidence of a pressure shift, at least within the accuracy of the frequency measurements (a few hundred kilohertz). According to the data reported in Ref. 3, we should have observed a frequency shift of about 3 MHz between the highest- and lowest-pressure measurements. A comparison between our experimental data and the pressure shift estimated in Ref. 3 is shown in Fig. 1.

The average of the values in Table 1 is 1 757 526.29, with a maximum deviation of 400 kHz. The remeasured frequency reported in this Letter is exactly coincident with that reported in 1975[5] from a different FIR-laser configuration (a 2-m waveguide) and is consistent with the less accurate value reported in 1974.[7] Furthermore, the results obtained here suggest that the accuracy and reproducibility of optically pumped FIR lasers are $\pm 2 \times 10^{-7}$ rather than the value $\pm 5 \times 10^{-7}$ assumed in Ref. 4.

We also performed frequency measurements of the 170.6-μm line at fixed CH_3OH pressure but on modes perturbed by oscillation of the 118-μm line. The frequency obtained in the presence of a 118-μm mode competing on the high-frequency side of the tuning range was typically 0.5 MHz lower than that with a competitive mode on the low-frequency side. We think that this evidence can explain the results reported in Ref. 3. In fact, a change of the pressure affects the mode pattern, and perturbing modes can be excited or de-excited, causing misleading determinations of the frequency. The presence of a competitive mode at the low-frequency side is likely to cause the asymmetry in the high-pressure scan of Fig. 1 in Ref. 3.

Of course this spurious effect of the pressure is not limited to the case of the 170.6-μm line but can be observed on other lines, especially on weak lines competing with strong lines.

Our results confirm that pressure shifts can be neglected in cw optically pumped molecular lasers oscillating on a single mode. These results also indicate that frequency shifts at the pressure of operation of these FIR lasers can be predicted by the first-order-impact perturbation theory introduced by Anderson.[8] This theory yields pressure shifts of a few percent of the broadening (at least for transitions different from $J = 0 \rightarrow 1$) and hence shifts of only a few kilohertz per Pascal for CH_3OH.

In conclusion, we have confirmed that optically pumped lasers are reproducible and reliable frequency sources in the FIR if one uses care in selecting modes unperturbed by competitive FIR emissions.

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