# Measurement of Far-Infrared Laser Frequencies Generated by Optically Pumped $N_2H_4$ and $N_2D_4$

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## ABSTRACT

A three-laser heterodyne system has been used to measure the frequencies of four previously observed far-infrared laser emissions generated by optically pumping either hydrazine ($N_2H_4$) or its fully deuterated isotope ($N_2D_4$) with a continuous-wave carbon dioxide laser. These newly measured frequencies have fractional uncertainties of $\pm 2 \times 10^{-7}$ and correspond to laser wavelengths ranging from 63.9 to 158.4 μm. This work represents the first measurement of a $N_2D_4$ laser frequency.

## INTRODUCTION

Hydrazine ($N_2H_4$) has emerged as one of the most prolific sources of optically pumped laser emissions in the far-infrared (FIR) region. Beginning with the first observation of laser action from $N_2H_4$ in 1974 by Dyubko et al. [1], this molecule has been found to generate 293 laser emissions in the 49.2 to 1060.3 μm wavelength region [2-8]. In 1985, Shevyrev and co-workers investigated the fully deuterated isotope of hydrazine ($N_2D_4$) and discovered 31 FIR laser emissions ranging from 115.0 to 724.0 μm [9].

This work is a follow-up to our previous investigation of $N_2H_4$ and $N_2D_4$ [6], in which three new FIR laser emissions were observed for $N_2H_4$. Our objective was to measure the frequencies of these recently discovered laser lines as well as the frequency of the previously observed $N_2D_4$ laser emission.

## EXPERIMENTAL DETAILS

The FIR laser consisted of a tunable Fabry-Perot cavity, optically pumped in an X-V geometry by a 2 m long continuous-wave $CO_2$ laser [10]. The FIR cavity utilized a nearly confocal mirror system with one end mirror mounted on a micrometer to tune the cavity into resonance with the FIR laser radiation. Laser wavelengths were measured by traversing at least twenty adjacent longitudinal modes for a particular laser emission. The $N_2H_4$ sample was obtained from Sigma Aldrich while the $N_2D_4$, 98% $D_4$ enriched sample was obtained from Cambridge Isotope Laboratories.

The FIR laser frequencies were measured using the three-laser heterodyne technique, discussed in detail in Refs. [11-13]. Two $CO_2$ laser frequencies were combined in a Tungsten-Nickel metal-insulator-metal (MIM) point contact diode to create a difference frequency in the FIR region. These $CO_2$ reference frequencies were chosen such that their difference frequency nearly equals the FIR laser frequency calculated from the wavelength measurement. A beat between this difference frequency and the unknown FIR laser emission (also focused on the MIM diode) was then amplified and observed on a spectrum analyzer. If the separation between these frequencies was greater than the range of the spectrum analyzer, a microwave source was added to the MIM diode to decrease the frequency gap. The frequency of the beat was then measured by comparison with standardized frequency markers generated by the spectrum analyzer.

This measurement determined the unknown FIR laser frequency, $\nu_{FIR}$, through the relation

$$\nu_{FIR} = | n_1 \nu_{CO2(I)} - n_2 \nu_{CO2(II)} | \pm m \nu_{\mu wave} \pm \nu_{beat}, \quad \text{Eq. 1}$$

where $| n_1 \nu_{CO2(I)} - n_2 \nu_{CO2(II)} |$ is the difference frequency synthesized by the two $CO_2$ reference lasers, $\nu_{\mu wave}$ is the microwave frequency and $\nu_{beat}$ is the beat frequency. The integers, $n_1$, $n_2$ and m define the harmonics of the difference and microwave frequencies, respectively (first-order, second-order, etc.). The value of m and the ± signs in Eq. 1 were determined experimentally by tuning the FIR laser cavity and slightly varying the microwave frequency to obtain a small shift in the beat note on the spectrum analyzer.

The $CO_2$ reference lasers were frequency stabilized to their saturation dips in the 4.3 μm fluorescence signal from an external reference cell [14]. The one-sigma fractional uncertainty, $\Delta\nu/\nu$, of FIR laser frequencies measured with this technique was $\pm 2 \times 10^{-7}$. This uncertainty was derived from setting the FIR laser cavity to the center of its

gain curve. To minimize this uncertainty, the FIR laser was tuned across its gain curve, the center of which was measured on the spectrum analyzer using a peak hold feature. An iris in each of the laser cavities in the three-laser heterodyne system was used to eliminate higher order modes and reshape the gain curve to a symmetric pattern. The FIR laser frequency was calculated from the average of at least fourteen measurements recorded with varying microwave frequencies. In addition, these measurements were made with at least two different sets of $CO_2$ reference laser lines.

## RESULTS AND DISCUSSION

Table I lists the frequencies measured for the FIR laser emissions generated by optically pumped $N_2H_4$ and $N_2D_4$. These laser lines, varying in wavelength from 63.9 to 158.4 μm, are arranged by molecule in order of their $CO_2$ pump. All frequencies are new and were measured under optimal operating conditions. Along with their frequencies, these laser emissions are reported with their corresponding wavelength and wavenumber, calculated from the average frequency using 1 $cm^{-1}$ = 29 979.245 8 MHz, and their respective reference.

**Table I.** New FIR laser frequencies measured for optically pumped $N_2H_4$ and $N_2D_4$

| $CO_2$ Pump | Wavelength (μm) | Frequency (MHz) | Wavenumber ($cm^{-1}$) | Reference |
|---|---|---|---|---|
| $N_2H_4$ | | | | |
| 9*P*14 | 115.762 | 2 589 720.4 ± 0.6 | 86.3838 | 6 |
| 10*R*36 | 63.945 | 4 688 323.0 ± 1.0 | 156.3856 | 6 |
| | 83.520 | 3 589 481.1 ± 0.8 | 119.7322 | 6 |
| $N_2D_4$ | | | | |
| 9*P*36 | 158.409[a] | 1 892 518.2 ± 0.4 | 63.1276 | 9 |

[a] Previously reported as 159.5 μm [9].

## CONCLUSIONS

In conclusion, this three-laser heterodyne system has been used to measure four FIR laser frequencies from $N_2H_4$ and $N_2D_4$. The frequency for the 158.4 μm line of $N_2D_4$ was the first frequency measurement performed for a laser emission generated by this molecule. The measurement of FIR laser frequencies will continue to permit the spectroscopic investigation of the FIR laser medium (e.g. [15, 16]) as well as its use as a source of coherent FIR radiation for the spectroscopic investigation of unstable molecular species with the laser magnetic resonance technique (e.g. [17]).

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