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Frequency measurement of the formic acid laser 311- μm line

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The optically pumped formic acid (HCOOH) laser is a rich source of submillimeter laser lines. A total of fifty-nine distinct lines have been reported.¹⁻⁸ The frequencies of twenty-seven of these lines have been measured accurately by mixing the laser output with harmonics of various millimeter wave sources.⁶⁻⁸ The weaker lines have been observed in some experiments, but not measured accurately. This letter reports a frequency measurement of one of these weaker lines, the 311- μm line, pumped by the CO₂ laser 10R22 transition.

The optically pumped submillimeter laser used in this experiment was similar to one described by Hodges *et al.*⁹ A 1.45-m-long, grating tuned cw CO₂ laser produced up to 30 W of power at the 10R22 line used to optically pump the formic acid. The submillimeter laser consisted of a Pyrex waveguide 2 m long by 38 mm in diameter with the hole input coupling and hybrid output coupling. The input copper mirror had a 4-m radius of curvature to reduce the number of low-loss modes in the laser and facilitate cavity scans for wavelength measurement. The frequency of the output from this laser was measured by mixing the laser output in a point contact diode with the 311- μm line from a submillimeter HCN discharge laser (see Fig. 1). The HCN laser was tuned to its strong 311- μm line by means of cavity scanning. The lowest order beat note obtained from mixing the two laser outputs, which was near 2 GHz, was amplified directly with a GaAs FET amplifier and observed with a microwave spectrum analyzer. A dc bias was applied across the point contact diode

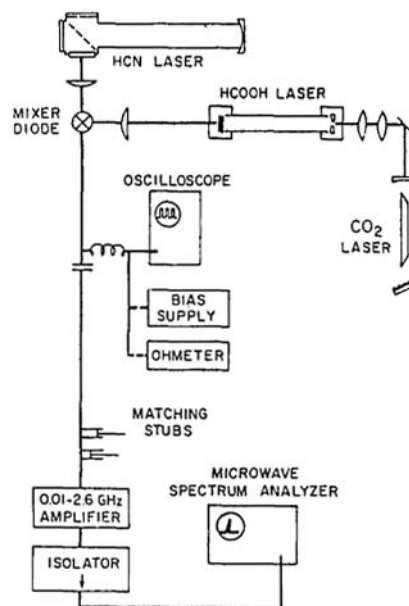


Fig. 1. Experimental apparatus for two-laser mixing and frequency measurement.

Table I. HCOOH Optically Pumped Laser Line Reports for CO₂ Laser 10R22 Pump Line

Wavelength	Reference
311.45	Wagner <i>et al.</i> ¹
311.0 ± 7	Dyubko <i>et al.</i> ²
309 ± 3	Plant <i>et al.</i> ³
311.554 ± 0.002	Present work

to optimize its mixing performance. Further details of the mixing arrangement may be found in Ref. 10.

The wavelength of the HCOOH laser was measured first by scanning the laser output coupler through a number of half-wavelengths (typically twenty). From the beat note observed on the spectrum analyzer, the wavelength was determined to better than 0.1%. This measurement confirmed that the HCOOH frequency was below that of the HCN laser. After calibration of the spectrum analyzer, the beat frequency from the two lasers was measured to be 2.063 ± 0.005 GHz. The HCN frequency was taken to be 964.3134 GHz,¹¹ from which the HCOOH line was derived to be 962.250 ± 0.005 GHz. The principal source of uncertainty in this result was the spectrum analyzer calibration (± 3 MHz); additional errors were introduced in determining the line center of the laser output (± 250 KHz) and in reading the spectrum analyzer (± 1 MHz). The formic acid laser line could be tuned between 1 MHz and 3 MHz, depending on the size of the output coupler used in the submillimeter laser. This places a lower limit on the gain bandwidth of the HCOOH line. From the observed width of the beat note, the laser linewidth had an upper limit of 30 kHz.

Conversion of frequency to wavelength permits comparison to previous measurements of the 311- μm line (see Table I). Dyubko *et al.*² and Plant *et al.*³ reported low accuracies in their work. Wagner *et al.*¹ performed a more accurate wavelength measurement, but their uncertainty was not

clearly stated. In making frequency measurements, Dyubko *et al.*⁶ and Deldalle *et al.*⁷ failed to obtain values for the 311- μm line.

Direct mixing of two submillimeter lasers has been demonstrated as a useful means of making FIR laser frequency measurements. The present frequency measurement should be accurate enough to be useful in making an assignment of the formic acid line and in reaching a better understanding of the spectrum of this important submillimeter laser molecule.

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Spectroscopic analysis of the transverse excited $C^3\Pi_u-B^3\Pi_g(0-0)$ UV laser band of N_2 at room temperature: comments

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Recently Petit *et al.*¹ published a paper devoted to a high-resolution spectroscopy study of the 0-0 laser band in the Second Positive System of molecular nitrogen and explaining the optical gain formation on its different branches. All the results by Petit *et al.*¹ are equivalent to those previously published by Massone *et al.*² and are in accordance with theoretical predictions reported by Tocho *et al.*³ Summarizing, we would comment as follows.

In 1972, Massone *et al.* published a paper² devoted to accurate wavelength measurements in the First and Second Positive Systems emitted by a molecular nitrogen pulsed axial laser and to analyze the possible pumping mechanisms in accordance with their experimental results. These results confirmed the general validity of the conclusion of Kaslin and Petrash⁴ relating to the influence of temperature on the

properties of a pulsed diatomic molecular laser. Such a conclusion, which is more general than that reported previously by Patel,⁵ is: The dependence of the gain on J' is the same as the dependence of the intensity on spontaneous emission from the upper level. (J' is the quantum number of the upper level.) The Patel analysis corresponds to vibrational-rotational transitions in diatomic and polyatomic (linear) molecules, while Kaslin and Petrash referred to electronic-vibrational-rotational transitions. Both analyses assumed that the coupling case (a) of Hund is achieved. As is well known, both electronic states in molecular nitrogen, $C^3\Pi_u$ and $B^3\Pi_g$, conform to coupling case (a) for low and moderate J values and to case (b) at high J values. Because of this, Massone *et al.* study the gain dependence as a function of J taking into account gas temperature by using the rotational intensity factor i calculated analytically by Budó⁶ for the electronic transition $C^3\Pi_u-B^3\Pi_g$ corresponding to the UC Second Positive System. The i Budó factors are tabulated according to the Hund coupling cases. Massone *et al.* presented various diagrams for different branches belonging to the 0-0-UV band at 100 K and 365 K temperatures, and the results were in agreement with their experimental observations.

The observation of both components of the Λ -doublets in all the P_1 transitions, and in one of the P_2 , was the major spectroscopic difference between Massone *et al.*² and Parks *et al.*⁷ results. But as in the experiments conducted by Massone *et al.* an axial discharge tube was used, while in those by Parks *et al.* a transverse discharge tube was used, it was decided at La Plata to build a superradiant emitter similar to others reported up to 1974,⁸⁻¹⁰ in which the nitrogen gas and the discharge tube were cooled with liquid air. The results obtained with this equipment were published by Tocho *et al.*³ According to the level of excitation of the nitrogen molecules, their spectral analysis of the 0-0 uv band clearly showed that the alternating symmetry Λ -doublets components were observed. This main result led to a definitive solution of the spectral misinterpretation made by Parks *et al.*,⁷ which was critically discussed by Massone *et al.*² Also, Tocho *et al.*³ concluded that the comparison between the spectral observations of the "strong Λ -doublets lines," as Dieke and Heath¹¹ called them, in transverse laser excitation and the full resolved components of the Λ -doublets in axial laser excitation confirms that the Λ -doublets may be observed if the threshold excitation conditions of nitrogen molecules are fulfilled. This statement, of course, clearly indicated that the observation of the Λ -doubling is completely independent of the geometry mode of operation of the discharge tubes.

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