Optically Pumped FIR Lasers: Frequency and Power Measurements and Laser Magnetic Resonance Spectroscopy

KENNETH M. EVENSON, DONALD A. JENNINGS, F. RUSSELL PETERSEN, JOHN A. MUCHA, JUAN J. JIMÉNEZ, R. M. CHARLTON, AND CARLETON J. HOWARD

Abstract—Optically pumped FIR lasers are currently in use in both frequency metrology and laser magnetic resonance spectroscopy programs in the NBS Boulder labs. The laser for use in frequency metrology is a CW 71 μm methyl alcohol waveguide laser with over 100 mW output for frequency synthesis. Another laser with an intracavity absorption cell for laser spectroscopy has been constructed and is nearly transversely pumped. The metrology technique used to measure the frequency of these lasers is briefly reviewed and a unique power meter is described.

I. Frequency Metrology

THE chief goal of our work is to extend direct frequency measuring techniques to the visible portion of the electromagnetic spectrum. In this extension of frequency metrology towards the visible, the frequencies of a number of the methyl alcohol lines were measured [1] using a conventional waveguide laser, synthesized frequencies from two stabilized CO_2 lasers, and the tungsten-nickel point contact diode.

The highest frequency measured so far [2] is 148 THz (2.03 μm), the frequency of the He-Xe laser. The frequency of the methane line used to lock the 3.39 μm He-Ne laser has also been measured; [3] and when multiplied by the wavelength [4] has yielded the currently accepted 100 fold more accurate value of the speed of light [5] (299, 792, 458 m/s). More accurate frequency measurements of this methane stabilized laser will be attempted using a simplified chain of lasers employing a 71 μm methyl alcohol laser instead of the 337 μm HCN and 28 μm H_2O lasers previously employed.

II. 71 μm Laser

The 71 μm laser is 4 m long and uses a 14 mm inside diameter quartz (dielectric) waveguide. Flat mirrors are utilized and spaced 2 or 3 mm from the ends of the quartz tube. The moveable mirror is made of copper and has a 1 mm coupling hole to admit the CO_2 laser beam into the FIR cavity. The CO_2 beam is focused through this mirror with a 20 cm focal length lens. A fixed salt window 2 cm from the copper mirror makes the vacuum seal for this end of the laser. The copper mirror is suspended from a rod positioned on ball bushings 3 cm above the FIR laser axis.

The output mirror consisted of a plane parallel silicon reflecting wafer 1.2 mm thick separated by $\lambda/4$ (17.5 μm) from a 3.85 mm thick plane parallel reflecting wafer which also served as the vacuum seal. Both of these were chosen from several samples available to give maximum reflectivity at 71 μm. To block the small amount of residual 10 μm radiation which is transmitted by the silicon and also to further enhance the 71 μm reflectivity, a 0.4 mm crystal quartz disk was spaced at $\frac{3}{4}\lambda$ outside the thicker silicon wafer. The reflectivity of the entire multimirror was about 90 percent at 71 μm. The 17.5 μm separation was achieved by vacuum deposition of 3 spots of nickel to act as spacers on the 3.85 mm wafer. These were then epoxied together to form a permanent two layer mirror. The outer quartz layer was separated by 51 μm ($\frac{3}{4}\lambda$) with a 2 mil plastic spacer and was clipped in place with 3 spring fingers.

Over 100 mW of 71 μm power was obtained when the laser was pumped with 30 W of CO_2 laser radiation. The FIR power was measured with the power meter described next.

III. Broadband Power Meter

The power meter used to measure the power of the 71 μm laser is shown in Fig. 1. It is useful at wavelengths from 0.4 μm to at least 500 μm, is sensitive from 1 mW to 1 W, and is accurate to better than 10 percent.

The 0.04 mm thick copper cone absorber is lined with a colloidal graphite paint which serves as the absorber. At 337 μm less than 5 percent of the power was reflected from this cone. The temperature rise of the cone was measured at 3 positions with 3 pairs of number 30 copper-constantan thermocouple junctions. These were attached with thermally conducting, electrically insulating epoxy. The power meter was calibrated with an electrical heater formed with teflon coated number 26 nichrome wire uniformly spiraled onto the copper cone. The current and voltage were measured with a precision ammeter and volt meter. Calibration curves were then made and the power meter was checked in the visible against an NBS secondary standard. The two agreed within 5 percent.

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K. M. Evenson, D. A. Jennings, F. R. Petersen, J. A. Mucha, and R. M. Charlton are with the Laser Physics Section, National Bureau of Standards, Boulder, CO 80302.

J. J. Jiménez was with the Laser Physics Section, National Bureau of Standards, Boulder, CO. He is now with the Centre National de la Recherche Scientifique and Bureau National de Metrologie, Paris, France.

C. J. Howard is with the Aeronomy Laboratory, NOAA, ERL, Boulder, CO 80302.

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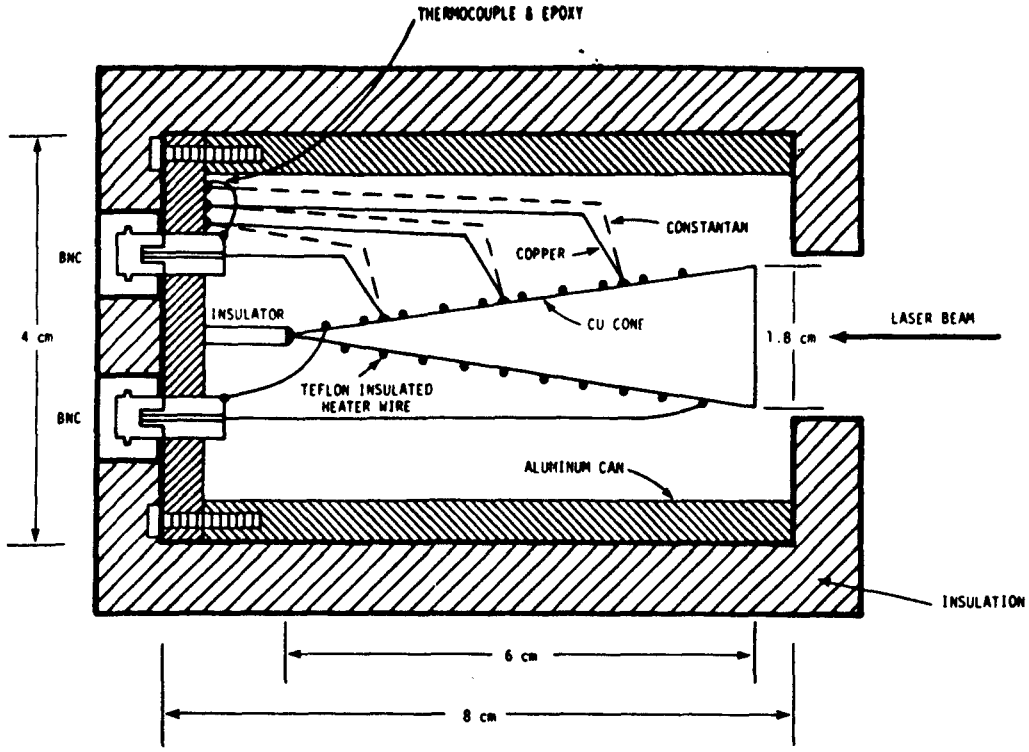

Fig. 1. Broadband CW laser power meter.

IV. Laser Magnetic Resonance

Laser magnetic resonance (LMR) is an extremely sensitive technique for the study of paramagnetic molecules (especially free radicals). Previous efforts have yielded spectra of OH [6], CH [7], HO_2 [8], O_2 [9], HCO [10], NO [11], NO_2 [12], NH [13], PH [14], NH_2 [15], PH_2 [16], and CH_3O [17]. Most of the spectra were observed with an intra-laser cavity absorbtion, H_2O or HCN laser spectrometer operating at 78-119 μm and 311 and 337 μm.

V. LMR Optically Pumped Spectrometer

To take advantage of the large number of optically pumped FIR laser frequencies, an optically pumped FIR magnetic resonance laser spectrometer has just been put into operation. Based on the observation of the $N = 1 \rightarrow N = 3$ spectrum of oxygen at 700 μm shown in Fig. 2 and the $N = 13 \rightarrow N = 15$ spectrum at 119 μm, the sensitivity would seem to be at least as good as that of the H_2O laser spectrometer.

The spectrometer is shown in Fig. 3. It uses nearly transverse pumping with the CO_2 laser radiation, and it has a conventional 2 mirror Fabry-Perot FIR laser cavity. The radii of the two mirrors were chosen so the beam waist is at the center of magnet. A polyethylene Brewster angle beam splitter separates the sample cell from the FIR laser medium. The beam splitter can be rotated to ensure polarization of the laser radiation either parallel or perpendicular to the magnetic field.

FIR powers of about 100 μW or less emerge from the $\frac{1}{2}$ mm coupling hole. The power levels are sufficient to saturate the detector and the exit hole is small enough so the laser cavity Q is not appreciably affected. The signal has been shown to be proportional to the cavity Q [18].

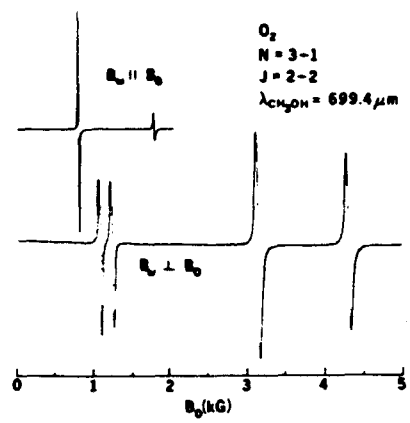

Fig. 2. LMR spectrum of oxygen taken at a pressure of 1330 pascals (10 torrs), modulation of 10^{-3} T (10 Gauss), and a time constant of 0.1 second.

The abundance of the FIR optically pumped laser lines should now enable spectroscopists to find the near coincidence required to observe laser magnetic resonance spectra of almost any desired paramagnetic molecule.

VI. Chemical Kinetics

A flow system has been added to our H_2O laser spectrometer so that the reaction rates of various free radicals with various atmospheric constituents can be measured. A number of reactions of OH with halogen substituted methane and ethane compounds have already been measured [19], [20], and they indicate that fluorocarbons FC 11 and FC 12 used commonly as spray can propellants and refrigerants have

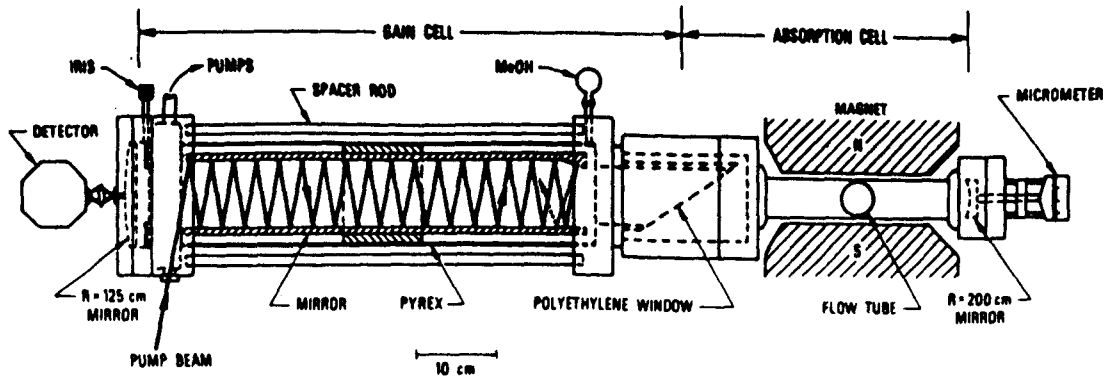

Fig. 3. FIR optically pumped laser magnetic resonance spectrometer.

extremely small reaction rates with OH. Thus, they are not "scrubbed" from the atmosphere by OH and are free to drift to the upper atmosphere where they are photo dissociated and begin the catalytic destruction of the ozone layer. We have shown that alternative propellants and refrigerants containing hydrogen do have appreciable reaction rates with OH and hence are "scrubbed" from the atmosphere before they drift to the stratosphere to destroy our planet's ozone layer.

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