

## ATOMIC OXYGEN FINE-STRUCTURE SPLITTINGS WITH TUNABLE FAR-INFRARED SPECTROSCOPY

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### ABSTRACT

We have accurately measured fine-structure splittings of atomic oxygen ( $^{16}\text{O}$ ) in the ground state using a tunable far-infrared spectrometer. The  $^3P_0-^3P_1$  splitting is 2,060,069.09 (10) MHz, and the  $^3P_1-^3P_2$  splitting is 4,744,777.49 (16) MHz. These frequencies are important for measuring atomic oxygen concentration in Earth's atmosphere and the interstellar medium.

*Subject headings:* infrared: spectra — interstellar: matter — laboratory spectra — planets: atmospheres

### 1. INTRODUCTION

Atomic oxygen plays an important role in the physics and chemistry of Earth's atmosphere. The  $^3P_1 \rightarrow ^3P_2$  fine-structure transition plays a major part in cooling the upper thermosphere (Bates 1951), and atomic oxygen is a key constituent in the chemistry of the ozone cycle. Possibly the best way to determine the oxygen concentration and thermosphere temperature is by monitoring far-infrared (FIR) fine-structure transitions (Sharma, Harlow, & Riehl 1988). The  $^3P_1 \rightarrow ^3P_2$  transition is also predicted to be the most important cooling transition for photodissociation regions (Tielens & Hollenbach 1985); it has been observed in the Galactic center (Lester et al. 1981) and other galaxies (e.g., Watson et al. 1984), as well as numerous other Galactic sources. Atomic oxygen is also thought to be a major constituent in the atmospheres of Venus and Mars (Chamberlain 1978).

Previous measurements of these transitions were performed using FIR laser magnetic resonance (LMR) (Davies et al. 1978; Saykally & Evenson 1979). The frequency reported by Saykally & Evenson (1979) was later corrected and appeared in a paper by Watson et al. (1984). We have improved upon those measurements by using a tunable source of FIR, thereby eliminating the external magnetic field used to tune the transitions in LMR. In addition, the absolute frequency of our FIR radiation is known to 35 kHz, as opposed to the 1 MHz uncertainty of the LMR laser frequency. The present transition frequencies are one order of magnitude more accurate than those from the LMR measurements and are limited by the noise in recording the absorption lines.

### 2. EXPERIMENTAL

The tunable far-infrared (TuFIR) radiation was generated from the difference between two  $\text{CO}_2$  lasers by nonlinear mixing in a metal-insulator-metal (MIM) diode (Evenson, Jennings, & Peterson 1984). For the high-frequency transition, microwave radiation was added to generate sidebands on the

$\text{CO}_2$  difference frequency. An in-depth description of the TuFIR spectrometer can be found in the paper by Jennings (1989). This technique has been used to study many astrophysically important molecules such as OH (Brown et al. 1986) and MgH (Zink et al. 1990), but this is the first application of the TuFIR technique to an atom. This was possible because of several changes in the spectrometer which have increased the sensitivity and also because of the relatively large amount of atomic oxygen produced.

Spectrometer improvements came from increased collection efficiency of the generated FIR, reduction of the standing wave between the MIM diode and the FIR detector, and the use of a double-pass absorption cell. Collection efficiency has been increased by the use of a right-angle rooftop reflector in combination with shorter diode whiskers (2–3 times the FIR wavelength) (Grossman 1988). This not only increases the FIR radiation collected by our optics, it also produces a smaller, better collimated beam that is nearly 100% linearly polarized (Zink et al. 1987). The second improvement is the use of a phase scrambler; this is a combination of mirrors composing an optical delay line, two of which undergo small, low-frequency oscillations. This modulates the optical path length between the MIM diode and the detector and virtually eliminates detected FIR power variations caused by standing waves.

The double-pass cell takes advantage of the FIR linear polarization. The FIR radiation passed through a wire mesh beamsplitter mounted at  $45^\circ$  to the beam propagation direction (Gottlieb, Vrtilek, & Thaddeus 1989). The mesh was oriented to pass the linearly polarized FIR radiation. The beam then entered the absorption cell and after traversing the cell was retroreflected by an internal polished rooftop copper mirror. This mirror was oriented to change the FIR polarization by  $90^\circ$  upon reflection; after traversing the cell, the return beam (with rotated polarization) is reflected by the beamsplitter to a liquid He-cooled Ge:Ga photoconductive detector.

Atomic oxygen was produced in a 3.8 m long, 22 mm inside diameter discharge cell in an atmosphere of 8 Pa of O<sub>2</sub> and 200 Pa of He or Ar with 40–60 mA current. Because of the efficiency of this production technique, the atomic oxygen concentration had to be reduced, by lowering the O<sub>2</sub> pressure, to prevent total absorption of the FIR radiation. The FIR radiation is frequency modulated at 1 kHz and phase-sensitively detected. The modulation depth for these measurements was 8 MHz, and the observed linewidth was about 10 MHz.

### 3. RESULTS AND DISCUSSION

For the  $^3P_0-^3P_1$  transition, the radiation from two CO<sub>2</sub> lasers was mixed in a W-Ni MIM diode. The FIR difference frequency was tuned by tuning one of the lasers over its gain curve. The signal-to-noise ratio was 40:1 with a 100 ms time constant, and six scans (three with increasing frequency, three with decreasing frequency) were averaged together. The measured transition frequency is 2,060,069.09 (10) MHz, where the number in parentheses is the 1  $\sigma$  uncertainty in the last digits.

The  $^3P_1-^3P_2$  transition was measured with microwave sidebands of a CO<sub>2</sub> difference frequency, generated in a W-Co diode (Evenson et al. 1988). In this case the TuFIR was tuned by changing the microwave frequency. Again six scans were

averaged with a 100 ms time constant, and the signal-to-noise ratio was also about 40:1. The measured frequency is 4,744,777.49 (16) MHz.

The  $^3P_0-^3P_1$  LMR measurement of Davies et al. (1978) is in agreement with our measurement within the LMR uncertainty; however, the present measurement is one order of magnitude more accurate. The other fine-structure transition frequency is 1.5  $\sigma$  different from the LMR measurement of Saykally & Evenson (Watson et al. 1984); the present measurement is 3.7 MHz higher. The difference is probably due to uncertainty in the FIR laser frequency used in the LMR experiment caused by different pumping geometries for the LMR laser and the laser used in the frequency measurement.

These fine-structure splittings are the first atomic transitions observed with our TuFIR spectrometer. The frequency uncertainty of 160 and 100 kHz provides more than adequate accuracy for astronomers and aeronomists to monitor atomic oxygen emissions.

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