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Accurate modeling of the diagnostic 118-GHz oxygen line for remote sensing of the atmosphere

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ABSTRACT

We report the results of laboratory investigations of the shape of the diagnostic atmospheric N = 1-oxygen line performed over a very wide range of pressures from 0.4 to 1000 Torr using two principally different spectrometers having complementary abilities. A spectrometer with a radio-acoustic detector of absorption was used for recording low pressure spectra spanning the 0.4–2 Torr range, and high pressure data from 250 to 1000 Torr were registered by a resonator spectrometer. The sensitivity of both instruments was improved significantly which allowed us to obtain signal-to-noise ratio at spectra recordings of the order of a few thousands. The spectra analysis enabled the first manifestation of the speed-dependence of the collision cross section of the line, along with considerable refinement of other parameters, including pressure broadening, intensity and line-mixing. The results are of primary importance for atmospheric applications.

1. Introduction

The present day methods of remote sensing demand high accuracy spectroscopic information about the diagnostic lines employed. The requirements for information accuracy are becoming increasingly more stringent. Line intensity and line width are known to be the most crucial parameters affecting accuracy of the retrieved atmospheric data. Another important issue is that a single measurement of a parameter cannot ensure the declared (usually statistic) uncertainty due to possible systematic errors (see, e.g., Figs. 15–17 in Ref. [1]). So, reliable information can be obtained only from the multiple studies of a target line by different spectroscopic laboratory techniques in a wide range of conditions.

A single fine structure oxygen line near 118 GHz is an important diagnostic line that serves in remote sensing as a “temperature and pressure sensor” (see, e.g. [2,3]). This line has been a subject of a large number of studies [4–13], including a series of our works [14–20]. We believe that the results of our studies provide the most accurate to date experimental values of line intensity [18], pressure broadening [19,20] and mixing coefficients [18] and their temperature dependences [20]. In the aforementioned studies of the 118-GHz line we used spectrometers having different measurement principles, significantly different working pressures and complementary abilities. Coincidence of some parameters (e.g., pressure broadening coefficients) measured by different methods allowed us to claim high reliability of the data obtained in those studies. The main limitation of those data concerns the use of the basic line shape models for fitting experimental spectra: the Voigt and Rosenkranz profiles respectively for low and high pressures. Indeed, it is well known [21] that in most cases these simple models fail to reproduce the experimental line profile recorded with a sufficiently high signal-to-noise ratio (SNR). Thus, deviation of the Voigt profile from the experimental one is typically about 1–2% of the peak absorption (see, for example [22–25],) reaching up to 8–9% [26,27] limiting accuracy of atmospheric profile modeling.

Achieving better accuracy requires taking into account first of all the dependence of collisional width and shift on the speed of absorbing molecules [28] called the speed-dependence (SD) or ‘wind’ effect, and velocity-changing collisions causing the Dicke narrowing [29]. For the lines of millimeter and submillimeter wavelength range, where Doppler broadening is relatively small, the SD-effect has been shown [22] to be the major cause of deviation of the experimental line shape from the Voigt profile, even at relatively low pressures corresponding to the conditions of the upper stratosphere. For the oxygen fine structure lines this effect has never been studied before and has not been taken into account in the models of atmospheric absorption of radiation by oxygen.

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However, even thorough modeling of the shape of the 60-GHz oxygen band formed by a large number of fine structure lines demonstrates [30] the difference of the calculated profiles from the experimental spectra by about 1–2%. This difference may be a result of neglecting the SD-effect in the model. Analysis of oxygen line shape at high enough pressures (hundreds of Torr) demands allowance for the cumulative effect of the SD and the line mixing that impedes such studies.

In this paper we report the results of laboratory investigations of the shape of the 118-GHz oxygen line performed over a very wide range of pressures (0.4–1000 Torr) using two principally different spectrometers having complementary abilities. The spectrometer with radio-acoustic detector [31] was used to record low pressure spectra spanning the 0.4–2 Torr range, and high pressure data from 250 to 1000 Torr were recorded with a resonator spectrometer [32]. The considerably improved sensitivity of both instruments allowed us to obtain signal-to-noise ratio at spectra recordings of the order of a few thousands. The spectra analysis enabled the first manifestation of the speed-dependence of collisional broadening of the line, along with considerable refinement of other parameters of the line, including collisional broadening, line-mixing and integrated intensity.

2. Low pressure experiment using RAD spectrometer

A spectrometer with a backward-wave oscillator (BWO) and a radio-acoustic detector of absorption (RAD spectrometer) [31] was used for studying the 118-GHz oxygen line in the 0.4–2 Torr pressure range. The spectrometer and the measurement method are similar to those used in our earlier studies [19,20,33], so we present here only their brief description and main changes in the setup and method. A copper gas cell ($\sim$10 cm long, $\sim$1.5 cm in diameter) was placed inside a double shield made of annealed permalloy to avoid distortion of the shape of the magnetic-dipole oxygen line by external magnetic fields. The cell was permanently connected with the Julabo FP-50 HE thermostat that provides stable temperature of a coolant inside the thermostat within $\pm$0.01°C around the chosen value of 23.7°C. The cell and both shields were additionally packed into a heat-isolating material minimizing temperature gradients and drifts. Four copper temperature sensors were mounted on the cell surface and allowed temperature control of the gas sample inside the cell to an accuracy of $\pm$0.5°C. It should be noted that for the current room-T study no gradients and drifts of the cell temperature were revealed within the T-sensors accuracy. Gas pressure in the cell was permanently monitored using a 10-Torr range MKS Baratron (Type 626B) gauge having a declared accuracy of 0.25% of reading.

In our previous studies of the oxygen fine structure lines using RAD spectrometer the signal-to-noise ratio of the recorded spectra was about a few hundreds for 1 s integration time constant [19,20,33]. However, this is almost an order of magnitude less than the SNR required for observation and study of the SD effect. To achieve the required SNR, some components and parameters of the previous RAD spectrometer were modified. In particular, for reducing the influence of external acoustic and mechanical noise the cell was weighted by about twenty five kilograms by the small lead balls 3-mm in diameter (initial cell weight was about 2.5 kg). This allowed a fivefold increase of the SNR of the experimental spectra. Analysis of the line recordings obtained at different amplitude modulation frequencies in the 70–290 Hz range revealed a possibility of further SNR improvement. The frequency should be outside unfavorable intervals that occur probably because of mechanical resonances in the setup elements (table, vacuum system, etc.). The use of the modulation frequency of 80 Hz instead of the previously used 180 Hz give a 25% increase of SNR.

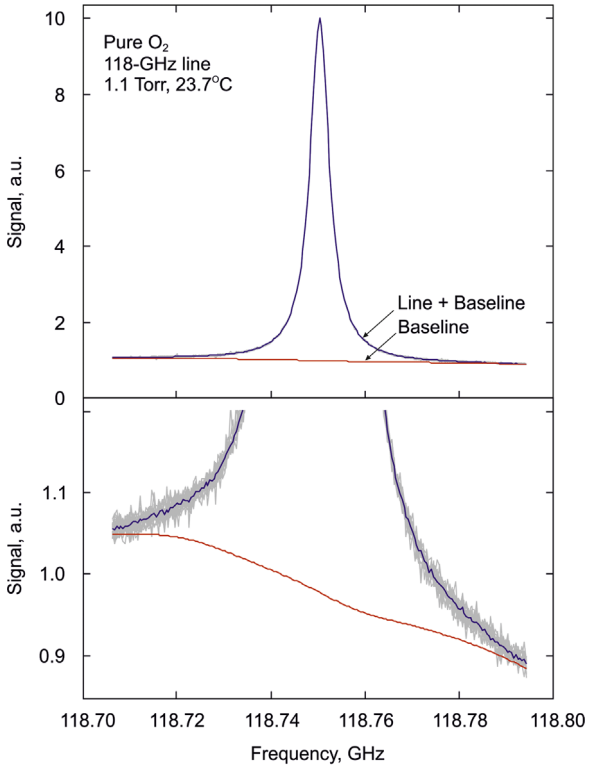

Fig. 1. (Color online) Experimental spectra of pure oxygen near 118.75 GHz. The lower plot is a zoomed-in part of the upper plot. Twenty spectra recorded at 1.1 Torr of pure oxygen are shown by grey. The blue curve is the result of their averaging. The red curve shows the baseline recorded for the same conditions with 1.1 Torr of pure nitrogen.

Finally, high stability of radiation parameters (frequency and power) and experimental conditions (room and cell temperature, pressure in the cell) allowed averaging a large number of repeated experimental recordings for each chosen pressure for achieving SNR of a few thousands. The number of averaged line recordings varied from 40 for lower pressure (lower sensitivity of the acoustic cell) down to 20 for higher pressures. An example of averaging twenty experimental spectra is shown in Fig. 1.

At the next step of experimental spectra treatment, the instrumental baseline was taken into account. The baseline arises from absorption of the radiation by the cell elements providing secondary gas heating and thus producing a microphone signal synchronous with modulation. Similarly to our previous studies the baseline signal was recorded at the same configuration of the waveguide line of the spectrometer and the pressure of nitrogen (negligibly absorbing in mm/submm range under these conditions) in the cell same as for oxygen spectra recording (see Fig. 1). Experimental baseline was subtracted from the sample spectra and the obtained spectra were then used for the line shape analysis.

3. High pressure experiment using resonator spectrometer

A modified version of the resonator spectrometer was employed for studying the shape of the 118-GHz line in the pressure range of 250–1000 Torr. A detailed description of the spectrometer and measurement method used in our earlier studies of the oxygen fine structure lines [16,18,33] is given in Refs [32,34]. The block diagram of the modified spectrometer is presented in Fig. 2.

The gas absorption measurement is based on determining the Fabry–Perot resonator Q-factor. The latter can be found as a ratio of eigenfrequency and resonance curve width, leading to the following