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Collisional broadening of oxygen fine structure lines: The impact of temperature

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ABSTRACT

The temperature dependence of self-, nitrogen- and air-broadening coefficients of oxygen fine structure lines was studied. Temperature exponents of a series of oxygen lines with rotational quantum number N ranging from 1 to 19 were measured with high accuracy for the first time. The obtained parameters were compared with the results of the earlier studies. This information is of high importance for atmospheric applications.

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1. Introduction

The significant role for different applications of the oxygen fine structure lines forming the 60-GHz atmospheric band is well known. In particular, the band is widely used for retrieving vertical pressure and/or temperature profiles from remote sensing data obtained by ground based, airborne and satellite instruments. These applications require knowledge of the line parameters with high enough accuracy [1,2]. Pressure broadening of the lines is known to be a crucial parameter affecting accuracy of the retrieved data. Broadening of the fine structure lines by pressure of the principal atmospheric gases (nitrogen, oxygen and water vapor) and rotational dependence of the corresponding coefficients have been accurately studied at room temperature in our recent works [3,4]. However, their temperature dependence has not been studied for most of the band lines. The majority of the papers devoted to measurements of the temperature exponent n_γ of the oxygen fine structure line width [5–8] (see Fig. 4) concerns the single $N=1-$ line at 118 GHz [6–8]. We are familiar only with two studies of the $9+$ fine structure line and the 425-GHz pure rotational line carried out by Liebe [5] and Drouin [7], respectively. The value of n_γ in all of the aforementioned measurements varies within the 0.61–0.9 range (corresponding to about $\pm$ 19% deviation from the mean value) for self-broadening and within the 0.79–1.04 range (about $\pm$ 14%) for nitrogen broadening. Results of the study of pressure broadening in oxygen A-band [9] demonstrate some smooth rotational dependence of the temperature exponent (see Fig. 4) which varies within 0.6–0.77 (about $\pm$ 12%) with the estimated uncertainty of about $\pm$ 15%. Such a large spread of available data and insufficiency of systematic measurements in both microwave and visible regions impede accurate evaluation of the rotational dependence of the temperature exponent.

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The present study is devoted to measurements of the temperature exponent for self-, nitrogen- and air-broadening of the oxygen fine structure lines.

2. Experimental details

The spectrometer with a backward-wave oscillator and a radio-acoustic detector of absorption (RAD spectrometer) was employed for the current investigation. The spectrometer design and the corresponding method of measurements are described in detail in Refs. [10,11]. Similar studies of pressure broadening performed at room-temperature are described in Refs. [3,4,12]. The copper gas cell ($\sim$10 cm length, $\sim$2 cm diameter) was placed inside a double shield made of annealed permalloy to avoid distortion of the shape of the magnetic-dipole oxygen lines by external magnetic fields. The cell was permanently connected with Julabo FP-50 thermostat (http://www.julabo.de/) that provides stable temperature of a coolant inside the thermostat within $\pm$0.1 °C of the chosen value. The cell as well as both shields were additionally isolated from ambient atmosphere by a foamed polyethylene case. Four copper temperature sensors of $\pm$0.5 °C stated uncertainty were mounted on the cell surface and allowed temperature control of the gas sample inside the cell. Temperature gradients along the cell were estimated to be less than 0.2 °C. 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.

Temperature dependence studies are time consuming, so some optimization of the number of studied lines and temperature points is unavoidable. Transitions for the study were chosen on the basis of a uniformly covered, maximum possible range of rotational quantum numbers limited from the large $|N|$ side by decreasing intensity and therefore insufficient signal-to-noise ratio for accurate study. The total of 12 transitions were selected ranging from 1 to 19.

Measurements of pressure broadening coefficients of oxygen lines were carried out at 12 temperatures in the range from $-$35 °C to $+$75 °C with 10 °C step. At each temperature, spectra were recorded at 8–10 pressures in the interval of 0.3–3.3 Torr. In the case of foreign broadening, partial pressure of oxygen in the mixture was set to be from 0.5 to 1 Torr depending on the studied line intensity. Then foreign gas was gradually added into the cell by steps of 0.3–0.4 Torr and line recording started after achieving the mixture equilibrium (the corresponding time was experimentally estimated to be about 15 min).

3. Data analysis

A typical example of the experimental data is shown in Fig. 1. The signal-to-noise ratio (SNR) for the majority of recordings was about 200–300, reaching 800 for the most intense lines. In quite a few cases (less intensive lines and high temperatures) SNR decreased down to 70. About 13,000 spectrum recordings for 12 oxygen fine structure lines were obtained and analyzed within the frames of the present investigation. The ratio of the Lorentz-to-Doppler width for oxygen lines of the 60-GHz band under the conditions of our experiment was at least 10, therefore the Lorentz profile was used for fitting the experimental spectra. For the 118-GHz line this ratio is twice less and a Voigt profile was used. The noise-like residuals shown in Fig. 2 confirm correspondence of the model function to the experiment. Pressure broadening coefficient was determined as a result of linear regression of the width pressure dependence (see Fig. 2).

One of the main sources of uncertainties in any spectrometer is its baseline originating mainly from the radiation interference effect. In the acoustic detection method the signal is produced by absorbed power only (the so-called “zero level detection”) and interference is efficiently averaged over the cell volume. The baseline is reduced considerably in comparison with the classical video-spectrometers but is still present. A false signal is produced by radiation absorption in windows and wall surfaces of a gas cell [10,11]. The major part of this signal can be removed by a repeated record of the spectrum with replacement of the sample by gas that is not absorbing in this frequency range [3,4]. Its remaining part is taken into account by additive plain terms to the model function. The part of the baseline related to frequency dependence of incident radiation can be taken into account by including a multiplicative term (linear frequency function was shown to be sufficient in most cases [3]) to the model function. The change of the cell position changes the interference pattern and, respectively, its contribution to the output signal. This may give significant systematic uncertainty to the fitting results if the model function is inadequate. In the current study pressure broadening coefficients were measured at several different positions of the cell relative to the radiation source. This allows not only minimizing the influence of standing waves on the retrieved line parameters but also assessing possible baseline-related systematic errors. The final value of the pressure broadening coefficient for each particular line at a given temperature was found by averaging the values obtained from the repeated measurements. The corresponding errors are combinations of statistical uncertainties and a standard deviation σ of the spread of parameters obtained in the repeated measurements. Experimental values of the self-broadening coefficients γ_{self} for the three selected lines are plotted in Fig. 3 as a function of temperature T which is described by the phenomenological power law

$$\gamma(T) = \gamma(T_0)\left(\frac{T_0}{T}\right)^{n_\gamma}$$

$$\lg(\gamma) = \lg(\gamma_0) + n_\gamma \cdot \lg\left(\frac{T_0}{T}\right), \quad (1)$$

where $\gamma_0 = \gamma(T_0)$; T_0 is reference temperature ($T_0 = 296$ K in our case). In total, pressure broadening coefficients $\gamma(296)$ and their temperature exponents n_γ were measured for 12 fine structure lines with the rotational quantum number N ranging from 1 to 19. Results are presented in Table 1 and in Figs. 4 and 5.

Air-broadening parameter $\gamma_{air}(296)$ and its temperature exponent n_γ^{AIR} were calculated for each oxygen line studied using the corresponding self- and nitrogen-broadening

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