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A collisional cooling investigation of the pressure broadening of the $1_{10} \leftarrow 1_{01}$ transition of water from 17 to 200 K

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ABSTRACT

An investigation of the $1_{10} \leftarrow 1_{01}$ rotational transition of water at 556 GHz pressure broadened by hydrogen, helium, nitrogen, oxygen and carbon dioxide has been completed. Using the collisional cooling technique the broadening of this transition by each gas was explored from 200 K down to the condensation point of the broadening gas or 17 K, which ever is lower. This marks the first time such an extensive investigation of the broadening of this rotational transition as a function of temperature has been completed. The results of this investigation will be presented including the exponential temperature dependence of the broadening, the lack of temperature dependence of the broadening observed in helium and the unique behavior of the hydrogen broadening at low temperature (<40 K). In addition, the broadening of the $1_{10} \leftarrow 1_{01}$ rotational transition of water by nitrogen and oxygen was recorded at room temperature and a direct comparison of this data with recent work will be discussed.

1. Introduction

Observing and understanding water in the interstellar medium is an essential and largely unstudied part of the physical conditions that dominate the processes of star and galaxy formation. Due to atmospheric opacity, the spectrum of water in the interstellar medium has been relegated to observations with a few limited balloon [1,2] and space-borne platforms [3–5]. These investigations have focused on bright nearby star formation regions such as the Orion nebula and have shown that water is a key constituent of interstellar molecular clouds. Water acts as a primary coolant and, as such, plays an important role in the evolution of the clouds and the regulation of star formation. In particular, water is very active in the phases leading to star formation [1]. The Herschel Space Observatory [6,7] will have access to a significant number of ortho and para water transitions and will be the first to arrive at a comprehensive picture of water in the interstellar medium.

In such studies, the spectrum of water is employed as a probe of the excitation dynamics and geometry of molecular clouds [6,7]. Interpretation of these observations requires radiative transfer models which utilize state-to-state collision rates of water and hydrogen. The models permit the radiative balance of the gas to be determined allowing structure, dynamics and ultimately the evolution of the molecular cloud to be predicted. Currently, the state-to-state rates are calculated from theory [8–12] and not validated with experiment. To take full advantage of water as an observation tool, the spectrum of water in interstellar environments must be fully understood. The best way to achieve a full understanding of the spectroscopy of water is to turn to laboratory measurements which will facilitate a complete understanding of the interstellar conditions.

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In a recent paper [13], we described a new collisional cooling experiment which has been constructed and implemented at the Jet Propulsion Laboratory (JPL). In this paper, we mentioned that the chief purpose of this system was to study species of astrophysical significance under interstellar conditions. In particular, the intent of the system was to observe gaseous water in an extremely cold environment.

The pressure broadening of the lowest energy transition of the ortho manifold of states ($1_{10} \leftarrow 1_{01}$) at 556 GHz was chosen for the first investigation of water using the new collisional cooling experiment. The broadening of this transition has been investigated previously by two separate research groups. The first study was completed in 1995 by Markov and Krupnov [14] who investigated the pressure broadening and pressure shift of this transition at room temperature. Using a backward wave oscillating (BWO) source and an absorption cell, the broadening and shift coefficients were determined for $1_{10} \leftarrow 1_{01}$ transition of water perturbed by oxygen and nitrogen. This work was followed by a complimentary investigation by Seta et al. [15] who used terahertz time-domain spectroscopy to explore the pressure broadening of the $1_{10} \leftarrow 1_{01}$ and $2_{11} \leftarrow 2_{02}$ transitions of water by nitrogen and oxygen at room temperature. In comparison with the work of Markov and Krupnov, this work found a 17% and 27% difference in the broadening coefficients of the $1_{10} \leftarrow 1_{01}$ transition at room temperature for nitrogen and oxygen, respectively. Recently, in an effort to explain the discrepancies in these two previous results, Golubiatnikov et al. [16] again turned their attention to the $1_{10} \leftarrow 1_{01}$ transition of water. Once more a BWO source was employed to study the pressure broadening and shift of this transition by nitrogen and oxygen at room temperature. The resulting broadening coefficients showed better agreement with the work of Seta et al. than with the other previous work. The discrepancy in the results from the same group was attributed to experimental conditions employed in the 1995 investigation.

This paper presents the results of our investigation of the pressure broadening of the $1_{10} \leftarrow 1_{01}$ transition of water. These results expand the previous work on two fronts. Firstly, the broadening gases studied have been expanded beyond nitrogen and oxygen to include helium, hydrogen and carbon dioxide. Secondly, using the collisional cooling technique the temperature range over which the pressure broadening has been investigated has been extended to cover the region of 17 K to 296 K. The pressure broadening and shift of the $1_{10} \leftarrow 1_{01}$ transition of water by each gas will be discussed as a function of temperature. The current results agree well with the work of Golubiatnikov et al. [16] and a comparison with these and the other previous results will be described.

2. Experimental

In our recent paper [13], we gave a full description of the new collisional cooling experiment at the Jet Propulsion Laboratory (JPL). The same experimental setup was employed in the current investigation of the pressure broadening of water and thus only an overview of the experiment will be presented here. At the core of the experiment is a collisional cooling cell which is housed inside a cryogenic chamber. The cell is cooled, to a low temperature of 17 K, via a helium cryo-refrigeration system. For temperatures above 17 K, the cooling of the refrigeration system is counteracted by heaters affixed to the cell. Using these heaters an experimental range of temperature for the cell of 17 to 200 K is achievable.

Once the cell has reached the desired temperature, it is filled with a fixed amount of broadening gas which quickly thermally equilibrates with the cell. Although the operational temperature range of the system is ~180 K, the actual temperature range varied with buffer gas. For hydrogen and helium, data was recorded over the entire experimental range (17–200 K). However for the other gases, the lowest temperature was restricted by the gases condensation points, 80, 80 and 150 K for oxygen, nitrogen and carbon dioxide, respectively.

When the experimental conditions have been set, water is introduced via an inlet tube and heated nozzle. The newly introduced water quickly thermally equilibrates (~100 collisions) with broadening gas and cell. Water is continuously introduced into the system and continually condenses on to the cell walls. In the time between reaching thermal equilibrium and condensation on the walls, the water spectrum can be recorded. For room temperature measurements, the collisional cooling cell was employed as a static cell with a set amount of water introduced into the system at the beginning of the experiment rather then a continuous flow. This stationary amount of water is then broadened by additions of buffer gas to both the outer cryogenic chamber and the inner collisional cooling cell.

Spectra were recorded in absorption using the multiplied and amplified output of a sweep synthesizer as a source and a Schottky diode detector. This source and detector were the same devices employed and described in full in our recent investigation of the pressure broadening of carbon monoxide [13]. Briefly, the multiplier chain consisted of a $\times 6$ multiplier (Millitech), two monolithic microwave integrated circuit amplifiers [17] and a $\times 2 \times 3$ cascaded multiplier chain (built at JPL). This source operates from 520 to 580 GHz with a maximum output power of 2 mW and a typical output power of 1 mW. In the case of the $1_{10} \leftarrow 1_{01}$ transition of water (556 GHz) the typical output power detected was ~0.5 mW. The detector is a VDI WR 1.5 zero biased Schottky diode detector. Spectra were recorded using the video spectroscopy technique and the signal was processed using a lock-in-amplifier. The spectral line was recorded using either amplitude modulation as a test for saturation or frequency modulation to record pressure broadening spectra.

For each buffer gas a series of water spectra were taken at various pressures and multiple temperatures. As will be explained in the next section, analysis of these successive spectra yielded the pressure broadening coefficient for water for each broadening gas at each temperature. The range of pressures chosen depended on the broadening ability of each buffer gas. In addition, the broadening ability of each gas increases as temperature decreases, such that the range of pressures