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CO_2 absorption coefficients at high pressure and high temperature: measurements and model

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Abstract

In this work we present the CO_2 experimental absorption coefficients recorded at the same conditions found in the deep atmosphere of Venus. Measurements were done according to a real vertical profile, varying the pressure from 1 to 30 bar and the temperature from 298 to 600 K. Each spectrum was acquired with a resolution of 2 cm^{-1}. The data has been compared with synthetic spectra obtained by taking into account the line mixing effect.

1. Introduction

The scientific goals related to this work are two: implement spectroscopic databases and support the space missions. Presently a wealth of spectroscopic data are present in several databases, such as HIgh-resolution- TRANsmission (HITRAN [1]), HIgh-TEMPerature (HITEMP) [2] Carbon-Dioxide-Spectroscopy-Databank (CDSD [3]). They provides a suite of parameters of molecular species at typically terrestrial conditions and are routinely used for the retrieval of many parameters concerning the atmosphere of the Earth. On the other hand, they do not or contain very limited information on the behavior of gases at extreme conditions, in particular, high pressure and high temperature.
These parameters are of major importance for the radiative transfer models of data coming from space mission, like the VIRTIS (Visible and InfraRed Thermal Imaging Spectrometer) instrument on board the ESA mission Venus Express [4]. In order to test the ability of sophisticated models in reproducing transmission spectra in Venus' conditions, we've built up an experimental setup to measure spectra for the characterization of the optical properties of CO2. The data recorded has been compared with a spectra simulation which takes into account the lines interferences due to inter-molecular collisions (or line-mixing), which have a major effect on the spectral shape.

2. Results

The experimental setup consists of a Fourier Transform InfraRed (FT-IT) interferometer able to work in a wide spectral range, from 350 to 25000 cm^{-1} (0.4 to 29 μm) with a relatively high spectral resolution, from 10 to 0.07 cm^{-1}. A special customized gas cell, designed to support pressure up to 350 bar and temperature up to 300 °C has been integrated inside the interferometer. To recreate the same conditions found in the deep atmosphere of Venus, we varied the CO_2 pressure and temperature according to a vertical profile measured by the Venera probes [5]. The experimental carbon dioxide absorption coefficients, recorded with a resolution of 2 cm^{-1}, reproduces the physical conditions from an altitude of 50 Km down to 16 km. In figure 1 we show some spectra acquired at different pressure and temperature. Measurements were compared with synthetic spectra obtained by a software, named Line Mixing Model (LMM) [6], that takes into account line-mixing effects for all bands present in the spectral region considered (from 800 to 10000 cm^{-1}), figure 2. This theoretical model was used also to calculate absorption in far wing region where absorption results essentially from contribution of far lines**.** For this region , χ correction factors have been determined to model the spectral shape. As additional effect, during our measurement campaign, we observed some CO_2 bands forbidden for the symmetric $^{16}O^{12}C^{16}O$ molecule. Even when the pressure is only a few bar, the interaction of the molecules with their neighbours induces indeed a dipole moment which yields to pressure-induced-absorption bands. The observed Collisional Induced Absorption (CIA) bands integrated intensities show a quadratic dependence versus density and depend on the temperature.

3. Figures

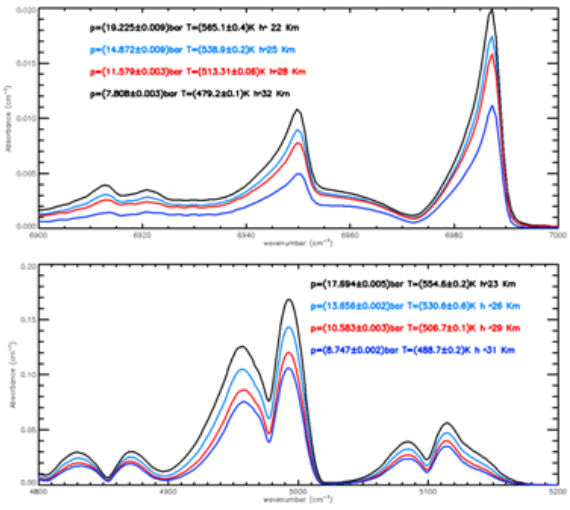

Figure1: CO_2 absorption coefficients measured at different pressure and temperature.

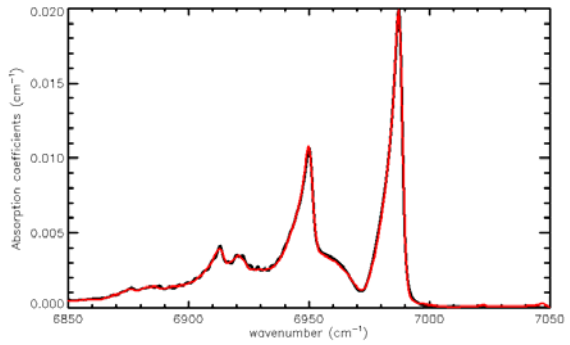

Figure 2: Comparison between Measured (black curve) and simulated (red curve) at p~19 bar and T~ 565 K

4. Conclusion

In light of the results presented in this work, many experimental data are now available obtained at real vertical venusian physical conditions, especially at low altitude, for a wide range of temperature, pressure and spectral domain (from 750 to 10000 cm-1). The comparison between measures and simulated spectra leads us to conclude that the new tool of simulation reproduces the data with an integral deviation better than 6% on the full range presently explored of the venusian deep atmosphere. Contrary to the widely used Lorentz shape, the LMM leads to have a good agreement with the experimental data in the central and near wing regions of the vibrational bands. For the far wings region, sets of temperature dependent χ factors have been constructed for the $\nu 2$, $\nu 3$ and $\nu 1+\nu 3$ bands wing regions.
Concerning Collision-Induced Absorption (CIA), it's found to be proportional to the square of the density, decreases with temperature, consistently with the collisional nature of this phenomenon.
More work has to be exploited in this direction in order to include this effect in the radiative transfer models and calculation.

References

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