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High pressure/high temperature spectra of CO_2; measured and simulated data

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Abstract

We have built up an experimental set-up to study the optical properties of gases at extreme conditions. A Fourier Transform InfraRed (FT-IR) interferometer has been integrated with a special customized high pressure-high temperature (HP-HT) gas cell and the system has been employed to measure CO_2 optical transmittance at the conditions typically found in the deep atmosphere of Venus. This set-up is able to work in a wide spectral range, from 350 to 25000cm^{-1} (0.4 to 29 μm), with a relatively high spectral resolution, from 10 to 0.07 cm^{-1}. The HP-HT gas cell is able to work at a pressure up to about 50 bar (real limitation being the CO_2 source) and a temperature up to 350°C. Measurements were done varying the pressure from 1 to 50 bar and the temperature from 298 to 550 K.
Measurements have been compared with synthetic spectra obtained using two different models. In our work we have also studied the Collision Induced Absorption (CIA) effect, which is due to the dipole moment induced by collision processes.
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1. Introduction

The modelling of the transmittance of gases at extreme conditions, in particular high pressure and high temperature, in some cases is not satisfactory as one would need for an accurate determination of the physical-chemical parameters in deep atmospheres. To better interpret the data coming from the remote sensing instruments flying in space, like Venus Express, we have built up a set-up in our laboratory to measure the optical properties of gases, in particular CO_2, at high pressure and/or high temperature. The CO_2 spectroscopic data available in the HITRAN, HITEMP and CDSD databases, are often not sufficient to simulate the optical properties of CO_2 in extreme conditions.

2. Results

The CO_2 transmittance measurement was done varying the pressure from 1 to 50 bar and the temperature from 298 to 550 K, similar to the Venusian deep atmospheric conditions (see figure 1). Measurements are compared with synthetic spectra obtained using two different models (see figure 2). The first is a straightforward radiative transfer program based on line by line calculation "ARS" [1]. The second, ''Solution'' [2],[3], takes into account several mechanisms which have an impact on the shape of the spectral lines; the so-called "line mixing", occurring when individual rotational levels interact, and the dense, short-path lengths intermolecular collisions, which change the lineshapes concerning the far wings of spectral lines. In particular, the absorption in the far wings of strong ro-vibrational bands at a few hundred cm^{-1} from the band center, is 10^3-10^4 times higher than predicted by the Lorentz profile. Inclusion of these mechanisms in the Solution program improves the agreement between measured and simulated data by about 4%. for pressures larger than 10 bar.

Another interesting phenomenon, occurring at high pressure is the collision induced absorption (CIA). Absorption bands which are strictly forbidden, having no electrical transition dipole moment, become weakly active due to collisionally induced dipole moments. Several CIA bands have been observed , some at relatively low pressures (a few bars), for the symmetric $^{16}O^{12}C^{16}O$ molecule. The integrated intensities of the observed CIA bands (see figure 3), show a quadratic dependence versus density .

2. Figures

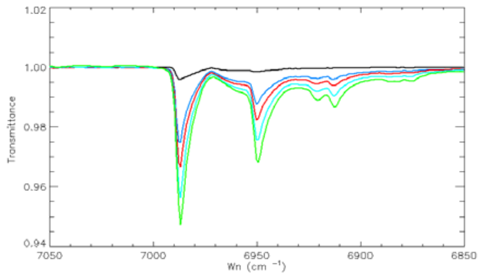

Figure 1:CO_2 transmittance in the Near InfraRed spectral range at different pressure and temperature

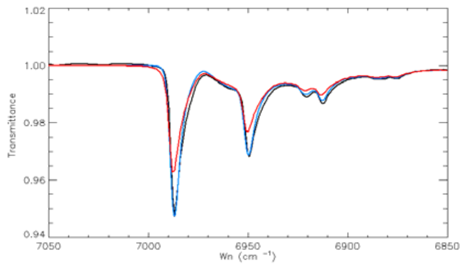

Figure 2:Comparison between measured (black curve) and simulated data. The green and red curves are the syntethic spectral obtained by ARS code and Solution software respectively

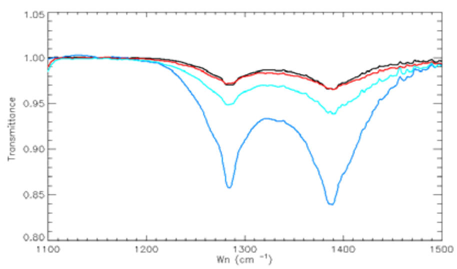

Figure 3: Fig.3 Bands of Collision Induced Absorption observed at differente pressure and temperature

Conclusion

First results of the CO_2 transmittance using real Venusian P-T atmospheric profiles have been obtained with a relatively high spectral resolution. A preliminary comparison leads us to conclude that in the real gases under pressure higher than only a few bars, the shape of the spectral lines no longer follows the conventional Voigt form**.** A remarkable agreement between measurements and synthetic spectra based on the strong collision approximation provides a strong argument in favour of this relatively simple line mixing model. An aspect which is probably underestimated in typical remote sensing methods of radiative transfer inverse calculation is the CIA effect, which has to be more deeply studied and modelled. The impact of CIA is currently under study.

References

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