New experimental measurements of the Collision-Induced Absorption of H_2–H_2 and H_2-He in the 3600 - 5500 cm^{-1} spectral range from 120 to 500 K

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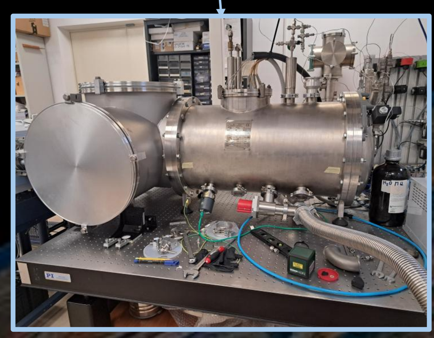
EXPERIMENTAL SETUP



High-resolution Fourier spectrometer
- Maximum resolution = 0,002 cm^{-1}

Coupled with

Simulation chamber called PASSxS (Planetary Atmosphere System Simulation x Spectroscopy)
- Maximum pressure = 70 bar
- Temperature range → [100, 550] K
- Optical path in vacuum
- MP cell aligned to reach an optical path of 3.27 m

Thanks to this experimental setup, it is possible to measure the transmittance of a desired gas mixture, according to the Lambert-Beer law.

$$T = \frac{I(\nu)}{I(0)} = e^{-kL}$$

- $I(\nu)$: Radiance measured with the cell filled with gas
- $I(0)$: Radiance measured with the cell empty
- L: Optical path
- k: Absorption coefficients

EXPERIMENTAL MEASUREMENTS

Previous experimental measurements of the H_2-H_2 and H_2-He binary absorption coefficients (Vitali et al., 2025, JQSRT, 330) performed in the [3600, 5500] cm^{-1} spectral range, revealed the presence of the so-called *interference dips*, which are not reproduced by CIA model simulations. Those represent a lack of absorption at specific frequencies.

To study their behavior with density, measurements at a resolution of 0,05 cm^{-1} have been performed at a fixed temperature and various pressures.

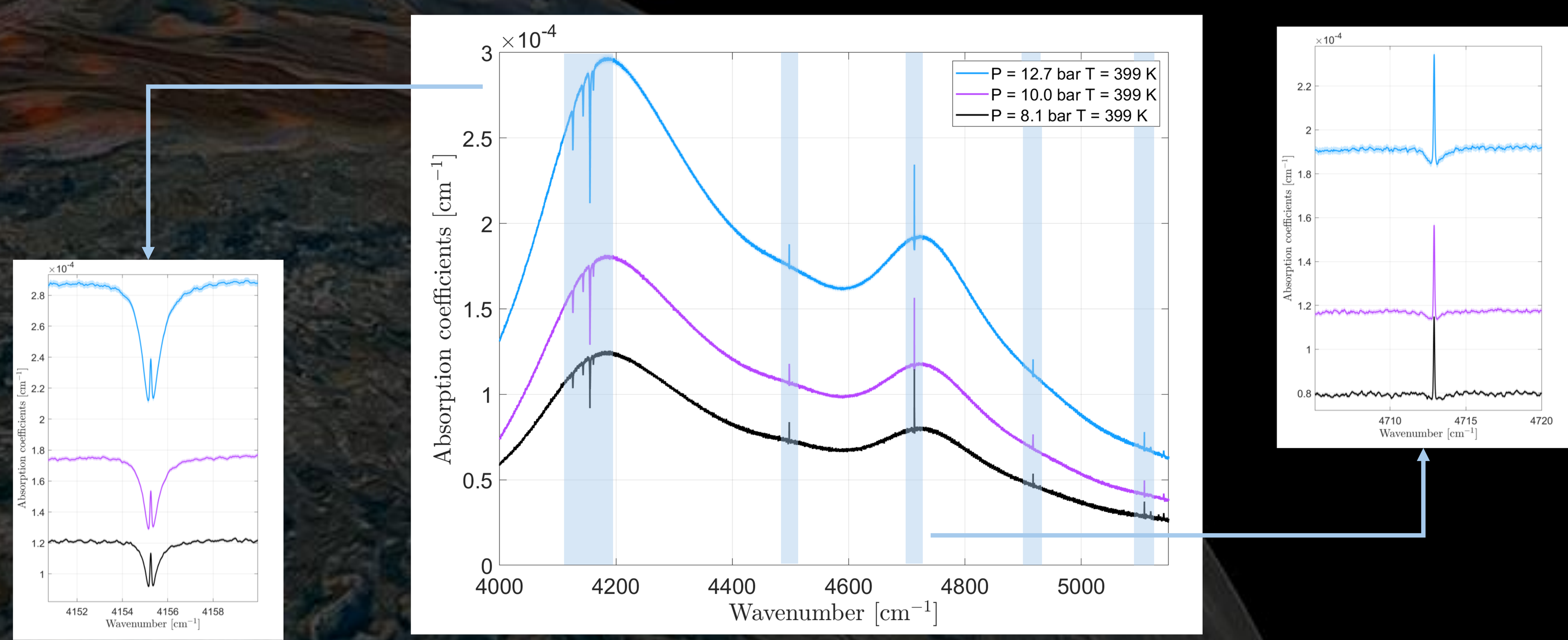


Figure 2: Experimental absorption coefficients measured at 399 K for three different pressures.

The interference dips have been observed at various frequencies (highlighted by the blue rectangles in Figure 2). The most evident ones rely on the main peak of the band. However, they are also present around 4500 cm^{-1}, 4700 cm^{-1}, 4900 cm^{-1}, and 5100 cm^{-1}, but are difficult to see because of the superimposition of some sharp absorption lines due to the H_2 quadrupolar transitions, present approximately at the center of the dips except that at 4161 cm^{-1}. The quadrupole absorption lines are very weak transitions that become observable due to the high density and the Dicke-narrowing occurring at high pressures. They vary linearly with the density, while CIA has a quadratic dependence on the density.

WHAT IS CIA?

The Collision-Induced Absorption lines are generated from the collision between two molecules or a molecule and an atom in a high-density environment. CIA of H_2 represents one of the main sources of opacity of the atmosphere of the gaseous giants mainly composed of H_2 and He, in the infrared part of the spectrum, particularly between 1 and 5 µm. In this spectral range, the CIA fundamental band of H_2 and its features have been experimentally investigated. Here we focus on the so-called interference dips.

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THE INTERFERENCE DIPS...

The phenomenon generating the interference dips has been previously investigated by Van Kranendonk (Van Kranendonk,1968, Can. J. Phys., 46,1173). They are caused by the interference of induced dipole moments in consecutive collisions.
Van Kranendonk calculated a theoretical profile to describe their shape in function of the intracollisional halfwidth δ and the frequency of the dip's peak ν_c.

Depending on its theory, δ is thought to follow a linear trend with density.

However, since an asymmetry of the main peak of the dip has been observed by Kelley and Bragg (Kelley and Bragg, 1984, Phys. Rev. A, 29), starting from the Van Kranendonk profile, they developed an asymmetric profile for the dips, by adding a phase α.

$$F(\nu) = \frac{1-\cos\alpha+\left(\frac{\nu-\nu_c}{\delta}\right)^2-\left(\frac{\nu-\nu_c}{\delta}\right)\sin\alpha}{1+\left(\frac{\nu-\nu_c}{\delta}\right)^2}$$

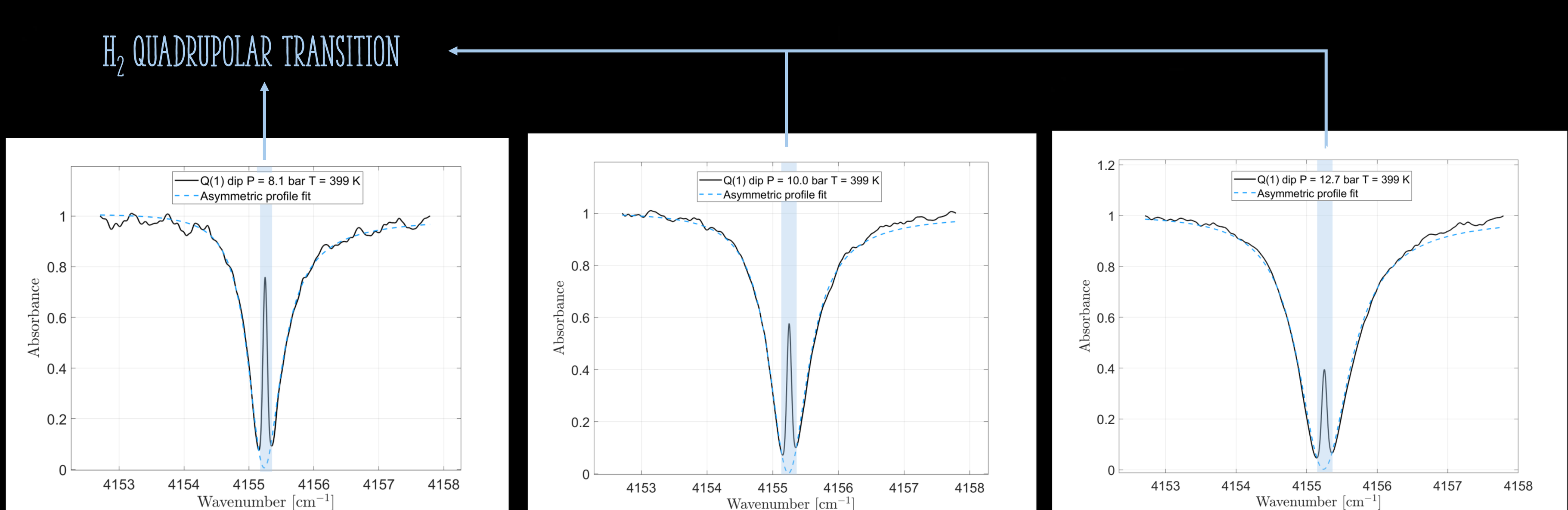

Figure 3: From left to right, the Q(1) interference dip (black solid line) measured at 399 k for an increasing pressure. The light blue dotted line represents the fit made with the asymmetric profile by J. D. Kelley and S. L. Bragg

...AND THEIR BEHAVIOR

The interference dips have been already observed in previous experimental works, but they have never been investigated at temperatures above 300 K. In this work the Q(1) experimental dip, at all the densities considered for the three temperatures investigated, 305 K, 399 K and 499 K, has been fitted with the asymmetric profile. Figure 3 shows the fit performed over the dips measured at 399 K at three different pressures.
Here, the dips have been normalized for their intensity, so that the absorbance shows a value between 0 and 1.

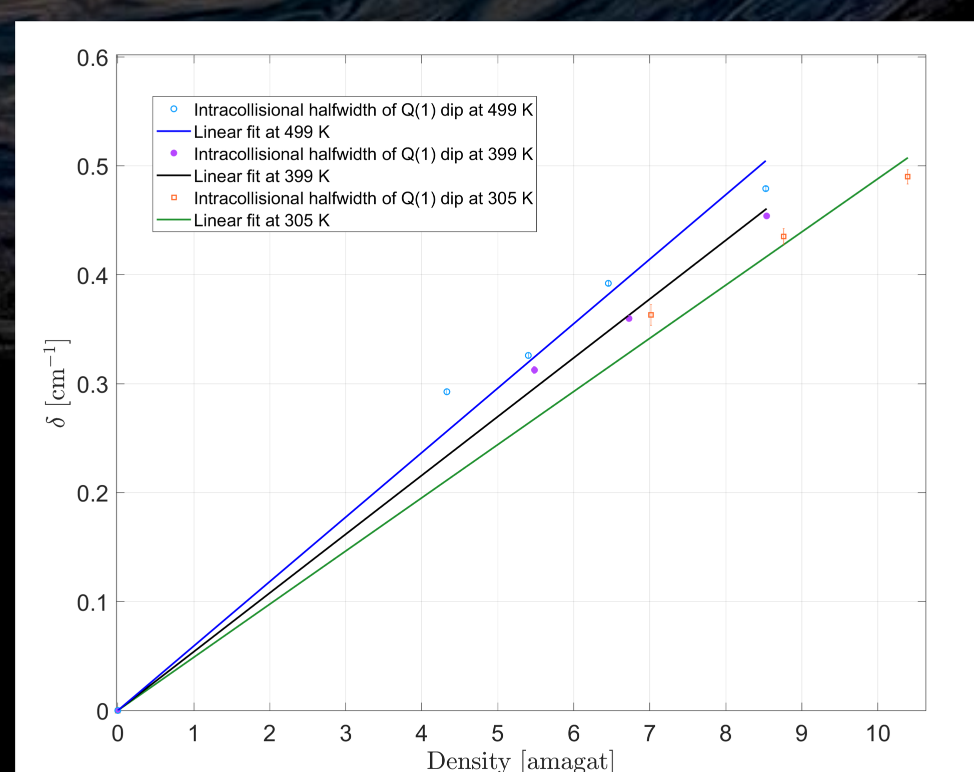

Figure 4: Behavior of the intracollisional halfwidth (δ) with density for the three temperatures considered

T [K]	m [cm^{-1} $amagat^{-1}$]
305,141 ± 0,017	0,0488 ± 0,0055
399,154 ± 0,003	0.0540 ± 0,0044
498,756 ± 0,005	0,0592 ± 0,0064

Table 1: Values of the angular coefficients (m) of the linear fits shown in Fig. 3

For all the temperatures, a linear behavior of δ with respect to the density has been found, according to Van Kranendonk's theory, as can be seen in Figure 4.
The angular coefficients obtained from the fits are reported in Table 1. As one can see from the table, the angular coefficients seem to follow a linear trend with the temperature