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## Experimental observation of the intercollisional interference effect on the $S_1$ pure rotation line of the collision induced spectra of the $H_2$ -He mixture (\*)

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**Résumé.** — On présente les coefficients d'absorption du spectre correspondant à l'interaction  $H_2$ -He à 300 K entre 200 et  $700\text{ cm}^{-1}$ . Un défaut d'absorption ou « dip » caractéristique des effets d'interférences collisionnelles est clairement visible à la fréquence de la raie rotationnelle  $S_1$  de la molécule libre  $H_2$ .

**Abstract.** — The absorption coefficients of the collision induced spectrum of the interaction  $H_2$ -He at 300 K between 200 and  $700\text{ cm}^{-1}$  are presented. A lack of absorption (dip) due to the intercollisional interference effect is clearly seen at the frequency of the pure rotation line  $S_1$  of the free molecule of  $H_2$ .

The collisional induced spectra of  $H_2$  and He- $H_2$  mixture have been the object of many experimental investigations [1-10] because they enable the collisional mechanisms to be studied: double transitions, overlaps, intercollisional interference effects; and also because they can be used to determine the composition of some planetary atmospheres.

Most of these investigations have been performed in the near infrared at about  $4\,500\text{ cm}^{-1}$  on the vibration rotational spectra of the  $H_2$  molecule. These investigations have shown the intercollisional interference effects which are dips around the rotational-vibrational frequency of the absorber. The theoretical explanation of these phenomena has been given by Van Kranendonk [11]; they are due to the fact that an effect of a collision is partly destroyed by the next one.

Up to now, the far infrared investigations [5, 7, 10] have been realized at too low resolutions ( $\approx 10\text{ cm}^{-1}$ ) to evidence the dips on the pure rotation spectra and the absorption coefficients of the  $H_2$ -He spectra derived from them are too inaccurate for the reconstitution of the planetary atmosphere spectra.

Moreover, the Jovian atmosphere spectra taken by Voyager I presents yet unexplained features [12] at  $354$  and  $587\text{ cm}^{-1}$  (transition frequencies of the free rotation of  $H_2$ : lines  $S_0$  and  $S_1$ ).

Considering these facts, we have begun a study of the  $H_2$ -He mixture spectra in the far infrared ( $200$ - $700\text{ cm}^{-1}$ ) with a higher resolution so as to obtain a better accuracy in the absorption

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coefficients and to show, in this spectral region, the lack of absorption due to the interferences between the different collisions which may explain the features of the Voyager I spectra.

1. **Experimental procedure.** — To obtain the absorption coefficient  $\alpha_{12}$  for the H<sub>2</sub>-He interaction alone, the contribution of the interaction H<sub>2</sub>-H<sub>2</sub> must be subtracted from the  $\alpha_m$  coefficient of the mixture experimentally determined by :

$$\alpha_m(\nu) = \frac{1}{Ld^2} \text{Ln} \frac{I_0(\nu)}{I_m(\nu)},$$

where

$\nu$  is the frequency

$L$ , the length of the absorption path

$d$ , the density of the mixture

$I_m$ , the intensity reaching the detector, the cell filled up with the mixture

$I_0$ , the intensity reaching the detector, the cell empty.

For this subtraction, the  $\alpha_{11}$  coefficients of the self-induced spectra of H<sub>2</sub> (experimental data) are used in the formula :

$$\alpha_{12} = \frac{\alpha_m - \chi^2 \alpha_{11}}{\chi^{(1-\chi)}},$$

with  $\chi$  = density of H<sub>2</sub>/density of the mixture.

In some parts of the spectra, especially around the S<sub>0</sub> and S<sub>1</sub> lines,  $\alpha_m$  and  $\chi^2 \alpha_{11}$  are very close together so that any inaccuracy in their determination leads to a great inaccuracy in  $\alpha_{12}$ .

In order to obtain a better accuracy of the data, we have not used the  $\alpha_{11}$  coefficients but we have replaced the reference spectra (empty cell) by that of the cell filled with hydrogen at the same pressure as the partial pressure of this constituent in the mixture. In these conditions :

$$\alpha_{12} = \frac{1}{d^2 L(1-\chi)\chi} \text{Ln} \frac{I_{H_2}}{I_m}.$$

Thus the noises and errors from two more reference spectra are avoided. The density of the mixture and  $\chi$  can be chosen so that  $I_m \in [0.2 I_{H_2}, 0.6 I_{He}]$  in order to obtain the best accuracy on  $\alpha_{12}$  [7].

2. **Apparatus.** — The instrument used is the Fourier Transform Spectro-interferometer described in [13] with some improvements.

— A three meter long pressure cell made of stainless steel has been added. This cell has a vacuum insulated cryostat. The inner surface of the pressure cell is lined with an aluminium pipe which conducts light. The windows are ICs crystals (12 mm thick ; free diameter : 10 mm). They are glued on their mounts which are themselves tightened on the body cell with indium gaskets.

A relatively fast scan of the interferogram has been adopted. It avoids the use of a mechanical chopper which always causes noise. This method provides the opportunity of taking several interferograms in a short time and of calculating a statistical average, thus improving the accuracy of the results. The speed of the scan has been chosen experimentally so as to obtain the best response of the detector at the top of the interferogram.

A micro computer (Zylog) has been connected with the instrument and controls it. Its numerous functions include the following :

1) Each time a fringe travels in the helium-neon laser interferometer which supervises the moving mirror, a sample is taken and put into a memory field looped on itself.

2) The nearest fringe of the zero path difference is identified by locating the maximum of the interferogram.

3) From this fringe, a sample is taken every step,  $h$ ; this step, imposed by the optic (beam splitter, filter, source), is measured by counting the corresponding number of fringes.

4) When the necessary amount of samples is reached, the samples, corresponding to the series whose elements are spaced from one another by the step  $h$  and which begins at the maximum of the interferogram, are extracted from the memory field. In this way the two-sided interferogram is rebuilt.

The four operations are repeated  $n$  times so as to obtain two sums of  $n/2$  interferograms each one corresponding to a travelling direction of the moving mirror.

The two lots of data are constituted both for the sample (H<sub>2</sub>-He) and for the reference (H<sub>2</sub>). A fast Fourier transform (Delouis algorithm [14] for a complex function) is effected on each lot of data; the absorption coefficients are computed and only after this step, the average of the results, corresponding to both directions of the movement of the mirror, is taken.

All the results (interferograms, spectra after fast Fourier transform, absorption coefficients) can be reproduced on a XY analogic plotter. For purposes of rapidity all the computer programs involved have been written in Z 80 assembly language.

3. **Experimental conditions.** — All the gases and mixtures used (15.6 % of H<sub>2</sub>) are Air Gas ultra high purity grade (6.0). To remove the residual H<sub>2</sub>O the gases pass through molecular sieves (Union Carbide type 13X) previously heated at 600 K and cooled at 77 K just during the flowing.

4. **Results.** — The normalized absorption coefficients  $\alpha_{12}/d^2$ , in absolute units, for the H<sub>2</sub>-He interaction at 300 K are shown in figure 1 (average of 40 interferograms for the sample and for the reference). The results are smoothed using a least square procedure (four degrees polynomial on nine points [15]).

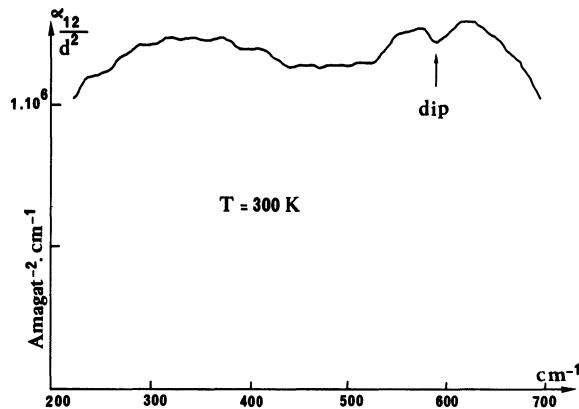


Fig. 1. — Absorption coefficient of the H<sub>2</sub>-He interaction spectrum.

A lack of absorption is clearly seen at  $587 \text{ cm}^{-1}$ . The enlargement of this dip is shown in figure 2. Its amplitude represents 6 % of the S<sub>1</sub> line; its half line width at half height is  $10 \text{ cm}^{-1}$  (density of the mixture : 109 Amagats). So this phenomenon is rather small and it does not seem possible to put it forward to explain the Jovian spectra features for the following reasons :

- the Jovian atmosphere is 90 % H<sub>2</sub>-10 % He ;
- the Voyager Spectra have been taken in the upper atmospheric layers where the total

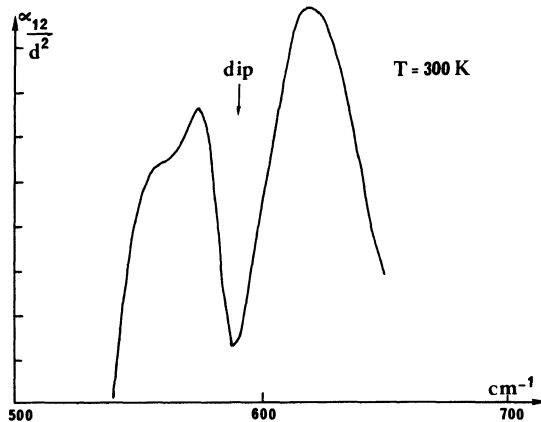


Fig. 2. — Dip of absorption due to the intercollisional interference effect.

pressure is about 200 mbar; therefore the dip should have a half width of  $0.01 \text{ cm}^{-1}$  since the dip width is proportional to the density of the mixture whereas the absorption coefficients are proportional to the square density;

— the resolution of the Voyager interferometer was  $3.5 \text{ cm}^{-1}$ : the shape of the dip is convoluted by an instrument response function one hundred times wider.

As the features shown are too big to be due to the interference effects between successive collisions  $\text{H}_2\text{-He}$ , their origin may be associated with the pure rotation spectra of  $\text{H}_2$ .

Our record of this spectra (cell of 5 cm, pressure of 150 bars, 300 K) does not show an effect as marked as in  $\text{H}_2\text{-He}$  spectra. There seems to exist a small depression (1 %) at about  $580 \text{ cm}^{-1}$  but it is not clear enough to be considered as an experimental proof of the reality of a dip.

In figure 1, a « dip » is not observed at the position of the  $S_0$  line, probably because of the weakness of this line comparatively with the  $S_1$  line, at room temperature.

**5. Conclusion.** — For the first time, at  $587 \text{ cm}^{-1}$ , transition frequency of the pure rotation of  $\text{H}_2$ , a dip is shown which is due to the interference effect between the successive collisions of  $\text{H}_2$  and He. But its low amplitude suggests that it cannot explain the feature observed at the same frequency on the Voyager's Jovian spectra. A search for the relationship between spectral parameters and temperature is now in progress.

### References

- [1] KISS, Z. J., GUSH, H. P., WELSH, H. L., *Can. J. Phys.* **37** (1959) 362.
- [2] COULON, R., Thèse, Paris (1958).
- [3] CHISHOLM, D. A., WELSH, H. L., *Can. J. Phys.* **32** (1954) 291.
- [4] MACTAGGART, J. W., WELSH, H. L., *Can. J. Phys.* **51** (1973) 158.
- [5] MACTAGGART, J. W., HUNT, J. L., *Can. J. Phys.* **47** (1969) 65.
- [6] POLL, J. P., HUNT, J. L., MACTAGGART, J. W., *Can. J. Phys.* **53** (1975) 954.
- [7] BIRNBAUM, G., *J. Quant. Spectrosc. Radiat. Transfer* **19** (1978) 51.
- [8] PADDI REDDY, S., VARGHESE, G., PRASAD, R. D. G., *Phys. Rev.* **15** n° 3 (1977) 975.
- [9] SEN, A., PRASAD, R. D. G., PADDI REDDY, S., *J. Chem. Phys.* **72** n° 3 (1980) 1713.
- [10] BACHET, G., COHEN, E. R., DORE, P., BIRNBAUM, G., *Can. J. Phys.* (à paraître).

- [11] VAN KRANENDONK, *Proceeding of international school of physics Enrico FERMI, Varenna (1978); Intermolecular spectroscopy and dynamical properties of dense systems* (North Holland Publishing Company) 1980.
- [12] HANEL, R., CONRATH, B., FLASAR, M., KUNDE, V., LOWMAN, P., MAGUIRE, M., PEARL, J., PINAGLIA, J., SAMUELSON, R., GAUTIER, D., GIERASCH, P., KUMAR, S., PONNAMPERUMA, C., *Science* **204** (1978) 979.
- [13] BACHET, G., COULON, R., *Infrared Phys.* **17** (1977) 359.
- [14] DELOUIS, H., Thèse, Paris (1973).
- [15] MINEUR, H., *Technique de Calcul numérique* (Librairie Polytechnique Ch. Béranger, Paris) 1952.
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