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On the hypothesis of H_2 as the carrier of the diffuse interstellar bands

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Abstract

In a laboratory experiment, employing an extreme ultraviolet/near-infrared double resonance technique, a recent hypothesis by Sorokin and Glownia on the carrier of the diffuse interstellar bands is tested. Quantum states $C\,^1\Pi_u^+$, $v=9$, $J=1$–3 of molecular hydrogen were populated by an extreme ultraviolet laser at 86 nm and used as absorber states for a second excitation step. In transitions to autoionizing doubly excited states of H_2 many resonances of strongly varying widths were observed. The frequency positions and linewidths are in general not consistent with the observed diffuse interstellar bands in the range 766–788 nm, nor with the predictions of Sorokin and Glownia. However, two lines are found in the present study that do coincide with diffuse interstellar bands.

1. Introduction

The mysterious 'diffuse interstellar bands' (DIBs) have intrigued and challenged spectroscopists and astronomers over the last 50 years. Along the line-of-sight of stars of various types more than 200 absorption features, in widths varying from 1 to 100 cm^{-1}, have been observed. The origin of these absorptions is disputed and the only point of agreement is that the carriers of these mysterious phenomena are species present in the interstellar medium. Even the question whether these unknown species are gas-phase molecules or atoms/molecules in or at surfaces of dust grains has not been resolved. In the past several gas-phase molecules have been proposed to cause the DIB absorptions: linear carbon chains [1], porphyrin molecules in general [2] and the chlorin molecule ($C_{20}H_{10}N_4$) in particular [3], polycyclic aromatic hydrocarbon molecules [4], free carbene molecules [5] and the C_{60}^+ ion [6].

Recently a model was proposed in which excited molecular hydrogen is predicted to be the carrier of the DIBs [7]. This hypothesis is particularly appealing, as H_2 is the most abundant molecule in interstellar space and no intricate chemistry is implied in the explanatory model. The hypothesis involves extreme ultraviolet pumping of H_2 and subsequent decay into high vibrational states in the $X\,^1\Sigma_g^+$ electronic ground state. Assuming a width of 50 cm^{-1}, Lyman-α radiation at $\lambda = 121.6$ nm is resonant in the $C\,^1\Pi_u$–$X\,^1\Sigma_g^+$ (9, 11) band for R(0)–R(3) lines. Thus the Lyman-α absorption acts as a state-selector that determines which $X\,^1\Sigma_g^+$, v'' levels are further excited and which highly excited states in H_2 are reached by this mechanism. The DIB features are predicted to be related to absorptions from the $C\,^1\Pi_u^+$,

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$v=9$, $J=1$–4 states. An important piece of support for the H_2 hypothesis is the frequency separations between observed DIB features that are found to coincide with known combination differences between P($J+2$) and R(J) states originating in the $C\,^1\Pi_u$ state. Since these states are located at 115000 cm^{-1}, by subsequent absorption of a single photon in the visible or near-infrared range doubly excited states of H_2 are reached that interact with dissociation and/or autoionizing continua. This coupling to continuum states may explain the strong variations in width as observed for the DIBs. Based on this model Sorokin and Glownia [7] have interpreted all twelve DIB phenomena within the interval 766–788 nm as transitions originating in excited H_2.

In the present work we have tested these predictions in a laboratory experiment. Quantum states $C\,^1\Pi_u^+$, $v=9$, $J=1$–3 are selectively prepared with a tunable XUV laser at 86 nm. Doubly excited states are probed with a second tunable laser in the wavelength range 766–788 nm. Because H_2^+ ions are detected, specifically the doubly excited states that couple to autoionization continua are recorded. The conclusion of the present work is that the overall spectroscopic structures observed do not coincide with the DIBs in this wavelength range. However in two cases features are observed that agree within error bars with known DIB absorptions.

2. Experimental

A tunable and narrowband XUV-radiation source is used for the state-selective preparation of single quantum states $C\,^1\Pi_u^+$, $v=9$, $J=1$–3 of H_2. Previously this XUV laser has been employed for high-resolution spectroscopic studies of the $B\,^1\Sigma_u^+$–$X\,^1\Sigma_g^+$ Lyman and $C\,^1\Pi_u$–$X\,^1\Sigma_g^+$ Werner bands systems in the range 91–98 nm [8,9]. Tunable radiation near 86 nm is generated via frequency tripling the output (7 mJ/pulse) of a frequency-doubled tunable dye laser at 260 nm in a xenon gas jet. The overlapping (spatially and temporally) XUV beam and the UV-fundamental perpendicularly cross a pulsed molecular beam, which enters through a skimmer into the interaction region. The H_2 density in the interaction zone is about 10^{-5} Torr. R-branch transitions of the C–X (9, 0) band are recorded via 1 XUV + 1 UV photionization. Ions are produced in a field-free zone and extracted by a pulsed electric field switched on 50 ns after the laser pulses. Both H_2^+ and H^+ resulting ions are detected on an electron multiplier after mass-separation in a time-of-flight zone.

For the recording of two-step excitation spectra the XUV wavelength is set on one of the transitions R(0), R(1) or R(2). A laser beam, obtained from a dye-laser operating on a mixture of styryl-8/9 dyes and tunable in the range 750–820 nm, enters the interaction zone counterpropagating the XUV/UV beams. The laser beams, with durations 3–5 ns, are also temporally overlapped in view of the short lifetime of the $C\,^1\Pi_u$ state (≈ 1 ns). Spectra of doubly excited states of H_2 are detected via enhancement of the H_2^+ signals. A major disadvantage of this technique is the strong background of the XUV + UV photoionization that gives rise to a noisy baseline. XUV and UV beams diverge into the interaction zone covering a diameter of 8 mm. The power density of the red laser beam in the interaction zone is ≈ 10–15 mJ/cm^2 in 5 ns pulses. Its bandwidth is 0.15 cm^{-1} or 0.1 Å. At 760 nm the wavelength was accurately calibrated by an echelle-grating monochromator, but at longer wavelengths extrapolation procedures had to be used yielding a calibration uncertainty of 0.4 Å for the wavelength readout. The uncertainty in the determination of resonances is further increased by the noise and broadness of the spectral resonances.

3. Observations

In a first step H_2 excitation spectra were recorded in the range near 86 nm. As an example a high-resolution spectrum of the R(0) line of the $C\,^1\Pi_u$–$X\,^1\Sigma_g^+$ (9, 0) band is shown in Fig. 1. Because of the crossed-beam configuration a sub-Doppler resolution of 0.4 cm^{-1} is obtained. The radiation at the fundamental wavelength of the first dye laser at $\lambda = 521$ nm is used for a simultaneous recording of an I_2 absorption spectrum, which allows for an accuracy in the determination of H_2 resonances within 0.15 cm^{-1}. Many narrow resonances were observed in the XUV-excitation scans that could all be assigned to lines in the $B\,^1\Sigma_u^+$–$X\,^1\Sigma_g^+$, $C\,^1\Pi_u$–$X\,^1\Sigma_g^+$, $B''\,^1\Sigma_u^+$–$X\,^1\Sigma_g^+$ and $D^1\Pi_u$–$X\,^1\Sigma_g^+$ systems. Similar XUV-

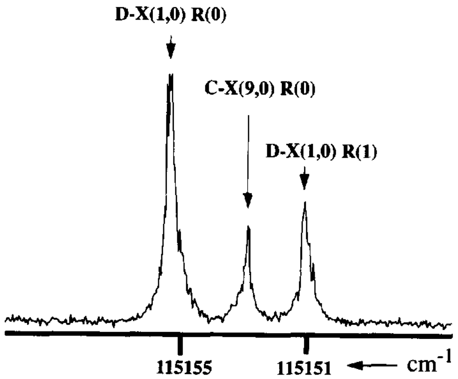

Fig. 1. XUV-excitation spectrum of H_2 in the wavelength range near 86 nm recorded in a crossed beam configuration. Detection of H_2^+-ions was performed upon 1 XUV + 1 UV photoionization.

excitation spectra were taken near $\lambda = 84.3$ nm, where lines of the $B'' \, ^1\Sigma_u^+ - X \, ^1\Sigma_g^+$ (7, 0) band were recorded and assigned by comparison with the analysis of Namioka [10].

Because the explanatory model of Sorokin and Glownia involves $C \, ^1\Pi_u^+$, $v = 9$, $J = 1–4$ states as the ground state levels for the DIB absorptions, we subsequently set the XUV laser to each of the R(0), R(1) and R(2) lines of the C–X (9, 0) band. Unfortunately the R(3) line was too weak to be observed in our molecular beam expansion, so the $J = 4$ state could not be used as an intermediate. In the second excitation step the near-infrared laser was scanned in the range 750–820 nm. In Fig. 2 the resulting spectra as obtained for $J = 1–3$ intermediate states are displayed in the specific wavelength range discussed by Sorokin and Glownia. Wavelength positions, uncertainties as well as widths of the most pronounced resonance features in the range 766–788 nm are determined and listed in Table 1. Throughout the entire wavelength range 750–820 nm covered by the tuning curve of the dye laser many more pronounced features were observed.

The observed spectra do not reproduce the DIB spectrum in the range 766–788 nm as given in the recent overview by Jenniskens and Désert [11] and for which an assignment was proposed by Sorokin and Glownia [7]. However, coincidences between listed DIB phenomena [11] and the recorded resonances are found. The feature observed at 7783.7 ± 1.0 Å (No. 3 in Fig. 2) with a width of 10 ± 4 Å might correspond to the DIB feature at 7782.2 ± 1.6 Å with a width of 3.6 ± 0.9 Å. At 7694.7 ± 0.5 Å a Fano-type line profile is observed (No. 5 in Fig. 2). By deconvolution with a Fano-profile a width of 1.6 ± 0.2 Å and a q parameter of 5 is deduced. In the work of Jenniskens and Désert [11] an absorption

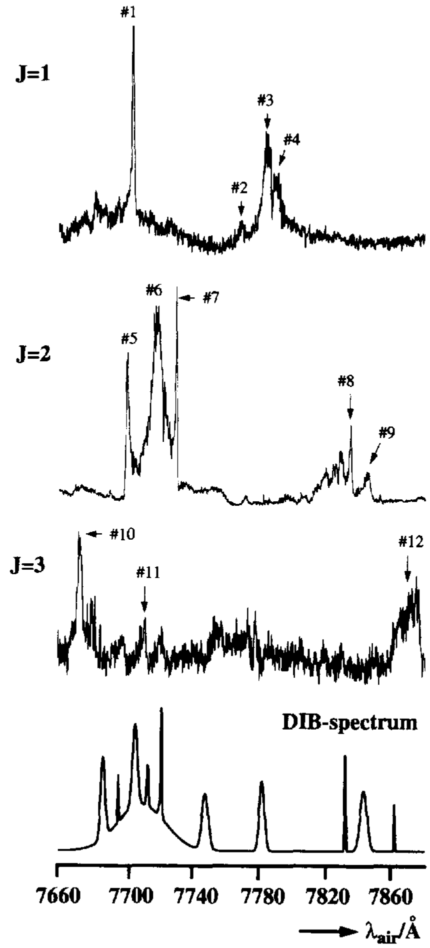

Fig. 2. Spectra from XUV/near-infrared double resonance excitation with the XUV-laser set to probe each of the $C \, ^1\Pi_u^+$, $v = 9$, $J = 1$, 2 and 3 states. The resonances are recorded by registration of signal-enhancement of H_2^+ as a function of wavelength of the near-infrared laser. The lower trace shows a reconstructed spectrum of the diffuse interstellar bands, in the same wavelength region, from the data of Jenniskens and Désert [11]. For convenience of comparison with astronomical data the wavelength is given in Å and in air.

Table 1
Observed autoionizing resonances in H_2 in the wavelength range 766–788 nm upon excitation from $C\,^1\Pi_u^+$, $v=9$, $J=1$, 2 or 3 intermediate states. In order to facilitate comparison with astronomical data the wavelengths are given in λ_{air} and in Å. The numbering of the lines refers to the lines in Fig. 2

Line No.	Wavelength (Å)	Width (Å)
1	7701.6 ± 0.5	1.1 ± 0.4
2	7768.4 ± 1.0	13 ± 8
3	7783.7 ± 1.0	10 ± 4
4	7790.4 ± 1.0	3 ± 2
5	7694.7 ± 0.5	1.6 ± 0.2
6	7713.4 ± 0.7	9 ± 1
7	7725.8 ± 0.4	0.7 ± 0.1
8	7834.9 ± 0.4	1.9 ± 0.2
9	7846.3 ± 1.0	4 ± 1
10	7671.6 ± 1.0	3.5 ± 1.0
11	7710.8 ± 1.0	3 ± 1
12	7870.7 ± 1.0	16 ± 3

phenomenon of possible interstellar origin is identified at 7695.92 ± 0.16 Å with a width of 0.67 ± 0.18 Å. Although feature No. 5 is close it does not match the DIB within the error limits.

A second DIB coincidence was found with the XUV laser set at the R(1) line of the $B''\,^1\Sigma_u^+ - X\,^1\Sigma_g^+$ (7, 0) band at 84.6 nm and the near-infrared laser scanned in the range 800–810 nm. A double resonance spectrum is shown in Fig. 3. Two strong resonances appear, one at $\lambda_{air} = 8038.4 \pm 0.5$ Å and another line at $\lambda_{air} = 8075.4 \pm 0.5$ Å. Both lines have a width of 2.0–3.0 Å. In the work of Jenniskens and Désert [11] an asymmetric line was observed that was deconvoluted into two lines. One component was listed at $\lambda = 8038.48 \pm 0.25$ Å with a width of 3.2 Å. So one of the observed features (8038.4 Å) shown in Fig. 3 coincides with a DIB, while the second (8075.4 Å) does not.

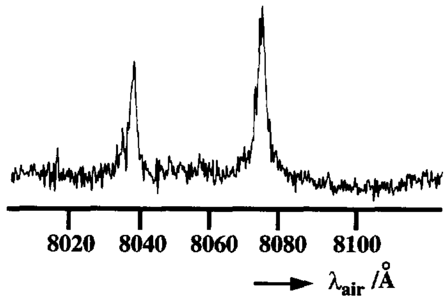

Fig. 3. XUV/near-infrared double resonance excitation with the XUV-laser set to probe the $B'\,^1\Sigma_u^+$, $v=7$, $J=2$ state and the near-infrared laser scanned in the range as indicated.

In the wavelength range 766–788 nm twelve more or less pronounced spectral features are observed.

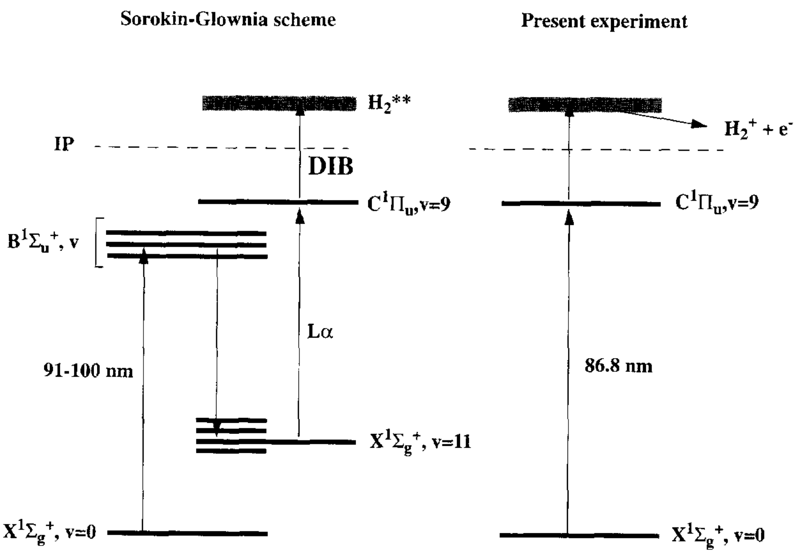

Fig. 4. Energy level scheme pertaining to the Sorokin/Glownia model of ref. [7] (left) and the level scheme pertaining to the present experiment (right).

This corresponds to the number of DIBs in this wavelength range [11].

4. Discussion

In the present experimental study a recent hypothesis concerning the origin of the diffuse interstellar bands is tested. The energy level scheme related to the pumping mechanism proposed by Sorokin and Glownia [7] as well as the excitation scheme used in our laboratory are depicted in Fig. 4. In the interstellar medium (or rather in circumstellar regions) molecular hydrogen would absorb radiation in the range 91–100 nm in excitation to $B\,^1\Sigma_u^+$, v states. Because the potential well of this valence state extends to large internuclear distances highly vibrational states $X\,^1\Sigma_g^+$, v'' will be populated after spontaneous decay. Lyman-α radiation, abundantly present with a large spectral width, is resonant with R(0)–R(3) lines of the $C\,^1\Pi_u$–$X\,^1\Sigma_g^+$ (9, 11) band. In the language of Sorokin and Glownia this Lyman-α resonance gives rise to enhanced two-photon excitation. Alternatively one may view the occurrence of the Lyman-α resonance as a mechanism to populate the $C\,^1\Pi_u^+$, $v=9$, $J=1$–4 states. In the present laboratory investigation analoguous state-selective population of the $C\,^1\Pi_u^+$, $v=9$, $J=1$–3 states is achieved directly by XUV laser excitation. For the Sorokin–Glownia hypothesis to be tested this makes no principal difference.

The observed spectra do not reproduce the DIB spectra although two matching resonances were found. It is not easy to explain why some lines observed in the laboratory would relate to DIBs and at the same time other lines would not. However, it should be noted that in fact doubly excited states of H_2 are excited in our experiment, that either dissociate or autoionize. The detection scheme used only probes the ionization channel and the relative absorption cross sections of the lines might be very different from the presently observed signal intensities.

In the XUV/near-infrared double resonance excitation spectra lines of various widths are observed. In autoionization as well as in predissociation a variety of widths is easily explained by different coupling strengths to continuum states. Previously the diffuseness of the DIBs was ascribed to underlying rotational band envelopes in molecular transitions. Also Fano-type profiles are observed. Such asymmetric phenomena in DIB features with a slight emission wing attached to an absorption line were in the past ascribed to absorptions by color centers in crystal grains of specific sizes [12]. All phenomena of varying widths and lineshapes are well known in autoionization of two-electron systems.

Most DIB phenomena listed in the recent overview [11] were not reproduced in excitation from the $C\,^1\Pi_u^+$, $v=9$, $J=1$–3 states. Investigation of the level structure of H_2 revealed that also the $B''\,^1\Sigma_u^+$–$X\,^1\Sigma_g^+$ (7, 14) band is resonant with Lyman-α radiation, and its Franck–Condon factor is even higher [13]. A DIB feature at 8035.5 Å is reproduced in excitation from the $B''\,^1\Sigma_u^+$, $v=7$, $J=1$ state. But then again a feature observed at higher intensity in the same excitation scheme does not relate to a DIB. If the Sorokin–Glownia model is correct then also the Lyman-α resonances with other transitions would have to be included. Apart from the B″–X (7, 14) band the D–X (1, 11) band is also resonant, but here the Franck–Condon factor is much smaller [13].

5. Conclusion

The recent hypothesis on the carrier of the diffuse interstellar bands, involving Lyman-α pumping of H_2 to $C\,^1\Pi_u^+$, $v=9$, $J=1$–4 states could not be unequivocally confirmed in a laboratory investigation. Since the detection technique presently used is biased in favor of monitoring autoionizing resonances, while it is not sensitive to dissociative resonances, the present result is not conclusive in rejecting the hypothesis. Although line positions are not reproduced the general pattern of the observed features resembles the DIB phenomena: lines of widths varying between 1 and 20 Å, some of them asymmetric, while the number of lines observed is nearly equal to the number of DIBs. The resemblance of the pattern as such supports the hypothesis of H_2 as a carrier for the diffuse interstellar bands.

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