



In situ measurement and modeling of hydrogen recycling and transport processes – the role of molecules

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Abstract

In TEXTOR-94 experiments have been performed which try to elucidate the nature and origin of the hydrogen released from plasma facing graphite surfaces. Density profiles and velocity distributions have been measured by means of passive and active spectroscopy on atomic and molecular line emissions for different surface temperatures and plasma conditions. It was found that until 1100 K hydrogen is predominantly released as H₂. Above 1100 K a reduction of the H₂-flux (50% at 1370 K) was determined. No flux density dependence in comparison to ion beam experiments were found. The photon rates for hydrogen atoms are larger than for the two atoms in the molecules. In the presence of a large fraction of molecules this may lead to an underestimation of the total hydrogen influx from Balmer line measurements. Comparisons with EIRENE-code calculations and molecular spectroscopic data show that the molecules are highly vibrationally excited, which leads to low energetic atomic dissociation products. The origin of these kind of molecules is probably not in the plasma but already at the graphite surface. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

The density, velocity distribution, and the atomic and molecular fractions of neutral hydrogen being released from plasma facing components in fusion devices determines largely the impact of these neutrals on plasma edge properties. It influences in general energy and particle transport at the edge and, therefore, plasma flow properties, momentum losses, fuelling efficiencies, the radiation characteristics of impurities and pumping efficiencies for helium are affected. The optimal design of the ITER divertor depends thus also on a detailed knowledge about the mechanisms of hydrogen recycling and release.

As a complication hydrogen can be released from the plasma facing components both as an atom and a molecule. In the first case a surface can emit the atoms directly, either by ion-induced desorption or by reflecting ions or neutrals. Except for very low energy impact conditions, where chemical bonding to the surface becomes important, these processes can reasonably be modelled via the TRIM-code [1], and the spectroscopic detection [2] of these atoms is a straight forward one and has already been applied in the modelling of the boundary layer for a long time. When the hydrogen is released in form of molecules, however, the physics becomes more complicated and the break-up atoms from re-emitted molecules are coupled to a variety of processes, which finally lead to a broad spectrum of velocity components. The molecules may leave the surface in vibrationally excited states, which can have a large impact on the dissociation channels and, hence, on

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the Franck–Condon energies. The relevance of these processes in boundary and divertor plasmas – although many of these have already been intensively studied under laboratory conditions [3] has presently experienced a growing interest as it became evident that, under certain plasma conditions, molecules may make up the majority of the released hydrogen. This leads to a strong need for optical spectroscopy of molecular hydrogen and its isotopes and its influence on the emission of atomic lines, which may also result in a revision of the interpretation of the latter in terms of fluxes derived from their intensities. In addition, present models, which describe the distribution of neutral hydrogen in front of wall and limiters (e.g. EIRENE [4]), should be revisited with respect to the molecular processes involved.

As many of these processes are spatially confined to regions of about 1 cm in front of plasma facing surfaces, a sufficient access to these components and the plasma boundary is needed for observation. This is provided in TEXTOR-94 by several limiter lock systems which allow to introduce test limiters (up to 12 cm in diameter) without breaking the vacuum system. The test limiters as well as parts of the wall are diagnosed by high resolution Doppler-spectroscopy, laser-induced fluorescence, atomic and molecular spectroscopy with high resolution in combination with gas-puff systems and 2D-mapping of line intensities. The surface temperature of special limiters can be controlled by external heating up to 1300 K. The interpretation of tokamak measurements are supported by ion beam experiments employing a 1 keV H^+ beam on heatable carbon containing targets.

The relative importance of the processes mentioned above at the plasma boundary of the tokamak TEXTOR-94 is the issue of this paper. This concerns in particular the local hydrogen particle density, its excitation state and velocity distribution. Special emphasis will be given determining the ratio of emitted atoms to molecules for different discharge and surface conditions.

On TEXTOR-94 we have recently performed D_α -line profile measurements in the boundary layer with a high resolution spectrometer [5] for different heating conditions and plasma radii. The surprising result of all these measurements had been the existence of a cold component of atomic hydrogen. Probably the specific observation geometry had been very helpful, by which line emission from inner plasma radii could be avoided. The deconvolution procedure yielded at least two different components with temperatures of 0.35 and 15 eV. Neither variations in the plasma radius nor heating conditions displayed a remarkable influence on the temperature of the components. Another evidence for the existence of such a low energy component came from laser induced fluorescence (LIF)-measurements on $Ly-\alpha$ [6], which also detected hydrogen atoms in a very shallow plasma region with energies below 1 eV and an insensitivity of the velocity distributions against the

plasma radius within the scrape-off layer plasma. So far measurements have only been done in front of the liner but could recently be extended to regions on front of test limiters [7].

2. Experimental set-up and methods

In order to determine the contribution of molecular hydrogen to the total hydrogen flux, high resolution emission spectroscopy of hydrogen molecules was performed. The emission spectra of a several bands [$3p^3\Pi_u \rightarrow 2s^3\Sigma_g^+$ (Fulcher) and $3d^1\Sigma_g^+ \rightarrow 2p^1\Sigma_u^+$] were measured in the ranges 600–650 nm and 400–440 nm, respectively. This was done both with a spectrometer with a resolution of about 0.2 Å [8] and a spectrometer equipped with an intensified CCD camera as detector (resolution ≈ 1 Å), which could be used for spatially (0.5 mm) and spectroscopically resolved measurements. The latter wavelength region was chosen particularly for extraordinary high limiter temperatures in order to minimize the influence of the continuum radiation in the radially resolving spectrometer. The position of the observation volumes was set at a fixed toroidal position conventionally chosen near the location of largest power loading or a hole for gas blow experiments 15 mm from the center. More experimental details can be found in Refs. [9,10].

The graphite limiters (10 cm long, 6 cm wide and 5 cm high) with a toroidal and poloidal curvature of 8.5 and 6 cm, respectively, were introduced into TEXTOR-94 through a limiter lock from the bottom of the torus and had been positioned at the same plasma radius as the toroidal belt limiter ALT-II ($r_{Lim} = 46$ cm). A special limiter (surface 100 cm²) was resistively heated up to a temperature of 1300 K in order to become independent of the heating by the plasma with varying loads.

TEXTOR was normally operated under the following discharge conditions: $I_p = 350$ kA, $B_t = 1.75$ – 2.25 T and NBI-heating of 1.3 MW. Mostly hydrogen has been used as working gas because the properties of its molecular emission spectrum are much better documented than for its isotopes. The TEXTOR vessel itself was at a temperature of 620 K and routinely boronized.

3. Results

3.1. Atom/molecule ratio

It has been speculated whether hydrogen molecules and their break-up into atoms may cause these extraordinary low atom energies. For this purpose the relative amount of released molecules and their contribution to the emission of the atomic Balmer lines has to be determined. During the past decade several attempts

have been made to resolve the individual contributions to the overall hydrogen energy distribution using Doppler spectroscopy in front of limiters [11,12] and walls [13,14]. Many of the authors noticed a growing velocity of the atoms in plasmas near the density limit, but could not give a satisfactory explanation. However, it has been speculated that a growing number of hydrogen molecules released from the walls could produce this effect.

On TEXTOR-94 these measurements have been accomplished by spectroscopy of hydrogen molecules, which have been either blown through a limiter into the plasma or by influencing the atom/molecule ratio via limiter heating or change in the plasma density. The first of these possibilities has been demonstrated by Frazen and Vietzke [15] in ion beam measurements, where the amount of re-emitted molecules could be brought down to zero for carbon target temperatures above 1800 K. However, in these experiments the impinging hydrogen fluxes was about three orders of magnitude smaller than in the plasma boundary of a tokamak. It has been speculated that a possible flux dependence of this effect as predicted in a simple model would shift the transition temperature to values well above 2000 K. It was also in the scope of this work to test the validity of this dependence.

Fig. 1 displays examples of the measured molecular hydrogen spectra for limiter temperatures of about 600 K. One can notice that a resolution $\lambda/\delta\lambda$ of > 15000 is necessary (Spek_H) to detect the majority of the molecular lines individually. However, for special purposes a resolution of $\lambda/\delta\lambda \approx 3 \times 10^4$ (Spek_L) may be sufficient. In the case of Fig. 1(b) (which should be preferred for measurements when the limiter becomes hot) the spectrum also contains – besides the molecular bands of H_2 and CH – the atomic Balmer γ - and δ -lines. They were used to measure the atomic radiation – originating both from atoms by direct and molecules by dissociative excitation as they fit into the same dynamic range as the molecular lines. The population of the excited levels may be influenced by the plasma electrons [16]; however, as the plasma conditions were not changed during the heating of the limiter the influence of the electrons remains constant and can, therefore, be neglected.

Fig. 2 shows the time traces of CH, H_2 (Σ Q1–3), and H_δ from the spectral lines indicated in Fig. 1(b). Also plotted is the limiter temperature which has been derived from the continuum radiation appearing in the spectral range shown in Fig. 1(a). Before the discharge the limiter is uniformly heated to 670 K. During the ohmic part of the discharge, the surface temperature increases only slightly. After starting the NBI heating the temperature increases to 2000 K within 1 s. In this phase one can already notice a drop in the H_2 -, an even steeper one in the CH-, and an increase (!) in the H_δ -emission. After the NBI-heating the signals recover nearly to the same

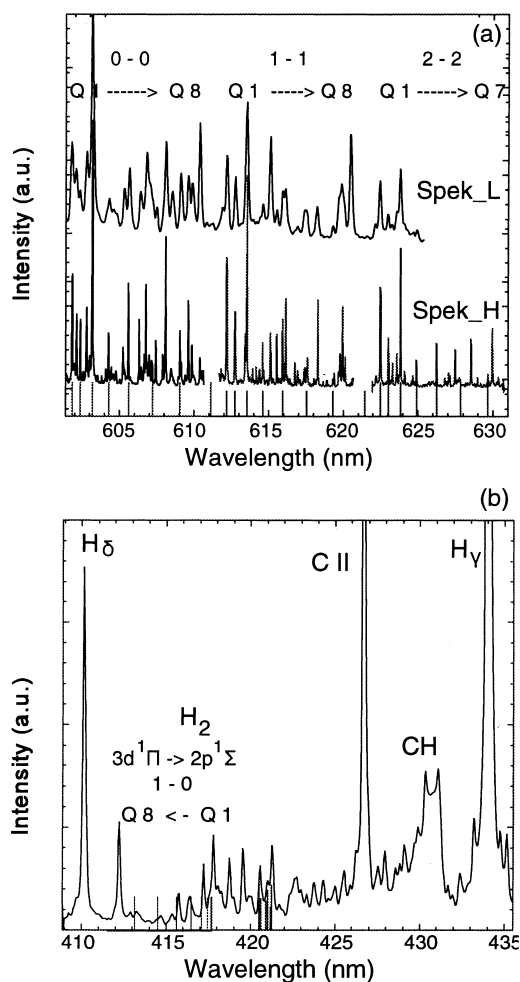


Fig. 1. Spectra obtained in front of a graphite test limiter with two spectrometers of different resolution. (Spek_H): 0.19 Å/pixel; (Spek_L): ≈ 1 Å/pixel showing (a) for H_2 : Q-branches of the (0-0), (1-1), (2-2), and (3-3)-bands of the transition $3p^3\Sigma_u \rightarrow 2s^2\Sigma_g$ (Fulcher band) and (b) H_2 : $3d^1\Sigma_u^+ \rightarrow 2p^1\Sigma_u^+$.

amount – except for the CH-band, because the surface temperature is still too high for its maximum formation rate. The latter behaviour has, under stationary heating conditions, already been published in Ref. [10]. However, the temperature rise during NBI heating is restricted to the highly loaded limiter parts and a quantitative interpretation has to be done with caution since the light emission is line integrated.

In Fig. 3 the changes of the H_2 -, CH-, and $H_{\delta,\beta,\alpha}$ -emission as a function of the limiter temperature in the Ohmic part of the discharge is shown. Whereas the drop for CH occurs again at about 950 K, the intensity of H_2 starts to decrease at about 1100 K. Simultaneously the atomic line emissions from atomic Balmer lines increase. For a better understanding the expected emission be-

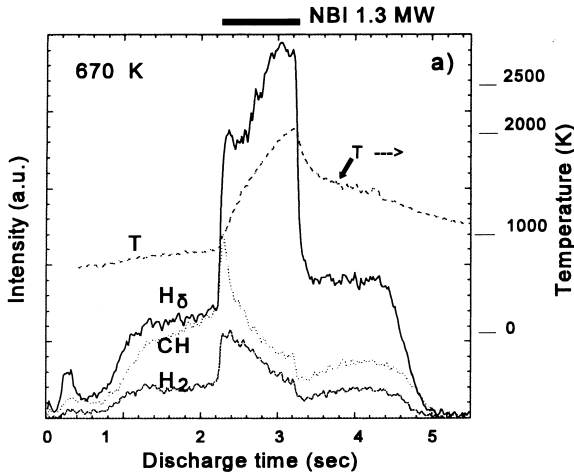


Fig. 2. Time traces of CH, H₂ (ΣQ_{1-3}) (Fig. 1(b)), and H _{δ} for the different limiter temperatures.

behaviours are extrapolated until the complete disappearance of the hydrogen molecules. One should note that the summation of only a smaller number of individual Q_i -components (x) would lead to an earlier drop of about 200 K; however, the general behaviour remains very similar.

3.2. Rotational and vibrational populations

The different temperature of the surface is also reflected in the population of the rotational H₂-bands, which can in our case be represented by the line ratio

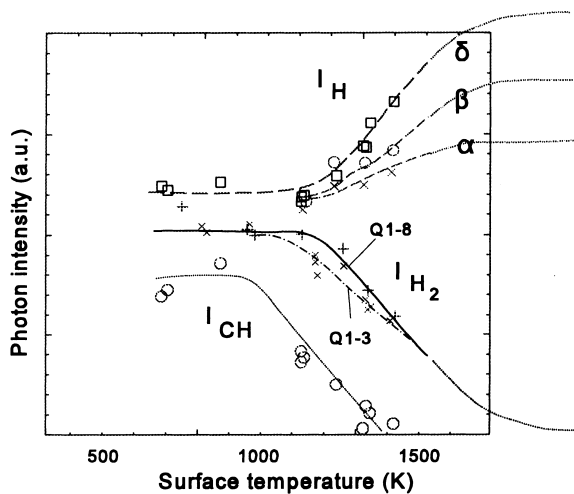


Fig. 3. Variations of the H₂-, CH-, and H _{δ,β,α} -emission (\square, \circ, \times resp.) as a function of the limiter temperature in the Ohmic part of the discharge (uniform heating). A smaller sum of individual Q_i -components is represented by (x).

Q_5/Q_1 of the (0–0) band. Fig. 4(a) shows this behaviour for the ohmic phase of the plasma between 1.4 and 1.9 s as a function of the limiter temperature. The change of the line intensity ratio displays also the problems in the

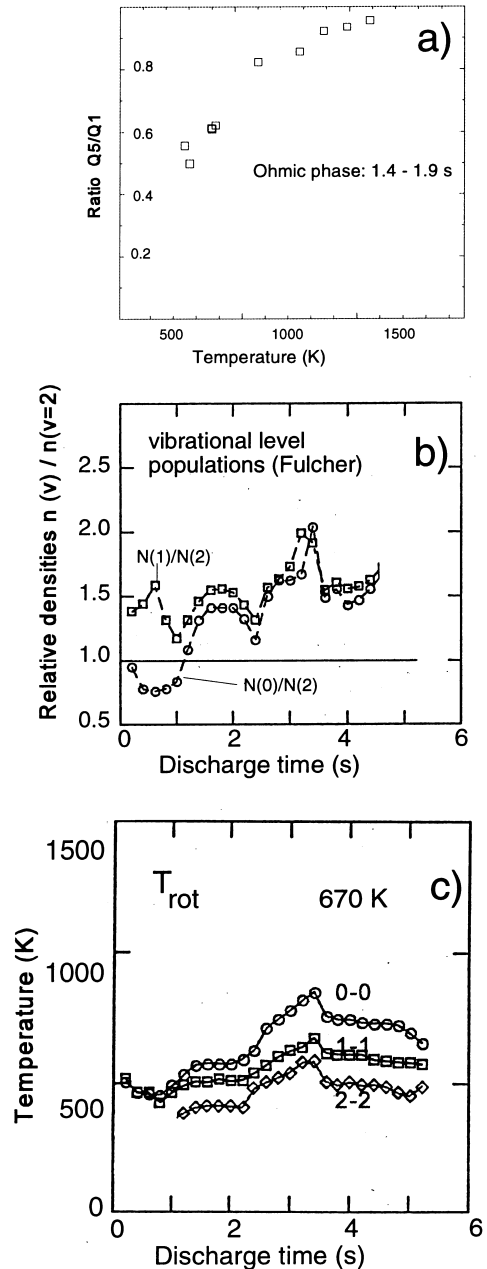


Fig. 4. (a) Change in the rotational temperature of the (0–0) transition in the Fulcher band represented by the line ratio Q_5/Q_1 during the ohmic phase for the experiments shown in Fig. 1. (b) Vibrational level population of the (0–0), (1–1), and (2–2) transition and (c) rotational temperatures during the discharge shown in Fig. 1.

molecular spectroscopy of H_2 . In order to avoid misinterpretations due to changing rotational temperatures it is often necessary to integrate a large number of lines within one band, which – in the case of H_2 – can be rather tedious. From a more refined analysis, which takes a number of rotational lines (also from different vibrational bands) into account, the temporal behaviour of the rotational temperature for the discharge in Fig. 2 is derived (Fig. 4(c)), which also reflects the behaviour during NBI. However, T_{rot} decreases with growing vibrational numbers. The corresponding variation of the vibrational populations is displayed in Fig. 4(b).

3.3. Comparison of radial profiles for $H_{\alpha,\delta}$ with EIRENE

In order to elucidate the nature of the hydrogen release, experimental 2d-recordings of the atomic and molecular light emissions have been performed for the type of discharges shown in Figs. 2 and 3 (Fig. 5). A 1D-scan of the measurements, which shows the radial line intensity distribution of H_2 and H_δ for different surface temperatures is displayed in the bottom graph. For comparison in the right part of the upper picture results of the H_α -line emission from EIRENE-code calculations for this limiter geometry and plasma conditions are added. The processes, which have been taken into account are outlined in Ref. [17] for a limiter and in Ref. [18] for a cold gas divertor surrounding.

4. Discussion

4.1. Atom/molecule ratio

By comparing the results of Fig. 3 with the ion beam measurements of [15] (Fig. 6), one can notice that the drop in the H_2 -production occurs at nearly the same surface temperatures and a flux density dependence is only marginally present. However, the thermal origin of the majority of the released hydrogen is still valid and also proven by the penetration depths of H_δ for the two different temperatures of 670 and 1370 K, which is very equal (≈ 1.5 cm) and not very much different from the molecular one (Fig. 5 bottom).

From the intensity behaviour of the hydrogen lines in Fig. 3, it is possible to derive the atom/molecule ratio for this plasma condition. For example the atomic H_δ -intensity increases by 50% when the molecular H_2 -intensity drops by 50% from 1100 to 1370 K, although the plasma parameters are unchanged and the total hydrogen flux (protons from molecules and atoms) from the limiter is constant for all limiter temperatures. Fig. 7 displays the replot of $I_{H_\delta}^0$ vs. I_{H_2} from Fig. 3 and the dependencies of their intensities as a function of hydrogen atoms and molecules. From these one can after

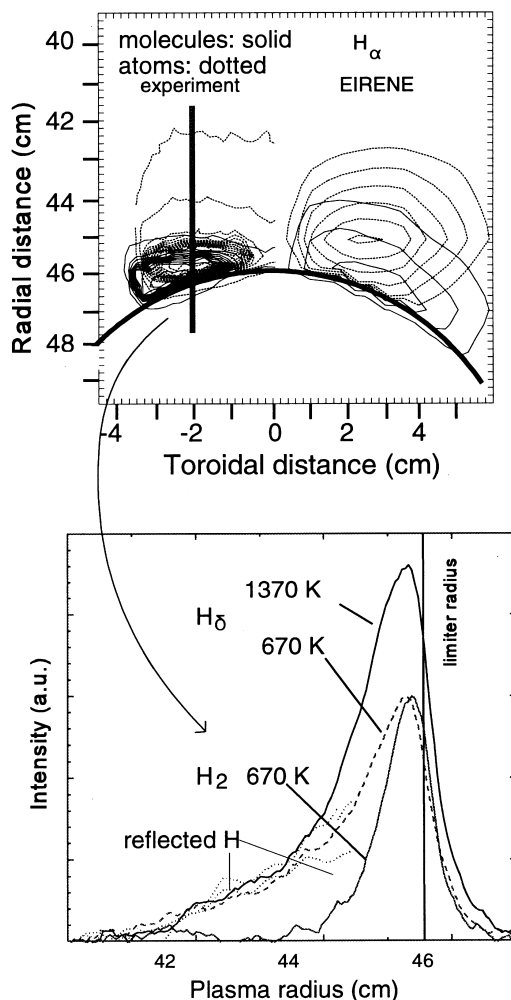


Fig. 5. (Upper left): 2D-pictures of the atomic (H_α) (dotted) and molecular light [$Q1-3$ from (0-0)] distribution on front of a graphite test limiter (solid) for the conditions of Fig. 2. (Upper right): Comparison with EIRENE-code calculations: H_2 from atoms (dotted), from electronically excited H_2 (solid) lower: penetration depths of H_δ for the two different limiter temperatures (within the bar indicated). The light intensity from the higher energetic (probably reflected) atoms is in both cases equal.

some algebra derive an equation for the ratio of atomic to total hydrogen flux [19]

$$\Gamma_H/\Gamma = [2(k_1/k_2)(I_{H_\delta}^0/I_H^0 - 1)]/[2(k_1/k_2) - 1], \quad (1)$$

where k_1 and k_2 are the conversion factors of atomic and molecular fluxes into Balmer line intensity I_H . The quantity I_H^0 is the value for high limiter temperatures, when the hydrogen flux contains only atoms and can be derived from $I_H = f(I_{H_2})$ as shown in Fig. 7 (≈ 0.5 for this discharge condition). If $k_1/k_2 = 1$ (smaller values are not possible!) the release of hydrogen from the limiter sur-

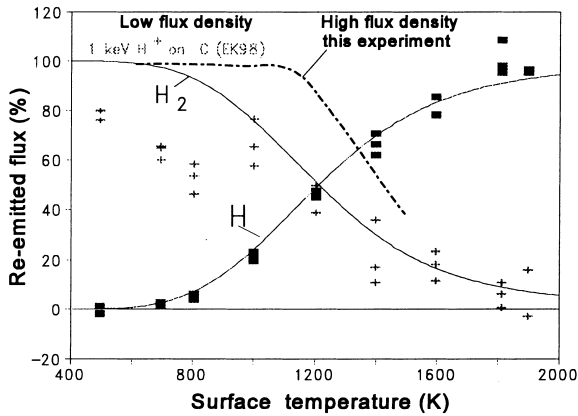


Fig. 6. Comparison of the H₂-decrease with ion beam measurements of Ref. [15].

face below 1100 K would practically be *only* by molecules. However, for $k_1/k_2 \approx 2$, which may be a reasonable value [11], the molecular fraction is still 67%, but never less than 50% for all possible values of k_1/k_2 . The remaining fraction of atomic flux can be represented by the dotted parts in Fig. 5 (bottom graph), which shows the difference between intensity from the thermal and the total H_δ-intensity. Since these are mainly directly reflected hydrogen atoms, their penetration depth does not change for the two different limiter temperatures. When the fraction of molecules is reasonably large, one has, however, to be cautious not underestimate the hydrogen flux as it might happen by the use of the model in Ref.

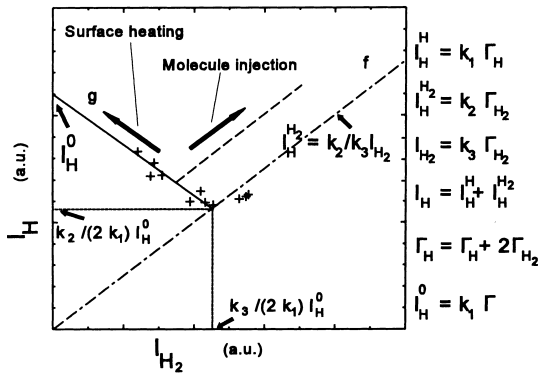


Fig. 7. Derivation of the ratio of the atomic to total hydrogen fluxes and photon efficiencies for the atomic line (H_δ) from atoms and molecules from limiter heating experiments ($\Gamma_{\text{total}} = \text{const!}$) and gas blow ($\Gamma_{\text{H}} = \text{const!}$) experiments. The proportional constants k_i connect the measured intensities with the respective fluxes Γ_i . For limiter heating experiments ($\Gamma = \text{const}$) one moves along the line g and for H₂-gas blow experiments ($\Gamma_{\text{H}} = \text{const}$) along the line f. The experimental conditions for the experimental points (x) were: $n_e (r_{\text{Lim}} = 45.5 \text{ cm}) = 6.5 \times 10^{13} \text{ cm}^{-3}$, $T_e (r_{\text{Lim}} = 45 \text{ cm}) = 45 \text{ eV}$.

[2] applying the numbers for the conversion of H_{α,β,γ,δ}-photons (see Fig. 3). For our plasma conditions the use of H_δ would have led to an underestimation factor of about 2. It is less for H_α as can be seen from Fig. 3 and also theoretically explained in Ref. [16]; for larger fractions of molecules as in our case the error in the hydrogen flux can be considerable even by the use of H_α (up to a factor 2).

4.2. EIRENE-code calculations

From Fig. 5 (top) one can notice immediately that the measured H_α-line emission profile cannot solely be explained by a pure atomic hydrogen source under these plasma conditions. However, the intensity profile, which stems from electronically excited H₂ (the one which fitted the measurements best) represents well the measured H₂-light intensity pattern. This had been in principle expected. But surprisingly the H_α-emission from vibrationally excited molecules came out practically negligible from the code calculations, where a “collisional radiative” model for hydrogen molecules had been added to EIRENE, which also included vibrationally excited states [20]. However, the calculations may look different when the molecules start already vibrationally excited from a surface (Section 4.3) and are followed individually in the code. Such calculations are presently underway.

4.3. Excited hydrogen molecules

Since the hydrogen molecules play a significant role in the overall hydrogen flux from surfaces, we may look for mechanisms, how dissociative excitation can produce both low and high energetic atoms. From Fig. 7(b) there is a possibility to derive the vibrational population of the molecule in the ground state ($X^1\Sigma$) yielding T_{vib} from the measured Fulcher-band intensities by assuming that this population is transferred by electron collisions into the excited states (e.g., $d^3\Pi$). This has been done in Ref. [21] and from the respective figure there one can derive a $T_{\text{vib}} > 5000 \text{ K}$ demonstrating that a remarkable population of levels up to $v = 10$ has to be considered. Recently a “collisional radiative” model has been developed, which predicts these populations as a function of T_e and is in good agreements with the measurements on TEXTOR-94 (Fig. 4(b)) and ASDEX-Upgrade [21].

These highly populated levels may now lead to important consequences: Both via the repulsive state $b^3\Sigma$ of H₂ and the states $X^2\Sigma$ and $2p\sigma$ of H₂⁺ it is possible to obtain (excited) atoms with low ($\approx 0.5 \text{ eV}$) and higher ($\approx 10 \text{ eV}$) energies as indicated by the arrows in the potential curve diagram (Fig. 8) both for low energy divertor ($< 5 \text{ eV}$) and high energy ($> 18 \text{ eV}$) boundary plasmas. The simple reason for this is that the proba-

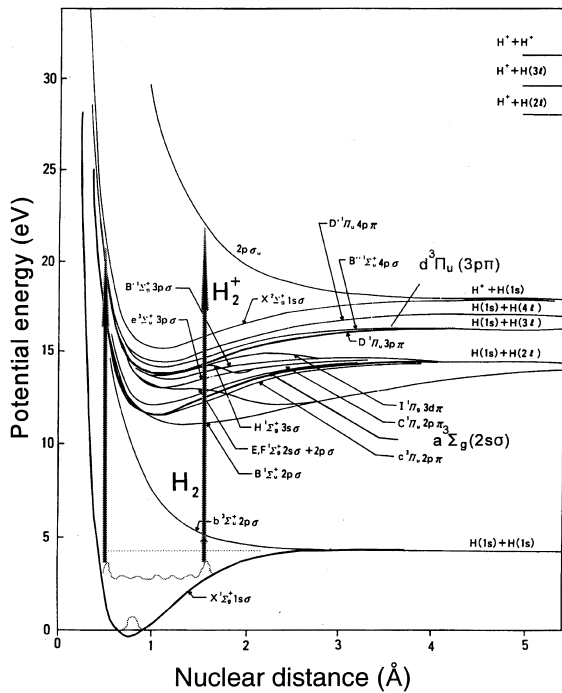


Fig. 8. Some potential curves of the H₂-molecules. Some excitations from excited vibrational levels of the ground state, which may lead to low energetic atoms are indicated by arrows.

bility for excitation is the highest within the turning points. Dissociation from $v = 0$, however, can only lead to energies of about 2.2 eV. This may now also explain why at the density limit, where the number of molecules is the largest – the dependence of the atom/molecule fraction as a function of density is indicated in Fig. 9 –

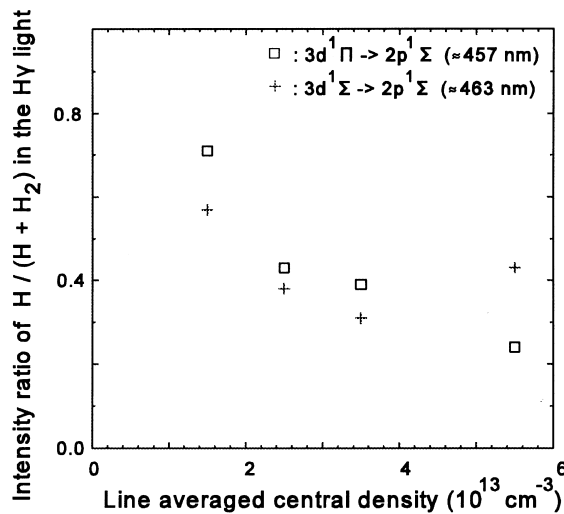


Fig. 9. Variation of the atomic part of H_γ to the total light as a function of n_c derived from gas blow experiments.

the energy of the dissociation component increases (see above).

Finally we have to understand how the molecules can achieve these high vibrational populations. The “collisional radiative” model mentioned above may be an explanation but need not be the only mechanism. Since the relative population is independent from the surrounding plasma conditions [5], it might also be that the molecules already leave the surfaces vibrationally excited, for which well known reaction channels exist: Eley–Rideal, Langmuir–Hinshelwood reactions etc. A strong argument that these processes may have some importance is the fact that obviously a conservation of energy takes place when it is shared between vibrational and rotational energy – i.e. the one grows whilst the other decreases (Fig. 4(c)).

5. Conclusions

For the hydrogen recycling on plasma facing materials in fusion plasmas the following conclusions can be drawn:

- until 1100 K hydrogen is predominantly released as H₂;
- above 1100 K: reduction of the H₂-flux (50% at 1370 K);
- no flux density dependence in comparison to ion beam experiments (only a slight shift to higher temperatures similar as for CH is found);
- the large fraction of molecular hydrogen may lead to different energy exhaust mechanisms in the boundary or divertor of a fusion plasma;
- photon rates for hydrogen atoms are larger than for the two atoms in the molecules; in the past this has led to an underestimation of the total hydrogen influx from Balmer line measurements;
- LiF on H₂ is necessary for a more exact identification of the different processes and their dependencies e.g. as a function of surface material.

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