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Impact of vibrationally and electronically excited H$_2$ on the molecular assisted recombination rate in detached plasma regimes

R. Chandra $^*$, A. Holm, M. Groth

Aalto University, Espoo, Finland

**Abstract**

The vibrational redistribution of H$_2$ molecules via the $B'$ $\Sigma^+_u$ and $C'$ $\Pi_u$ states are included in the determination of molecular assisted recombination (MAR) rates and are found to have negligible impact in detached plasma regimes. In high recycling and detached divertor plasma conditions, MAR occurs through the higher levels ($\nu \geq 4$) of vibrationally excited H$_2$ which are primarily populated through direct electron impact excitation or the electronic excitation of H$_2$ molecules and the subsequent radiative decay to another vibrational state. We use the collisional-radiative model CRUMPET to evaluate MAR rate coefficients with electronic transitions included, which are applied to detached plasma parameters found in the linear plasma device Magnum-PSI.

1. Introduction

Plasma recombination in the divertor region is considered to be an explanation for the decrease of plasma particle flux to the divertor targets in detached plasma regimes [1]. Plasma recombination occurs mainly from radiative and three-body recombination, which are collectively termed electron-ion recombination (EIR) [2]. EIR becomes relevant only at electron temperatures of 1 eV or below. The presence of molecules contributes to the plasma recombination via molecular assisted recombination (MAR) processes, which are relevant at electron temperature higher than EIR ($1 < T_e < 5$ eV) [1,3]. For hydrogen in divertor plasma conditions, MAR occurs through the higher levels ($\nu \geq 4$) of vibrationally excited H$_2$. Hence, the population distribution of the H$_2$ vibrational states influences plasma recombination through MAR. The vibrational states of H$_2$ are primarily populated through electron impact excitation of the vibrational number $\nu$ to a higher number $\nu'$ via two channels [4–6]:

$$\text{H}_2(\nu) + e \rightarrow \text{H}_2(\nu') + e$$

$$\text{H}_2(\nu) + e \rightarrow \text{H}_2(B' \Sigma^+_u, C' \Pi_u) + e \rightarrow \text{H}_2(\nu') + e + h\nu$$

The first reaction is a resonant mechanism involving the temporary formation of H$_2^*$ [6,7]. It is the vibrational excitation process commonly included in vibrationally resolved recombination rate coefficients [8] used in coupled fluid-kinetic edge codes such as SOLPS [9], EDGE2D-EIRENE [10,11], or B2.5-Eunomia [12]. The second reaction excites the vibrational levels indirectly through electronic excitation of the molecule followed by radiative decay [5,6]. To the authors knowledge, the second reaction was not commonly included in the derivation of effective recombination rate coefficients (e.g. AMJUEL [8]) used in fluid-kinetic edge codes.

This paper evaluates the effect of (2) to the vibrational population distribution of H$_2$ and consequently to the MAR rate for electron densities and temperatures expected in the detached plasma regime of a tokamak. The collisional-radiative model CRUMPET [3] is used to solve the population distribution of the H$_2$ vibrational states for certain plasma parameters and is explained in Section 2. Using the H$_2$ vibrational state distribution obtained from CRUMPET, the effective recombination rate coefficients of MAR processes are re-evaluated with (2) included and the results are presented in Section 3. The effect of electronic transitions of H$_2$ to the particle balance in detached plasma regime is evaluated using data from fluid-kinetic code simulations of plasmas in the linear plasma device Magnum-PSI and is shown in Section 4. We summarize our conclusions in Section 5.

2. CR model species and reactions

The population distribution of H$_2(\nu)$ (vibrationally excited H$_2$ in the electronic ground state) is calculated using the collisional-radiative model CRUMPET [3]. CRUMPET allows flexibility in $N$ number of species considered in the equation [13]:

$$\dot{n} = M \dot{n} + \Gamma$$

where $\Gamma$ is the source term, $n$ is the density of species considered, and the $N \times N$ rate matrix $M$. For this paper, the species considered are:

* Corresponding author.

E-mail address: ray.chandra@aalto.fi (R. Chandra).
1. 15 vibrational levels of the electronic ground state of H₂, H₂(2Σ\(^+\)\(^1\)\(g\), \(v\) = 0 – 14
2. 37 vibrational levels of the first electronically excited singlet state of H₂, H₂(2Σ\(^+\)\(^3\)\(g\), \(v\) = 0 – 36
3. 14 vibrational levels of the second electronically excited singlet state of H₂, H₂(2Π\(^+\)\(^1\)\(g\), \(v\) = 0 – 13
4. the molecular ion H₂⁺
5. the negative ion H⁻
6. the electronic ground state of H

The applicability of CRUMPET is only limited by the availability of reaction rate coefficients necessary to construct M [3]. The collisional processes included in M and the available cross-section or rate coefficient data are summarized in Table 1. Direct electron impact vibrational excitation (1) is treated as ladder-like, where only single step transitions are allowed. For electronic excitation of H₂ (2 and 3 in Table 1), all possible transitions between vibrational levels are included. The same applies for optical transitions (4 and 5 in Table 1). The H₂(v) population can be depleted through dissociation, ion conversion (IC), and the dissociative attachment (DA) process (6,7 and 8 in Table 1). The reaction rates are calculated assuming Tₑ = T. The direct ionization of H₂ is not included in CRUMPET for the purpose of comparison with simulation data [14] later in Section 4, which also does not include this process. It is advised to include direct ionization in the future, especially when the electron temperature can exceed 4 eV, where it starts to compete with ion conversion and dissociation.

Two different CRUMPET inputs are created to demonstrate the effect of electronic transitions to the population distribution of H₂(v). The first input considers only the electronic ground state of H₂, H₂(2Σ\(^+\)\(^1\)\(g\), \(v\) = 0, \(v\) = 0 and 1, respectively. The first input will be referred to as no electronic transitions, both through the IC and the DA channel. The effective rate matrix R. Chandra et al. effect of H₂ transitions is not included, as shown by the population distributions of H₂(v) for electron temperatures Tₑ = 1, 3, and 5 eV in Fig. 1. Indeed, the reaction rates for the electronic transition to the singlet B and C states, as described in reaction number 2 and 3 in Table 1, are expected to be dominant for higher electron impact energies [4].

The shift towards higher vibrational population of H₂ starts to improve the MAR rates for H₂ when the electronic transitions of H₂ is included in the model. This trend is reversed when electronic transitions are not included, as shown by the population distributions of H₂(v) for electron temperatures Tₑ = 1, 3, and 5 eV in Fig. 1. Indeed, the reaction rates for the electronic transition to the singlet B and C states, as described in reaction number 2 and 3 in Table 1, are expected to be dominant for higher electron impact energies [4].

The shift towards higher vibrational population can potentially increase the effective recombination rate through the MAR process, both through the IC and the DA channel. The effective rate coefficient for these two channels can be calculated as follows:

\[ \langle \sigma v \rangle_{eff} = \sum_{i=0}^{14} (\sigma v)_i P_{H_2(v)} \] (4)

Table 1

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction formula</th>
<th>Data ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>H₂(2Σ(^+)(^1)(g), (v) = 0) + e → H₂(2Σ(^+)(^1)(g), (v) = 1) + e</td>
<td>H2VIBR 2.l0/1+1 [15–17]</td>
</tr>
<tr>
<td>2</td>
<td>H₂(2Σ(^+)(^1)(g), (v) = 0) + e → H₂(2Σ(^+)(^1)(g), (v) = 2) + e</td>
<td>MCC 18</td>
</tr>
<tr>
<td>3</td>
<td>H₂(2Σ(^+)(^1)(g), (v) = 0) + e → H₂(2Σ(^+)(^1)(g), (v) = 3) + e</td>
<td>MCC 18</td>
</tr>
<tr>
<td>4</td>
<td>H₂(2Σ(^+)(^1)(g), (v) = 0) + e → H₂(2Σ(^+)(^1)(g), (v) = 4) + e</td>
<td>[19]</td>
</tr>
<tr>
<td>5</td>
<td>H₂(2Π(^+)(^1)(g), (v) = 0) + e → H₂(2Π(^+)(^1)(g), (v) = 1) + e</td>
<td>[19]</td>
</tr>
<tr>
<td>6</td>
<td>H₂(2Σ(^+)(^1)(g), (v) = 1) + e → H₂(2Σ(^+)(^1)(g), (v) = 2) + e</td>
<td>H2VIBR 2.l2 [15,17]</td>
</tr>
<tr>
<td>7</td>
<td>H₂(2Σ(^+)(^1)(g), (v) = 2) + e → H₂(2Σ(^+)(^1)(g), (v) = 3) + e</td>
<td>H2VIBR 2.l2 [15,17,20]</td>
</tr>
<tr>
<td>8</td>
<td>H₂(2Σ(^+)(^1)(g), (v) = 3) + e → H₂(2Σ(^+)(^1)(g), (v) = 4) + e</td>
<td>H2VIBR 2.l3 [7,15,17]</td>
</tr>
</tbody>
</table>

Table 2

<table>
<thead>
<tr>
<th>Nr.</th>
<th>Reaction formula</th>
<th>Name</th>
<th>Data ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>H₂⁺ + e → 2e + H + H²⁺</td>
<td>MAI</td>
<td>AMUJEL 2.2.11 [6]</td>
</tr>
<tr>
<td>2</td>
<td>e + H + H²⁺</td>
<td>MAD-IC</td>
<td>AMUJEL 2.2.12 [8]</td>
</tr>
<tr>
<td>3</td>
<td>H + H</td>
<td>MAR-IC</td>
<td>AMUJEL 2.2.14 [8]</td>
</tr>
<tr>
<td>4</td>
<td>H²⁺ + e → H + H + e</td>
<td>MAD-DA</td>
<td>AMUJEL 7.2.3b [8]</td>
</tr>
<tr>
<td>5</td>
<td>H + H</td>
<td>MAD-DA</td>
<td>AMUJEL 7.2.3a [8]</td>
</tr>
</tbody>
</table>

where \( P_{H_2(v)} = H_2(v)/H_2(v = 0) \) is the relative population fraction of \( H_2(v) \). The IC and DA processes do not necessarily result in plasma recombination and depend on the subsequent reaction of the ionic products, H₂⁺ for IC or H²⁺ for DA. These subsequent reactions are summarized in Table 2. Taking into account these follow-up reactions, a factor can be added to Eq. (4) such that an effective MAR rate can be defined as:

\[ \langle \sigma v \rangle_{eff} = \left( \sum_{i=0}^{14} (\sigma v)_i P_{H_2(v)} \right) \left( \frac{\langle \sigma v \rangle_{eff}}{\langle \sigma v \rangle_{ref}} \right) \] (5)

where \( \langle \sigma v \rangle_{eff} \) is the ratio between rate coefficients of reactions resulting in recombination and the total rate coefficients of all possible reaction depleting the ionic products H₂⁺ or H⁻. The effective MAR rate coefficients through the IC and DA channel are evaluated for 1000 Tₑ values evenly spaced in the logarithmic scale between 0.1–1000 eV (Fig. 2). By including the electronic transitions of H₂, the change in the population of the higher vibrational states of H₂(v) alters the effective recombination rate coefficients starting from Tₑ = 3 eV. For the ion conversion channel (MAR-IC), the rate coefficient is enhanced by a factor of 3 at around Tₑ = 5 eV, and by more than an order of magnitude at around Tₑ = 15 eV. The impact of including the electronic transitions of H₂ is even more profound for effective recombination via the dissociative attachment channel (MAR-DA). This pathway becomes a relevant contributor of plasma recombination for Tₑ ≥ 2 eV, contrary to when no electronic transitions of H₂ is considered. Furthermore, the rate coefficient is larger by 2 orders of magnitude at around Tₑ = 10 eV. The electronic transitions of H₂ starts to improve the MAR rates for Tₑ above 2–3 eV. However, in this temperature range, direct dissociation and ionization deplete the electronic ground state of H₂ as
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Fig. 2. Effective MAR rate coefficients via ion conversion (MAR-IC) (a) and via dissociative attachment (MAR-DA) (b) pathways calculated with CRUMPET. The rate coefficients are evaluated when the electronic transitions of H$_2$ are included (red solid line) and neglected (red dotted line). As a comparison, the effective MAR rates from AMJUEL, where electronic transitions are neglected, is shown in black. The relative difference between the CRUMPET rate coefficient and the no el. rate coefficients of AMJUEL (black) and CRUMPET (red) are shown for MAR-IC (c) and MAR-DA (d). The rate coefficients are calculated for $n_e = 10^{20}$ m$^{-3}$. The AMJUEL rate coefficient collapse faster than CRUMPET at very low temperatures which resulted in the very high ratio numbers shown in (c) and (d). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 3. The rate coefficients of processes that deplete the electronic ground state of H$_2$. The MAR precursors (IC and DA) compete with direct dissociation (black circle) and ionization (blue square) for $T_e \geq 2$ eV even when the electronic transitions of H$_2$ are included. The rate coefficients are calculated for $n_e = 10^{20}$ m$^{-3}$. There is no ionization rate with electronic transitions as it is not included in CRUMPET.

A MAR precursor [1]. Direct dissociation and ionization of H$_2$ are still dominant for $T_e$ above 2 eV even when electronic transitions can significantly enhance MAR rates as shown in Fig. 3. Hence, evaluating the overall impact of electronic transitions of H$_2$ to the global particle balance requires information on the H$_2$ ground state density which is coupled with the electron density and temperature.

4. Evaluation of MAR rate in detached plasma regimes

SOL plasma codes, such as SOLPS, EDGE2D-EIRENE, and B2.5-Eunomia, solve the Braginskii equations for the plasma species, and the Boltzmann equation for the neutral species using Monte Carlo methods. They provide self-consistent information of the plasma density, temperature, and neutral densities. A detachment study using B2.5-Eunomia [14] was recently conducted in the linear plasma device Magnum-PSI [23]. Plasma detachment is achieved by increasing the H$_2$ gas pressure exclusively in a chamber housing the target of the plasma beam using gas puffing [24]. The plasma was observed to be in a varying state of detachment depending on H$_2$ pressure that is applied within the chamber [24]. We use the plasma density, temperature, and the H$_2$ density in the electronic and vibrational ground state from B2.5-Eunomia simulations of the several detached plasma states for H$_2$ pressures of 0.27–2 Pa. The plasma density, temperature, and the H$_2$ density associated with the different H$_2$ pressure values are illustrated in Fig. 4. These plasma conditions cover the temperature range where the effect of electronic transitions of H$_2$ to the effective recombination rate becomes noticeable ($T_e \geq 2$ eV), based on the analysis on the effective MAR rate coefficients in Section 3.
Fig. 4. \( T_e, n_e \) and \( n_{H_2} \) distributions of the Magnum-PSI plasma beam within the target chamber. The values are obtained from B2.5-Eunomia simulations of varying \( H_2 \) pressures of 0.27, 0.53, 1.0 and 2.0 Pa [14], measured near the pumping surface far from the plasma (\( R = 0.2 \) m). The target plate is located at the east boundary (\( Z = 0.09 \) m).

Fig. 5. Global MAR-IC (a) and MAR-DA (b) rates calculated using data shown in Fig. 4. The rates are shown for each simulated pressure level representing the varying stages of plasma detachment. The rate uses \( \langle \sigma v \rangle_{MAR} \) with electronic transitions of \( H_2 \) included (black) and neglected (red). The impact of electronic transitions of \( H_2 \) to the MAR rate is negligible in the detached regime. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The plasma density, temperature, and the \( H_2 \) density in the electronic and vibrational ground state from B2.5-Eunomia are used to calculate the effective recombination rate \( S_{rec} \) following (5):

\[
S_{rec} = \sum_c \left[ \langle \sigma v \rangle_{MAR} n_e n_{H_2}(v=0) V \right]_c
\]

where \( n_e \) is the electron density, \( n_{H_2}(v=0) \) is the hydrogen molecule density in the electronic and vibrational ground state, \( V \) is the plasma volume, and \( c \) indicates the quantities to be local in a grid cell of the finite-volume B2.5 code. The grid cells are limited to the plasma region inside the target chamber of Magnum-PSI. The global recombination rate from MAR-IC and MAR-DA pathways for each level of pressures are shown in Fig. 5. By including electronic transitions of \( H_2 \), the MAR rates are enhanced marginally in the ‘attached’ plasma regime for which \( T_e \) is around 3–4 eV. The MAR-IC pathway is enhanced by approximately 7% while the MAR-DA pathway shows a larger increase of around 16%. The impact diminishes towards detached plasma regime and becomes negligible at the highest pressure for MAR-IC and MAR-DA, at 0.7% and 2%, respectively.

The same analyses are repeated for the MAD pathways (nr. 2 and 4 in Table 2). While these processes do not produce plasma recombination, they can remove electron energy from the plasma. The electron cooling rate can therefore be higher when electronic transitions are included. Indeed, this increase is readily observed from the increase of global MAD-IC and MAD-DA rates shown in Fig. 6. The MAD-IC and MAD-DA pathways are increased by, respectively, 18% and 59% in the attached plasma regime. Similar with MAR, the impact diminishes towards detached plasma regime to around, respectively, 1% and 3%. However, MAD is expected to be relevant at attached plasma temperatures, and so the electronic transitions should be included in determining MAD rates.

5. Conclusions

The impact of electronic transitions to the singlet B and C states of \( H_2 \) on the MAR rate is evaluated using the CR model CRUMPET and data of electron density, temperature and \( H_2 \) density from B2.5-Eunomia simulations of plasma detachment in the linear plasma device Magnum-PSI. The impact is highest in the attached plasma regime where the electron temperature is around 3–4 eV, as expected from the modified effective MAR rate coefficients shown in Fig. 2. However, the enhancement diminishes for further plasma detachment, and so remains insufficient to change the system significantly. On the other hand, MAD rates is enhanced substantially in the attached plasma regime where it is relevant, and thus can have major impact in the plasma energy balance. The additional electron cooling may affect plasma recombination indirectly by driving the temperature further down. While this paper uses simulation data from a linear plasma device, these effects are expected to occur in tokamak divertors with similar plasma and neutral conditions, as the effect is dependent mainly on the plasma density, temperature and neutral densities. It is important to note that reactor divertors are subject to other physics such as drifts and complex magnetic geometries which are absent in a linear plasma device. The same approach presented in this paper is readily available for tokamak simulations performed using SOLPS, EDGE2D-EIRENE or other codes, which included the aforementioned physics. These studies should provide more insight in the effect of electronic transitions of \( H_2 \) to plasma recombination, and consequently, their effects on plasma detachment.
Data availability

Data will be made available on request.

Acknowledgements

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References


