Observation of Molecular and Atomic Ions in Recombination Plasma

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Measurements of the densities of the molecular and atomic ions \((n_\text{H}^+, n_{\text{H}_2^+}, n_{\text{H}_3^+})\) were carried out in hydrogen recombination plasma in the linear divertor plasma simulator, TPD-SheetIV. The molecular and atomic ion currents were detected using an omegatron mass analyzer. The ground-state vibrational temperature of hydrogen molecules \(T_{\text{vib}}\) is obtained by measurement with VUV emission spectroscopy. Taking into account of \(T_{\text{vib}}\), a zero-dimensional model using the relevant rate balance equations was found to predict the observed dominant ion.

Keywords: atomic and molecular process, recombination, omegatron mass analyzer, vibrational temperature, TPD–SheetIV

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1. Introduction

The production mechanism of atomic and molecular processes in hydrogen plasma has become increasingly important for plasma processing, space plasma, and fusion reactors. In a recombination plasma at low temperature, vibrationally excited hydrogen molecules \(\text{H}_2(\nu)\) persist in dissociation and ionization processes of the plasma volume. Thus, the plasma volume recombination associated with \(\text{H}_2(\nu)\) molecules in the recombination plasma is effective in enhancing the reduction of ion particle flux [1,2]. The \(\text{H}_2(\nu)\) molecules contribute to plasma volume recombination owing to the following chain reactions: \(\text{H}_2(\nu) + \text{H}^+ \Rightarrow \text{H}_2^+ + \text{H}\) followed by \(\text{H}_2^+ + e \Rightarrow \text{H} + \text{H}\) (dissociative recombination: DR) or \(\text{H}_2^+ + e \Rightarrow \text{H}_3^+ + \text{H}\).

However, the role of molecular ions in the hydrogen plasma is still under discussion [3–7]. It was shown in a previous paper that the role of molecular ions in the hydrogen plasma for lower plasma density has been discussed by E.M. Hollmann et al. [4] On the other hand, it has been pointed out that even the contribution of recombination is not critical for a reduction in ion flux [8]. Recently, it has been reported that in a typical high-density plasma, \(\text{H}_2(\nu)\) molecules assist dissociation (MAD) and ionization (MAI) before they contribute to recombination [9]. For high-density and low-temperature plasma, the MAD always dominates DR. It is thus required that experiments which will aid the understanding of the role of DR are carried out. Molecular processes with \(\text{H}_2(\nu)\) molecules have not been reported clearly for high-density plasma.

In this study, we have carried out the experimental observation and modeling of molecular ions in hydrogen plasma in a linear plasma device, TPD-SheetIV [5,6]. Measurements of the densities of molecular and atomic ions were carried out in hydrogen plasma with a hydrogen gas puff. The molecular and atomic ion currents were detected using an omegatron mass analyzer [7], while the electron density and temperature were measured using a Langmuir probe and a double probe. The ground-state vibrational temperature of hydrogen molecules \(T_{\text{vib}}\) is obtained by measurement with VUV emission spectroscopy. The zero-dimensional plasma model used to predict the measured densities of hydrogen ion species is discussed.

2. Experimental Apparatus and Method

The experiment was performed in the linear plasma device TPD-SheetIV, shown in Fig. 1 [5,6]. Ten rectangular magnetic coils formed a uniform magnetic field of 0.8 kG in the experimental region. The hydrogen plasma was generated at a hydrogen gas flow of 75 sccm, with a discharge current of 30-100 A. The neutral pressure \(P\) in the experimental region was controlled between 0.1 and 20 mTorr with a secondary gas feed. The sheet plasma flowed from the plasma source along the magnetic field to an endplate located about 1.0 m downstream. Electron density and electron temperature were measured using a planar Langmuir probe and double probe, which were located 3 cm in front of the endplate.

An omegatron mass analyzer, situated behind a small hole (\(\Phi\ 0.5\) mm) in the endplate with a differential pumping system, is used for analyzing ion species [4,7]. The omegatron mass analyzer is electrically isolating and is water-cooled to allow the entire housing to be inserted into the plasma column. Inside the diagnostic body of this mass analyzer, electrons are stripped away from the particle beam with a pair of electron repeller grids, which are typically dc-biased to \(V_c \approx -150\) V. The resulting ion...
beam is then slowed by an ion repeller grid and an RF chamber which are dc biased to have slightly higher than the plasma potential. The relative densities of the molecular and atomic ions were determined from the collector current of the mass analyzer. The peaks appear in the collector current of the analyzer when the frequency of the applied RF electric field is equal to the ion cyclotron frequency \( f_c \equiv eB/(2\pi m) \). The current of an ion species \( j \) entering the mass analyzer should be proportional to the corresponding ion peak current \( I_{p,j} = e_n \nu_{i,j} S \), where \( \nu_{i,j} \) is the thermal velocity, and \( S \) is the area of the small hole in the end plate.

To model the ion density in this experiment, a simple zero-dimensional model is developed for solving the system of rate balance equations for ion and gas species [4, 7, 10]. The processes included in the model are

\[
\begin{align*}
H + e & \rightarrow H^+ + 2e, & \text{(el)} \\
H_2 + e & \rightarrow 2H + e, & \text{(eD)} \\
H_2 + e & \rightarrow H + H^+ + e, & \text{(eDn)} \\
H_2(v) + H^+ & \rightarrow H_2^+ + H, & \text{(CNV)} \\
H_2 + e & \rightarrow H_2^+ + e, & \text{(eI2)} \\
H_2 + e & \rightarrow H^+ + H + 2e, & \text{(elI)} \\
H_2^+ + e & \rightarrow H^+ + H, & \text{(DR2)} \\
H_2^+ + e & \rightarrow H^+ + H + e, & \text{(eIDI2)} \\
H_2^+ + e & \rightarrow 2H^+ + 2e, & \text{(elI2)} \\
H_2^+ + H_2 & \rightarrow H_2^+ + H, & \text{(CNV2)} \\
H_2^+ + e & \rightarrow H_2^+ + H + e, & \text{(eI3)} \\
H^+_2 + e & \rightarrow 3H + \begin{cases} 3H_2 + H, \quad \text{(DR3)} \\
H_2 + H, \quad \text{(BCNV2)} \end{cases}
\end{align*}
\]

Here, we have ignored radiative and three-body recombination, as well as dissociative electron attachment (H\(^+\) formation), as these reactions are believed to be negligible for the conditions studied here. The particle H\(^+\) refers to electronically excited atomic hydrogen. In the H\(^+_2\) recombination reaction (DR3), we have assumed a 50% branching ratio between the two reaction channels [11]. For the purposes of calculating the average velocities (\( \nu_{r,ij} \)), the neutral velocities are assumed to be negligible compared with the ion and electron velocities. The ion temperatures are taken to be \( T_j = T_e \) \((j = 1, 2, 3)\), where \( T_e \) is the electron temperature. The molecular ion density of H\(^+\), H\(^+_2\), and H\(^+_3\) are described by the steady-state rate balance equations with the charge neutrality \( n_e = \sum j \nu_j \) [4]. The neutral densities of hydrogen molecules is obtained from the measured neutral gas pressure \( P \). In evaluating the rate coefficients, the reactions involving H\(_3\), H\(_2^+\), and H\(^+_3\) are vibrationally resolved in the model. These rate coefficients are calculated by the available cross-section data [4, 10–13].

We obtain the distribution of ground-state quantum numbers from measured relative intensities of the H\(_2\) molecular radiation between the upper electronic state \( B^2\Sigma_u^+(v') \) and \( C^1\Pi_g(v') \) with vibrational level [14, 15]. The corona model used to calculate the population distribution of the vibrational levels resulting from a Boltzmann population distribution in the ground state characterized by a temperature of hydrogen molecule. Assuming a thermal (Boltzmann) population of ground state \( (X^1\Sigma_g^+) \) vibrational levels, and given the Franck-Condon matrix for electron-impact excitation from the ground state to the upper state, a ground-state vibrational temperature of hydrogen molecule \( T_{e,\text{vib}} \) can be found which results in a best fit to the observed relative intensities of VUV spectrum.

3. Experimental Results

Figure 2 shows the ion densities (\( n_{H^+} \), \( n_{H_2^+} \), and \( n_{H_3^+} \)), the electron density \( n_e \), and the electron temperature \( T_e \) plotted against hydrogen gas pressure \( P \) at the discharge current \( I_d \) of 50 A. Using a small amount of secondary hydrogen gas puffing into a hydrogen plasma, \( n_e \) has a maximum value of \( 5.0 \times 10^{18} \text{ m}^{-3} \) at \( P \sim 4 \text{ mtorr} \) and \( T_e \) decreases rapidly from 11 to 3 eV owing to ionization. Above \( P \sim 4 \text{ mtorr} \), \( T_e \) falls below 3 eV, and \( n_e \) gradually decreases. \( n_{H^+} \) shows the same tendency as the characteristics of \( n_e \) with increasing \( P \) and has a maximum value of \( \sim 4.5 \times 10^{18} \text{ m}^{-3} \) at \( P \sim 4 \text{ mtorr} \). On the other hand, \( n_{H_2^+} \) and \( n_{H_3^+} \) decrease and have a minimum values of \( \sim 2.0-3.0 \times 10^{17} \text{ m}^{-3} \) at \( P \sim 4 \text{ mtorr} \). After that, \( n_{H_2^+} \) and \( n_{H_3^+} \) have a maximum value at \( P \sim 6 \text{ mtorr} \) and decrease with increasing \( P \).

By defining the ion density ratio as the individual val-
Fig. 2 The ion densities (n_H^+, n_H2^+, and n_H3^+), the electron density n_e, and the electron temperature T_e plotted against hydrogen gas pressure P at the discharge current I_d of 50 A in hydrogen plasma.

Fig. 3 Measured the ion densities ratio (H^+, H2^+, and H3^+) and the calculated ion densities ratio ⟨H^+⟩, ⟨H2^+⟩, and ⟨H3^+⟩) plotted against gas pressure P at the discharge current I_d of 50 A in the hydrogen plasma.

ues of the ion densities divided by the sum of the densities of H^+, H2^+, and H3^+, we can express the effect of molecular processes on the hydrogen plasma. Figure 3 shows the measured ion densities ratio (H^+, H2^+, and H3^+) and the calculated ion densities ratio ⟨H^+⟩, ⟨H2^+⟩, and ⟨H3^+⟩) plotted against gas pressure P at the discharge current I_d of 50 A in the hydrogen plasma. In the reactions involving H2, H2^+, and H3, a ground-state vibrational temperature of hydrogen molecule are T_vib = 0 K and T_vib ~ 3000 K in the model. The ion density ratio of H^+ is larger than that of H2^+ or H3^+ in the low gas pressure (< 6 mtorr). With increasing P, the ion density ratio of H^+ has a maximum value of 0.8 at P ~ 4 mtorr and gradually decreases from 0.8 to 0.4. On the other hand, the ion density ratio of H2^+ rapidly increases from 0.1 to 0.5 with increasing P and saturated up to P ~ 10 mtorr. The density ratio of H3^+ remains nearly constant at around 0.1-0.2 with increasing P. In case of T_vib = 3000 K in the reactions involving H2, H2^+, and H3, the characteristic of (H^+), (H2^+), and (H3^+) show the same tendency as that of the measured ion density ratio. (H^+) has a maximum value of 0.8 at P ~ 4 mtorr and gradually decreases with increasing P. (H2^+) gradually decreases with increasing P and (H3^+) increases above P ~ 10 mtorr. On the other hand, in case of T_vib = 0 K, the characteristic of the calculated ion density ratio shows the different tendency as that of the measured ion density ratio. (H^+) has no maximum value and gradually decreases with increasing P. In particular, (H2^+) are one-half magnitude smaller than that of measured H2^+ above P ~ 8 mtorr. The value of (n_H2^+) gradually decreases with increasing P and (n_H3^+) increases. This discrepancy may be attributed to the fact that the reactions involving H2, H2^+ are not vibrationally resolved in the model for solving the system of rate balance equations for ion and gas species. Neglecting vibrational excitation of H2 would have resulted in a significant understimation of the molecular ion density in these experiments.

Figure 4 shows the measured the ion densities ratio (H^+, H2^+, and H3^+), calculated the ion densities ratio ⟨H^+⟩, ⟨H2^+⟩, and ⟨H3^+⟩) with T_vib = 3000 K, the electron temperature T_e, and the electron density n_e plotted against discharge current I_d at gas pressure of 8 mtorr. In this condition, n_e increases with increasing I_d, while T_e is nearly constant value of 0.5-1.0 eV and the very bright visible light emission in front of the endplate were observed in the detached plasma. In the low discharge current (I_d < 40 A), the measured ion density ratio of H^+ is larger than that of H^+. Above I_d ~ 50 A, the ion density ratio of H^+ is smaller than that of H^+ for the high density plasma. Also, the vibrational resolved model was found to predict the observed dominant ion density ratio.
4. Conclusion

We have carried out the experimental observation and modeling of molecular ions in hydrogen plasma in a linear plasma device, TPD-SheetIV. From a zero-dimensional model using the relevant rate balance equations, the calculated molecular ion densities of H^+ and H^+_2 was found to predict the observed dominant ion density ratio, demonstrating the importance of vibrational temperature of hydrogen molecules. For high density plasma, the effect of molecules on recombination tends to decrease and contributes to assist dissociation and ionization. The conversion of H^+_2 into H^+_3 (CNV2), however, tends to increase and the plasma will contain H^+_3 for low electron temperature using a massive secondary hydrogen gas puffing. Through H_2(v) + H^+_2 or H^+_3 collisions are ignored here, it is conceivable that the creation and destruction of H^+_2 and H^+_3 play a significant role by contribution to the observed high level of H_2(v) internal excitation [4, 9]. Despite the reasonably agreement observed here between our predictions and the experimental data, a need for future modeling to improve understanding of the high level of H_2(v).

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