Experimental Evidence of Molecular Activated Recombination in Detached Recombining Plasmas

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Molecular activated recombination (MAR) has been clearly observed for the first time in a divertor plasma simulator. A small amount of hydrogen gas puffing into a helium plasma strongly reduced the ion particle flux along the magnetic field, although the conventional radiative and three-body recombination processes were quenched. Careful comparison of the observed helium Balmer spectra with collisional radiative atomic and molecular data indicates that the population distribution over the atomic levels with relatively low principal quantum numbers can be well explained by taking the MAR effects into account. [S0031-9007(98)06573-9]

Recently, volumetric plasma recombinations have attracted considerable interest in detached plasmas observed in tokamak magnetic divertors and in linear divertor plasma simulators [1–5]. The plasma recombination is expected to play an essential role in strong reduction of ion particle flux along the magnetic field, resulting in a decrease in the heat flux to plasma-facing components [6]. Continuum and series of visible line emissions from highly excited levels were observed in detached plasmas in these devices. The analysis of these spectra shows that the radiative and three-body recombination (EIR) is important for divertor plasma conditions and gives the electron temperature T_e of less than 0.4 eV in detached pure helium plasmas in linear devices [5] and around 1 eV in detached hydrogen plasmas in tokamaks [3].

On the other hand, the importance of another recombination process associated with molecular reactions, that is, the molecular activated recombination (MAR) involving a vibrationally excited hydrogen molecule such as $H_2(v) + e \rightarrow H^- + H$ followed by $H^- + A^+ \rightarrow H + A$, and $H_2(v) + A^+ \rightarrow (AH)^+ + H$ followed by $(AH)^+ + e \rightarrow A + H$, where $A^+(A)$ is the hydrogen or the impurity ion (atom) existing in divertor plasmas, was pointed out in theoretical investigations and modeling [7–9]. MAR is expected to lead to an enhancement of the reduction of ion particle flux, and to modify the structure of detached recombining plasmas because the rate coefficient of MAR is much greater than that of EIR at relatively high T_e above 0.5 eV as shown in Fig. 1 [9]. Therefore, in order to control a huge amount of ion particle and heat fluxes to the plasma-facing components in next generation fusion devices intended to have a long pulse or a steady state operation, a deep understanding of such a detached plasma regime associated with MAR effects is one of the most urgent issues in a magnetic confinement fusion research. However, no clear experimental evidence of MAR has been reported so far in a relevant plasma to the divertor condition, in which the plasma density is more than about 10^{19} m^{-3} and the neutral gas pressure is around 10 mtorr.

Studies of weakly ionized plasmas with high neutral pressure, such as discharge for gaseous lasers [10] and plasma jets [11], also show that the conversion of atomic ions into molecular one as a result of ion-neutral reactions, and electron attachment to the neutrals may drastically increase the plasma sink due to fast dissociative and charge-exchange recombination. However, these conditions are quite different from the divertor plasmas.

The present paper gives the first experimental evidence of MAR observed clearly in hydrogen/helium mixture plasmas. A low concentration of hydrogen molecules exceeding some critical level in a helium plasma leads to a strong reduction of ion particle flux along the magnetic field. We have analyzed the population of the excited states of the helium atom by using collisional radiative atomic molecular data model (CRAMD code [9]) to show that the intensities of the observed helium Balmar spectra
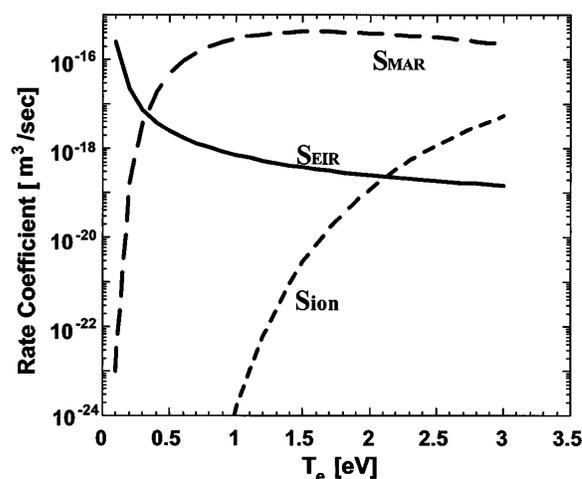
FIG. 1. Rate coefficients of collisional processes as a function of electron temperature; electron-ion recombination (radiative and three-body recombination) S_{EIR}, molecular activated recombination S_{MAR}, and electron impact ionization S_{ion}, where the electron density is assumed to be 5×10^{18} m^{-3}.

in a hydrogen/helium mixture plasma can be explained by the effect associated with MAR, because the MAR produces neutrals in a low excited or ground states in contrast to the case of EIR where originated neutrals are in highly excited states.

The experiment was performed in the linear divertor plasma simulator NAGDIS-II [12] as shown in Fig. 2. Helium plasmas are produced by the modified TP-D type dc discharge [12]. The neutral pressure P in the divertor test region can be controlled from 1.0 to 20 mtorr by feeding a secondary gas and/or changing the pumping speed. The change of P in the divertor test region has no effect on the plasma production in the discharge region due to 3 orders of magnitude pressure difference between the discharge and the divertor test regions. Spectra of visible light emissions are detected at two different axial positions of $X = 0.92$ and 1.58 m from the discharge anode. Two sets of fast scanning probes are also installed at the same positions to measure plasma parameters.

First we generated the helium plasma at a discharge current $I_d = 50$ A without any secondary gas puff, where the electron density n_e and T_e in the upstream ($X = 0.92$ m) are 6×10^{18} m^{-3} and 3.5 eV, respectively. P is kept to be 5.0 mtorr, which is a critical value for the plasma to start to detach from the target plate, by controlling the pumping speed. In this detached helium plasma regime, our recent experimental studies have shown that the electron-ion energy exchange process followed by the ion-neutral charge exchange is a key to reduce T_e along the magnetic field to a temperature less than 1 eV, where the EIR occurs, leading to the detached plasmas [13,14].

Figure 3 shows the change in spectrum of visible light emission from 310 through 370 nm observed in the downstream ($X = 1.58$ m) with a hydrogen or helium gas puff. For a pure helium plasma, a continuum and a series of visible line emissions from highly excited levels due to the conventional EIR were observed as shown in Figs. 3(c) and 3(d) [5,12]. Detailed analysis of the population distribution over the highly excited levels shows that T_e is about 0.4 eV by using the Boltzmann relation, because the
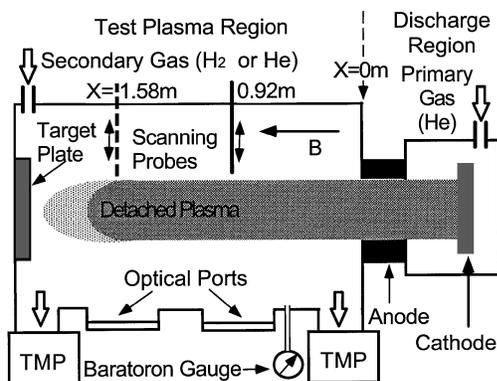
population follows the Saha-Boltzmann distribution; that is, those excited states above a critical quantum number ($n \sim 5$) are in local thermodynamic equilibrium (LTE) with free electrons in the plasma [5]. Analysis of the photon energy dependence of the continuum emission intensities also provides the same value of T_e [5].

When a hydrogen gas was introduced into the helium plasma at a low hydrogen gas concentration as shown in Fig. 3(a) at the total pressure of $P \sim 5.5$ mtorr, corresponding to the partial pressure of hydrogen gas ~ 0.5 mtorr, the continuum and the series of visible line emissions become weak, but can still be observed. They are similar to that in pure helium as shown in Fig. 3(c). On the other hand, the spectrum is found to be changed dramatically in Fig. 3(b) when the partial pressure of hydrogen gas exceeds a critical level ~ 1.4 mtorr. There are neither continuum nor series of visible line emissions. It shows that EIR does not occur at all in this plasma condition, which means that T_e goes up or n_e goes down with the hydrogen gas puff. Radial profiles of the ion flux measured both in the upstream and the downstream are illustrated in the insets of Figs. 3(a)–3(d). The reduction of the ion flux along the magnetic field due to the EIR is found in the right-hand insets of Figs. 3(c) and 3(d). The reduction rates of the ion flux from the upstream to the downstream in Figs. 3(c) and 3(d) are almost the same, because of a very small helium pressure difference. On the other hand, in the inset of Fig. 3(b), we can see a strong reduction of the ion flux by the addition of a small amount of hydrogen gas associated with the complete quenching of the EIR processes. Moreover, the ion flux in the upstream has already decreased compared to that in the case of the pure helium detached plasma. This means that some plasma volumetric recombination process already starts to occur in the upstream, where T_e is relatively high. This is well reproduced by the one-dimensional fluid simulation for the present configuration [15,16]. From these experimental results, we can conclude that there is the plasma volumetric recombination process coming from the effect of the molecular hydrogen (MAR) in our helium/hydrogen mixture plasma.

The observed helium Balmer series spectra were analyzed with the CRAMD code [9] by adjusting the source of the population of the excited state of helium atoms, corresponding to (i) EIR, (ii) electron impact excitation from the ground state of atoms, and (iii) MAR. It should be noted that this analysis does not directly involve kinetics of long living vibrationally excited states of hydrogen molecules, where an applicability of local collisional-radiative approximation is questionable [17]. Precise comparison between the experimental spectra and the relative line intensities obtained with the CRAMD code gives us which is the dominant population mechanism among the above three candidates.

First of all, we start to analyze the helium Balmer series spectra in *pure* helium detached plasma, corresponding to Fig. 3(c) because the basic characterization of bulk

FIG. 2. Schematic of experimental apparatus NAGDIS-II. The running magnetic field strength is 0.25 T. The length of the plasma in the test plasma region is 2 m and the diameter of the vacuum vessel is 0.19 m.
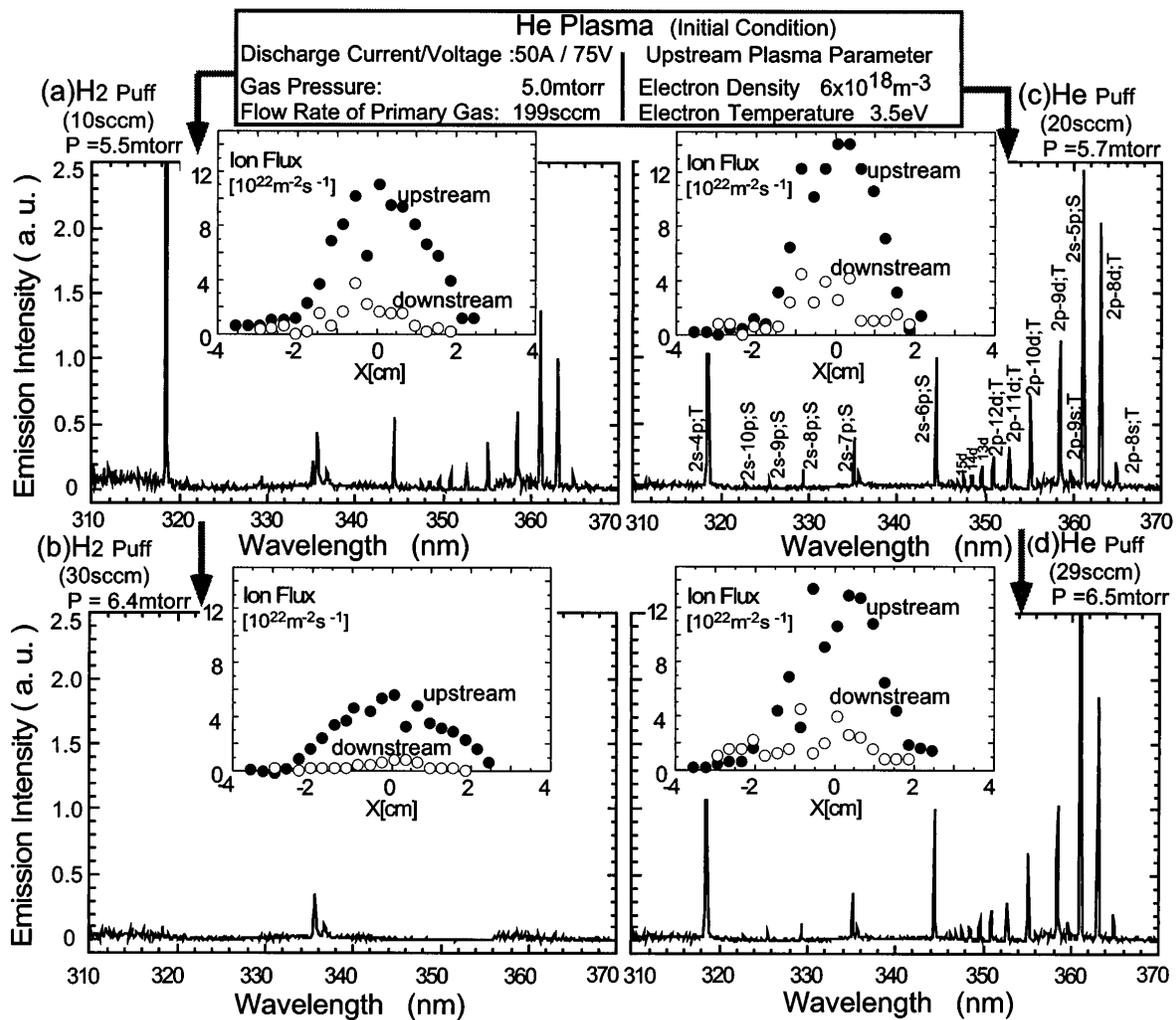
FIG. 3. (a) and (b): Visible light emission spectra from helium plasmas with hydrogen gas puff and [(c) and (d)] with helium gas puff. The insets show the radial profiles of the ion particle flux. Closed circles and open ones are obtained in the upstream ($X = 0.92$ m) and in the downstream ($X = 1.58$ m), respectively.

helium plasmas in the present device should give the basis for the later analysis on the helium Balmer series spectra in helium/hydrogen *mixture* plasmas. Solid circles in Fig. 4(a) show the observed intensities of the helium Balmer series from $n \sim 3$ to 7 normalized by the intensity at 668.3 nm ($n \sim 3$) in the *pure* helium detached plasma. In this case, only EIR and the electron impact excitation from the ground state contribute to the population in excited states. Open circles in Fig. 4(a) show the calculated results with the CRAMD code at $T_e \sim 0.5$ eV, in which the variation of intensities from $n = 3$ through 7 is much weaker than that observed experimentally. It means that the EIR alone cannot reproduce the observed distribution of intensities in such a low T_e. The high population of low n ($n \sim 3$ and 4) relative to that of high n ($n \sim 6$ and 7) corresponds to that the excitation from the ground state with relatively high T_e. However, such a high T_e contradicts the observation of EIR as shown in Fig. 3. Therefore, the observed spectrum cannot be matched with a single Maxwellian distribution for electrons. However,

in the dc discharge system like the present device, the existence of a very small amount of electron beam components, which can be generated by the primary electrons with the energies somewhat below 100 eV from the cathode surface entering to the divertor test region directly, is thought to be a realistic situation, although the gas pressure in the discharge region is quite high. The assumption about a small fraction of the fast electrons allows drastically to improve the matching of Balmer series spectra in the *pure* helium detached plasma. Calculated intensities are shown by open triangles and squares in Fig. 4(a) for the electron beam density n_b of 0.1% of n_e with the beam energy E_b of 25 eV, corresponding to $n_e \sim 10^{18}$ and 10^{19} m^{-3}, respectively. Moreover, a further analysis shows a very weak dependence of relative Balmer series line intensities on E_b within the range $\sim$10–50 eV, which is reasonable from the value of the discharge voltage, roughly 100 eV. The ratio n_b/n_e changed from 0.1% to 0.001% also gives almost the same variation of line intensities over n.
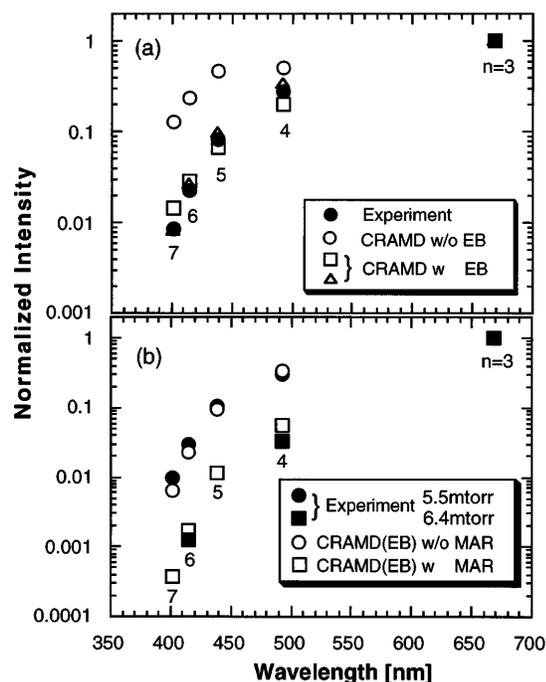
FIG. 4. Emission intensities of helium Balmer series from (a) pure helium detached plasma at $P \sim 5.7$ mtorr; (b) helium and hydrogen mixture detached plasma. In (a), solid circles are experimental data. Open circles are obtained with the CRAMD code assuming $T_e \sim 0.5$ eV without any nonthermal electrons. Triangles and squares are obtained by taking account of the electron beam component, whose density n_b is 0.1% of $n_e \sim 10^{18}$ and 10^{19} m^{-3}, respectively. In (b), solid circles and solid squares are measured intensities at $P \sim 5.4$ and 6.4 mtorr, corresponding to Figs. 3(a) and 3(b). Calculation by taking account of the electron beam component at $n_b/n_e \sim 0.1\%$ gives open circles. Open squares are obtained by taking account of MAR as well as the effect of the same electron beam component as in the pure helium case.

We now consider the analysis on the Balmer series spectra in helium plasma with the hydrogen gas puff. Figure 4(b) shows the normalized intensities of helium Balmer series in this condition. Hydrogen molecules are assumed to be vibrationally excited by the bulk electrons with $T_e \sim 0.5$ eV, because the rate coefficient of the vibrational excitation peaks around $T_e \sim 1.0$ eV. On the other hand, the electron beam component has little effect on the generation of the vibrationally excited hydrogen molecules, because of the low electron beam density n_b of 0.1% of n_e and the high beam energy E_b of 25 eV. Solid circles and solid squares are experimental data corresponding to Figs. 3(a) and 3(b). The distribution of intensities at a low concentration of hydrogen gas at $P \sim 5.5$ mtorr is found to be in good agreement with the CRAMD results shown by open circles, taking the electron beam component into account. On the other hand, at the concentration of hydrogen gas above a critical level where the total pressure is 6.4 mtorr, the line emission intensity from $n \sim 4$ has already decreased by more than an order of magnitude comparing to $n \sim 3$. This kind of strong drop of emission intensity with an increase in n contra-

dicts the CRAMD results with the EIR and the electron beam effect but without the MAR. On the contrary, an inclusion of the MAR effect, corresponding to open squares in Fig. 4(b), gives a very good agreement with experimental data. Thus, the analysis of experimentally observed Balmer series spectra in the helium/hydrogen mixture plasma also support the fact that the MAR becomes dominating in the experimental environment with the hydrogen gas pressure above 1.4 mtorr.

In conclusion, we have performed the experiments on hydrogen gas puffing to the helium plasma to show clearly the MAR for the first time in the divertor plasma condition. At a hydrogen gas pressure above some critical level in the helium plasma, the continuum and line emissions due to the conventional EIR disappeared, but the strong reduction of ion particle flux was observed over a relatively long path length along the magnetic field associated with the MAR. Detailed analysis on the intensities of helium Balmer series spectra also indicates that the observed rapid decrease of intensities with the principal quantum number can be explained only by taking account of the MAR.

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