Measurement of ro-vibrational temperatures using Fulcher-α band in H_2 and D_2 mixture plasmas

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Introduction

In the divertor region of fusion relevant magnetic confinement devices, significance of ro-vibrationally excited hydrogen molecules has been emphasized. For divertor detachment operation, recombination rate coefficient can be enhanced through molecular assisted recombination (MAR) process. Meanwhile, production of so-called Franck-Condon atoms, which have higher energy (typically 2 to 3 eV) compared to those produced by electron-ion recombination (< 1 eV), determines the neutral penetration depth. Ro-vibrational temperatures of hydrogen molecules have also been utilized for diagnostics. In a simple negative-ion source system, translational temperature was estimated from the rotational temperature based on the translation-rotation relaxation[1], and a possibility of extension to surface temperature deduction in tokamak devices has been shown[2, 3]. In general, the dominant processes which control the ro-vibrational temperatures are (i) electron impact processes in plasma, (ii) surface recombinations (Eley-Rideal reaction), and (iii) internal energy transfer due to the inter-molecular collision. However, in the fusion application, less attention to the processes (ii) and (iii) has been paid. In this work, we performed experiments using a hollow-cathode glow discharge chamber aiming at clarifying the contributions of the processes (i) to (iii). The ro-vibrational temperatures were evaluated from Q-branch spectra of H_2 Fulcher-α transition ($d^3\Pi_u^- \rightarrow a^3\Sigma_g^+$) with changing the plasma parameters and cathode surface temperature. Isotope effect on the observed ro-vibrational temperatures was also investigated using H_2 and D_2 mixture plasma.

Coronal model

Since ro-vibrational temperatures in electronic excited states of hydrogen molecules are usually not in the thermal equilibrium with the ground state ($X^1\Sigma_g^+$), the coronal model was used for the evaluation of the ground state ro-vibrational temperatures[4, 5]. The model deals with a balance between the electron-impact excitation from X to $d-$state and radiative de-excitation from d to $a-$state as

$$n_{dv'N'} = n_e \sum_{v,N} [n_{XvN} R_{XvN}^{dv'N'}] / \sum_{v''N''} A_{av''N''}^{dv'N'},$$

(1)

where v and N are the vibrational and rotational quantum numbers, respectively, n_{XvN} is the X-state population, $R_{XvN}^{dv'N'}$ is the electron-impact excitation coefficient, and $A_{av''N''}^{dv'N'}$ is the spontaneous emission coefficient. Assuming the thermal equilibrium for ro-vibrational distribution in the X-state, the population n_{XvN} is written as

$$n_{XvN} = n_{X_0} \exp\left[-\frac{\Delta G(v)}{kT_{\text{vib,X}}}\right] \frac{g_{\text{as}}(2N+1)\exp\left[-\frac{\Delta F(N,v)}{kT_{\text{rot,X}}}\right]}{\sum_N^\infty g_{\text{as}}(2N+1)\exp\left[-\frac{\Delta F(N,v)}{kT_{\text{rot,X}}}\right]}, \tag{2}$$

where n_{X_0} is the population in the vibrational ground state, g_{as} is the nuclear-spin statistical weight, $\Delta G(v)$ and $\Delta F(N,v)$ are the vibrational and rotational energy from their ground states, respectively, and $T_{\text{vib,X}}$ and $T_{\text{rot,X}}$ are the X-state vibrational and rotational temperature, respectively. $T_{\text{vib,X}}$ and $T_{\text{rot,X}}$ can be deduced using Eqs. (1) and (2) from the experimentally observed excited state population $n_{dv'N'}$.

Experimental setup

A hollow-cathode discharge chamber employed for the experiment has been described in detail previously[6]. DC-glow discharge was sustained between an aluminum cathode and a copper anode using working gases of H_2 (21, 75, 120, 180 Pa) or equal mixture of H_2 and D_2 (41 Pa). The volume of generated plasma which corresponds to the cathode dimension is about 60 mm in length and 19 mm in diameter. In order to observe the dependence on the plasma parameters as well as on the cathode surface temperature, the discharge current was changed from 10 to 70 mA. A double probe made of molybdenum electrodes and glass insulator was used for the measurement of electron temperature and density, while a 1 m Czerny-Turner spectrometer equipped with a 2400 Grooves/mm holographic grating and a photo-multiplier tube (Hamamatsu R928) was used for optical emission spectroscopy. Measurements were performed with a slit width of 60 μm, which gives a wavelength resolution of $\Delta\lambda_{\text{fwhm}} \simeq 0.018$ nm. For monitoring the cathode surface temperature, a K-type thermocouple was adopted. The thermocouple voltage was converted to temperature using NI cRIO-9211 module (National Instruments) with a resolution of 24 bit.

Relation between ro-vibrational temperatures and surface temperature

Figure 1 shows the evaluated plasma parameters. Electron temperature (T_e) is varied between 3 to 9 eV, while electron density (n_e) between 1×10^{15} to 4×10^{17} m^{-3}. The evaluated $T_{\text{rot,X}}$ with respect to the cathode surface temperature T_{cathode} is plotted in Figure 2. The processes which determine $T_{\text{rot,X}}$ are (i) electron-impact rotational excitation, (ii) surface recombination, (iii) rotation-translation energy transfer, and (iv) vibration-rotation energy transfer. Time scale
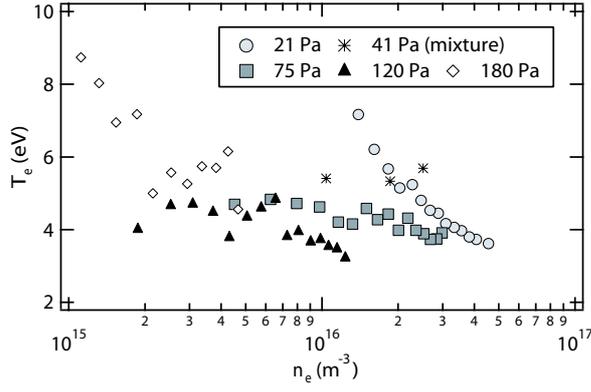
Figure 1: T_e and n_e evaluated by a double probe.
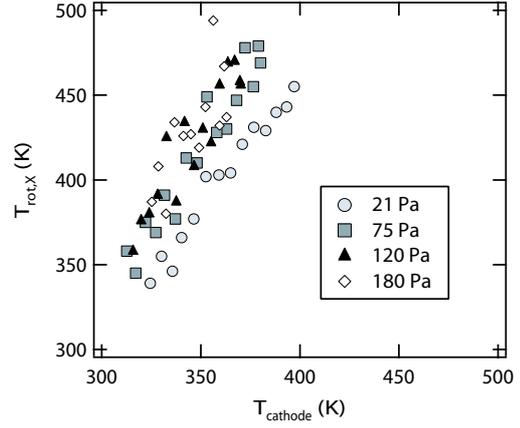
Figure 2: $T_{rot,X}$ with respect to $T_{cathode}$.

of rotational de-excitation ($N = 2 \rightarrow 0$ or $3 \rightarrow 1$) due to the process (iii) was evaluated based on a quantum mechanical calculation[7] as $\tau_{rt} = 1.3 \times 10^{-4}$ (21 Pa) to 1.4×10^{-5} (180 Pa) sec. The residence time of molecules, on the other hand, was estimated using a three-dimensional random-walk model to be $\tau_{res} = 3.2 \times 10^{-2}$ (21 Pa) to 2.9 (180 Pa) sec. Since the relaxation time between the translational and surface temperature is sufficiently short, clear correlation between $T_{rot,X}$ and $T_{cathode}$ in Figure 2 implies the rotation-translation energy transfer ($\tau_{rt} \ll \tau_{res}$). An increase in $\Delta T_{rot,X} = T_{rot,X} - T_{cathode}$ with increasing n_e was observed for each pressure condition, indicating the contribution of the electron-impact excitation. Moreover, vibration-rotation energy transfer accounts for a tendency that the absolute value of $\Delta T_{rot,X}$ becomes larger for higher pressure. The effect of surface recombination is not clear from the present results, and verification of it is a task for the future study.

The relation between measured $T_{vib,X}$ and $T_{cathode}$ is shown in Figure 3. In contrast to $T_{rot,X}$, no correlation was observed, and only weak dependence on n_e can be seen. The processes which determine $T_{vib,X}$ are (i) electron-impact processes including excitation and dissociative attachment, (ii) surface recombination, (iii) vibration-translation energy transfer, and (iv) vibration-rotation energy trans- fer. Time scale of vibrational de- excitation ($v = 1 \rightarrow 0$) via the process (iii) was evaluated from the quantum mechanical calcula- tion as $\tau_{vt} = 3.8$ (21 Pa) to 0.4 sec (180 Pa), which are compara- ble to the residence time ($\tau_{vt} \sim \tau_{res}$). However, experimental re-
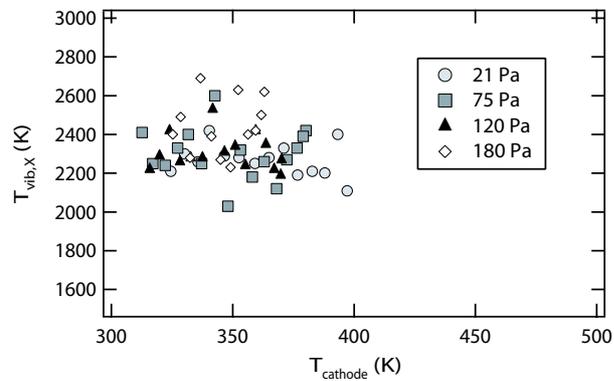
Figure 3: $T_{vib,X}$ with respect to $T_{cathode}$.

sults suggest that the effect is not large. The effect of the process (iv) is as small as the order of 10 K. The weak dependence on T_{cathode} and n_e, therefore, may indicate the importance of the surface recombination process.

Isotope effect

The difference of ro-vibrational temperatures in H_2, HD, and D_2 isotopes was investigated. Dependence of $T_{\text{rot,X}}$ and $T_{\text{vib,X}}$ on the cathode surface temperature was observed to be similar for all the isotopes. Table 1 shows the ro-vibrational temperatures normalized to the rotational (B_e) and vibrational (ω_e) molecular constants. These ratios are relative values to H_2. The dependence of $T_{\text{rot,X}}/B_e$ on the isotopes was observed, suggesting the results of the relaxations. The almost equal value of $T_{\text{vib,X}}/\omega_e$ indicates that the difference in $T_{\text{vib,X}}$ can be attributed to the difference of the molecular mass. In other words, the total vibrational energy is almost equal for all the isotopes and the relaxation processes (iii) and (iv) are ineffective.

T_{cathode}	$T_{\text{rot,X}}/B_e$ for H_2	HD	D_2	$T_{\text{vib,X}}/\omega_e$ for H_2	HD	D_2
327 K	1.0 (390 K)	1.0 (300)	1.8 (360)	1.0 (2570 K)	1.1 (2360)	1.2 (2140)
357 K	1.2 (450 K)	1.2 (350)	2.2 (420)	0.95 (2440 K)	1.0 (2290)	1.2 (2110)
390 K	1.3 (510 K)	1.6 (480)	2.1 (410)	1.0 (2680 K)	1.1 (2440)	1.3 (2300)

Table 1: Normalized ro-vibrational temperatures.

Conclusion

Ro-vibrational temperatures of hydrogen molecules were measured using a hollow-cathode glow discharge plasma. Observed correlation between $T_{\text{rot,X}}$ and T_{cathode} indicates the dominance of the rotation-translation energy transfer. For $T_{\text{vib,X}}$, on the other hand, no correlation can be seen. The experimental results for the isotope molecules may support these evaluations. The dependence of $T_{\text{rot,X}}/B_e$ on the isotopes indicates that the relaxation processes are effective, while the almost equal value of $T_{\text{vib,X}}/\omega_e$ suggests the opposite. Further elucidation including the surface recombination process is a task for the future study.

Acknowledgments

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