Measurement and modeling of hydrogen molecule rovibrational accommodation on graphite

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Molecular hydrogen is the working gas in most tokamak experiments and is widely distributed in the plasma edge [1]. Accurate modeling of the cold edge region of these devices therefore requires inclusion of collisions with hydrogen molecules, and these collisions rates are typically very sensitive to the internal energy (rotational temperature T_{rot} and vibrational temperature T_{vib}) of the H_2 molecule [2]. Experiments suggest that cooling of T_{rot} and T_{vib} dominantly results from H_2-wall collisions [3]. Measurements and modeling of the rovibrational energy accommodation rate of H_2 on various tokamak wall materials, such as graphite, is therefore desirable.
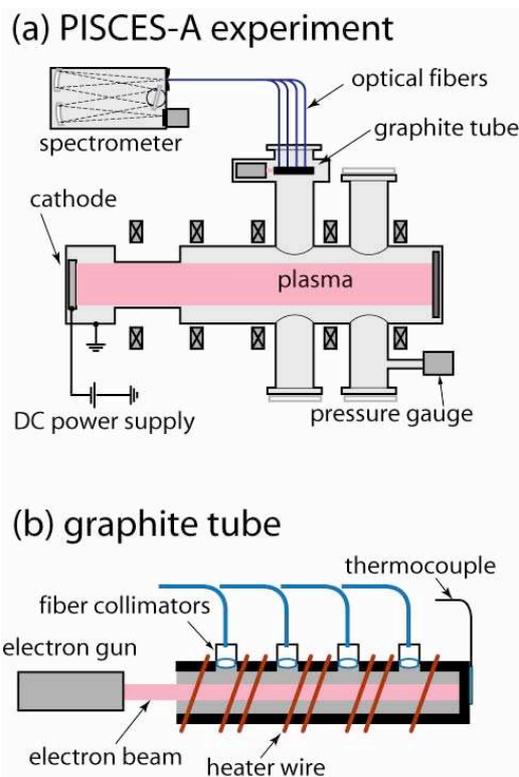

Fig. 1. (a) overview of PISCES-A and (b) close-up of graphite tube mounted in side port.

An overview of the experimental setup is shown in Fig. 1(a). The PISCES-A reflex-arc plasma discharge [4] is used to create rovibrationally excited hydrogen molecules which fill the vacuum chamber. The heated H_2 neutrals travel down a stainless steel side port and then enter a small graphite tube mounted in a side port. An electron beam is aimed down the hollow graphite tube to electronically excite a small fraction of the hydrogen molecules inside the tube. The resulting line emission is then measured at 4 positions in the tube. The local H_2 ground electronic state vibrational and rotational temperatures T_{vib} and T_{rot} at each fiber location can then calculated with measured line ratios of H_2 Fulcher band emission [5,6]. Additionally, H_2

kinetic temperature T_{kin} and density n_{H2} can be obtained from line broadening and line intensity, respectively.

Details of the graphite tube arrangement are shown in Fig. 1(b). Collimator lenses are used to increase the amount of light gathered into the four fibers. Heater wire wrapped around the graphite tube is used for heating. The graphite tube temperature (assumed to be uniform) is measured with a thermocouple at the end of the tube. The graphite tube is made of E-294 graphite from Electro-Tech Machining with density $\rho = 1.80$ g/cm^3 and average grain size of 0.10 mm. The tube dimensions are: length $L = 10$ cm, diameter $D = 3.8$ cm, and thickness $t = 0.64$ cm. Prior to taking data, the graphite tube was baked out for 4 hours at 1000 C to remove stored water. The graphite tube was not exposed to plasma prior to the experiments, so hydrogen in the walls arrives as neutrals, not as embedded ions.

The electron beam is created using a $D = 1.2$ mm Ta disk cathode emitter. Typical beam energies are 70 V; this is chosen to be well above the threshold for H_2 electronic excitation (~10 eV) so that signal brightness is not sensitive to small changes in beam energy. Beam currents (~1 mA) are small and heating of H_2 in the tube due to the beam is found to be negligible.

To interpret the measurements, Monte Carlo modeling of H_2 neutral trajectories is used [7]. The free parameters are the temperatures $T_{vib,0}$ and $T_{rot,0}$ at the tube entrance and the accommodation coefficients α_{rot} and α_{vib}. Wall collisions are assumed to be diffuse. The kinetic temperature is assumed to always equal the rotational temperature, $T_{kin} = T_{rot}$, as is seen in the experiments. Because T_{vib} is found to be much larger ($\approx 5 - 10 \times$) than T_{rot}, direct mixing between T_{rot} and T_{vib} in wall collisions assumed to be small and is neglected. Neutral-neutral collisions and rovibrational energy transfer between vibrational energy and kinetic/rotational energy in these collisions is included in the Monte-Carlo modeling. Neutral density is obtained from the brightness of the H_2 lines; this is found to depend dominantly on the temperature of the
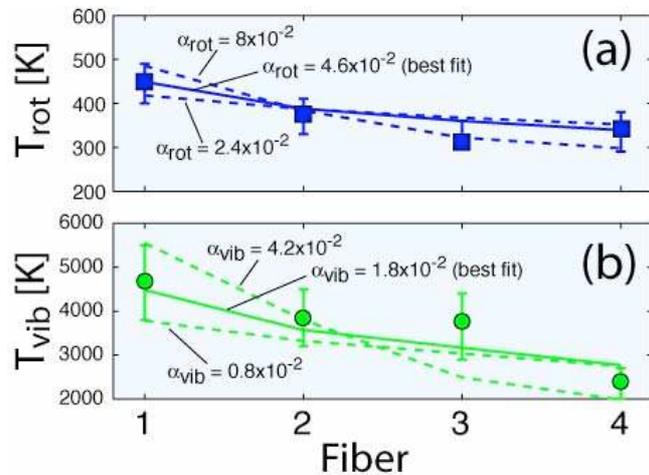

Fig. 2. Monte-Carlo fits to (a) T_{rot} vs axial position and (b) T_{vib} vs axial position.

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graphite tube. The standard definition of the accommodation coefficient is used [8], i.e. after a wall collision the new rotational temperature is $T'_{rot} = T_{rot} - \alpha_{rot}(T_{rot} - T_s)$, where T_s is the surface temperature. An analogous definition is used for α_{vib}.

Figure 2 gives an example of Monte-Carlo fits to the measured (a) T_{rot} and (b) T_{vib}. The experiment shown here was for room-temperature graphite. The free parameters $T_{rot,0}$, $T_{vib,0}$, α_{rot}, and α_{vib} were varied to obtain a best fit shown by the solid lines. The error in the accommodation coefficients was estimated by finding the highest and lowest values of α_{rot}, and α_{vib} which would still fit the data within the error bars; these extreme fits are shown by the dashed curves in Fig. 2. Error bars on T_{rot} and T_{vib} data are estimated from the quality of the single-temperature Boltzmann fits to the line populations.

Motivated by these measurements, we have recently undertaken a series of large-scale classical molecular dynamics (MD) simulations of H_2-carbon surface interactions. Our previous calculations [9] were focused on modeling chemical sputtering, implantation, reflections and dissociation processes of hydrogenated carbon from well-defined low-intensity ion beam experiments [10]. In this work, we extend our previous approach to model a high-density plasma-bombardment environment with randomization of the simulated particle impact parameters to represent the plasma. For each surface, after defining a hydrogenation level, surface temperature, and rovibrational, rotational, and kinetic temperatures of the ambient H_2, the irradiation is repeated for 1000 random independent trajectories. The center-of-mass, rotational, rovibrational and vibrational energies and corresponding temperatures, consistent with the degrees of freedom for each mode of the molecule motion, were determined for each reflected H_2 molecule, and averaged over all trajectories. The number of trajectories for each surface was satisfactorily large, reflected in less than 5% statistical uncertainty (one standard deviation) of the calculated averages.

The classical MD treatment of all degrees of internal motion of the H_2 molecule possibly leads to the enhanced coupling (mixing) of vibrational and rotational modes of motion, which is quantum-mechanically suppressed due to a large difference in energy of the vibrational and rotational quanta and the low level of vibrational excitation in the present experiments. Therefore, our simulation results are best considered in terms lf the total rovibrational energy accommodation coefficient $\alpha_{rovib} = \dfrac{\alpha_{rot} + Q\alpha_{vib}}{1+Q}$, where $Q = \dfrac{T_S \, \rho \, T_{0,vib}}{T_S \, \rho \, T_{0,rot}}$.

Since $T_{0,vib} \gg T_{0,rot}$, it follows $|Q| \gg 1$ and α_{rovib} is therefore is dominated by the vibrational

accommodation coefficient. A comparison between the measured (open points) and calculated (solid) coefficients α_{rovib} is shown in Fig. 3. Since the level of hydrogenization of the carbon surface is not known in the experiment, the calculation is shown for both a bare amorphous C surface (black line), and for a fully hydrogenated surface (red line). The dashed line indicates the average of the results from all studied surfaces. Overall, the measured accommodation α_{rovib} can be seen to agree reasonably well with the simulations at larger surface temperature. Within the scatter of the data, D_2 accommodation (green points) is similar to H_2 accommodation (blue points). A slight increase in accommodation with increasing surface temperature is suggested by the data. All values, measured and simulated, are much larger than expected by analytical phonon excitation estimates [11], demonstrating that hydrogen trapping and de-trapping (which is included in the MD simulations) is a crucial process here. Future work will attempt to improve both simulated and measured hydrogen accommodation coefficients.

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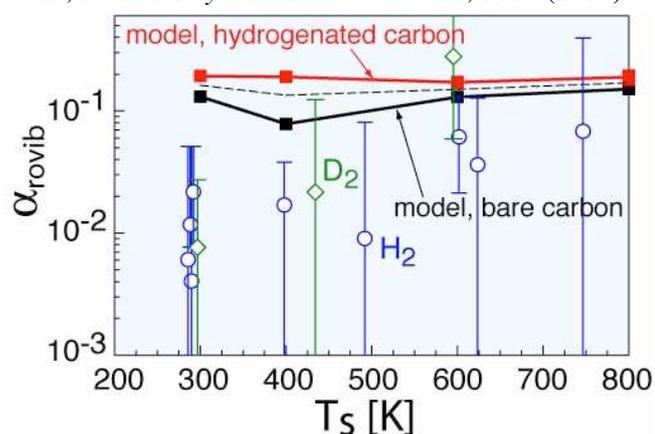[11] E. Ya. Kogan and V. N. Malnev, *Sov. Phys. JETP* **47**, 276 (1978).

Fig. 3. Measured (open symbols; blue for H_2 and green for D_2) and modeled (solid points) rovibrational energy accommodation coefficients α_{rovib}. Red points are for hydrogenated carbon surface; black points for bare carbon surface.