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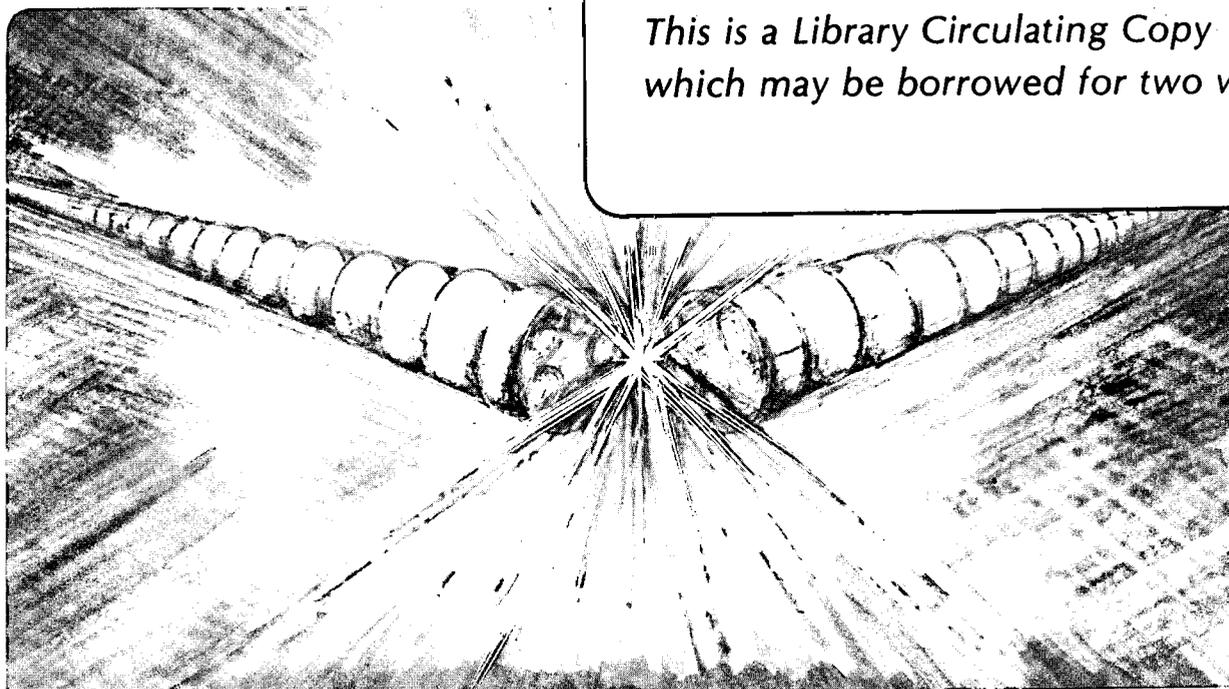
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***IN-SITU* DENSITY AND TEMPERATURE MEASUREMENTS OF
VIBRATIONALLY-EXCITED HYDROGEN MOLECULES IN ION SOURCE
PLASMAS***

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Abstract

The role of highly vibrationally-excited hydrogen molecules has been postulated to be of great importance in H⁻ ion sources. However the difficulty of making *in-situ* measurements has led to a paucity of direct determinations of these species within the plasma of these sources. Recently, vacuum-ultraviolet (VUV) laser absorption spectroscopy has been used to measure the H₂ rovibrational populations up to v''=5 and J''=8 in a medium-power hydrogen plasma. This work extends those measurements to v''=8 and to J''=13. The populations of the vibrational levels still appear to be almost Boltzmann. The theoretically predicted plateau is not observed up to the detection limit. The dependence of several vibrational levels on discharge current and filling pressure is shown.

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I. INTRODUCTION

High current H^- (D^-) ion sources are of interest to the fusion community for neutral beam purposes, as H^- but not H^+ can be efficiently neutralized at beam energies above 100 keV/amu.¹ H^- has been observed in hydrogen plasmas in large concentrations (1 to 20% of the electron density).² The proposed formation mechanism is a two-step process which involves dissociative attachment of electrons to highly vibrationally-excited H_2 molecules ($v'' \geq 5$).^{2,3} The distribution of the H_2 molecules within the many (15) vibrational levels of the electronic ground state, $X^1\Sigma_g^+$, has been the subject of many theoretical and several experimental investigations.⁴⁻¹⁰ Many different collision phenomena, both with other particles and the walls of the device, must be taken into account in order to accurately model the vibrational populations. Most of the theoretical works predict that a plateau occurs within the vibrational distribution, usually from $v'' \approx 5$ to $v'' \approx 11$. The vibrational distribution has been previously measured using VUV absorption spectroscopy up to $v'' \approx 5$. No evidence for a plateau was found, within the uncertainties of the measurement.⁷ This work extends those measurements to $v'' \approx 8$. The dependence of the population of several vibrational levels on discharge current and filling pressure is also presented.

II. EXPERIMENTAL

The method used to determine the quantum-state specific populations, VUV laser absorption spectroscopy, has been described previously in detail.¹¹ Briefly, narrow-band tunable radiation in the range of 80,000 to 86,000 cm^{-1} is produced by four-wave sum-frequency mixing of two pulsed dye laser beams in a mercury vapor

oven. The resulting coherent VUV beam is split into two beams using a diffraction grating. One beam passes through the plasma in the discharge chamber⁷ and is then detected with a microchannel plate (MCP) detector operating in the linear gain regime. The other beam is detected directly using another MCP detector. The ratio of the two signals is measured with and without the discharge, from which the transmission of the plasma can be directly deduced as a function of frequency. This is done in the neighborhood of numerous strong transitions which have $H_2(v'', J'')$ states of interest as their initial state. The integrated path density, hereby line density, and the translational temperature can be determined in a direct manner from the transmission data, if the oscillator strengths of the relevant transitions are known. The transitions used were $X\ ^1\Sigma_g^+ \rightarrow B\ ^1\Sigma_u^+$, $C\ ^1\Pi_u$, which have theoretically determined oscillator strengths which are stated to be accurate to 6%.¹² The bandwidth of the VUV radiation is narrow ($\sim 0.30\text{ cm}^{-1}$) compared to the Doppler linewidths of the H_2 absorption lines employed, so that corrections to the inferred line density(s) are unnecessary. Small corrections are necessary for the temperature determination however. The actual density of a given rovibrational state, $n(v'', J'')$, is computed by dividing the measured line density by the path length, which is well defined by differential pumping apertures.

III. RESULTS

A plot of the population of the $J''=1$ state of each vibrational level for $v''=1$ to 8 is shown in Fig. 1. The discharge parameters are 25 A at 120 V, with 8 mTorr of filling pressure. These were chosen to be typical of those used in H^- sources, within the limits of our power supplies.¹⁴ The error bars on most of the points are $\sim 15\%$, but are $\sim 25\%$ for $v''=7$ and 8. The vibrational distribution is well described by a temperature of 0.36 eV ($R^2 = 0.997$), in contrast to several theoretical models which predict a plateau in the distribution.^{5,6} The $J''=1$ population for $v''=7$ was estimated based on the

population for $J''=3$ and the corresponding ratio for $v''=6$, as coincidences obscured the Q and R $J''=1$ lines. Comparing only the $J''=1$ population of the different vibrational levels is valid because the rotational distributions in these discharges are generally independent of v'' , and are apparently insensitive to most macroscopic parameters, as has been shown previously by the authors and others.^{7,8,10} A graph of the measured rotational distribution for $v''=1$ is shown in Fig 2. The rotational temperature derived from the first four rotational states is 0.036 eV, which is close to previous measurements for similar discharges. The non-Boltzmann tail of the distribution is typical for these discharges.

The scaling of the population of $J''=1$ and 3 states of the $v''=4$ and 6 manifolds with discharge current is shown in Fig. 3. The population of both of the $J''=3$ states has been multiplied by 0.5 for the plot to avoid overlapping. The $v''=6$ data are clearly noisier, but the uncertainty after averaging is no larger than ~20%. The most salient point is that the population of $v''=4$ is almost independent of discharge current over the range studied and the peak is apparently at ~3A. Although it was not possible to get the low current data for $v''=6$, it is clear that the population thereof is at most constant with increasing discharge current. The data would indicate that these populations have a dominant destruction mechanism that scales roughly linearly with discharge current, such as electron density or atom density. The slight negative slope is probably mostly accounted for by depletion of the total H_2 density due to: a) dissociation, and b) increased pumping speed of the differential pumping holes caused by increased gas temperature at higher discharge currents.

The dependence of the density of the $H_2(v''=5, J''=1)$ state on filling pressure is shown in Fig. 4, for discharge parameters of 8 A at 120 V. The discharge current was chosen because it maximizes $n(5,1)$ at 8 mTorr. There is a broad maximum which peaks at roughly 12 mTorr. It is interesting to note that the optimum filling pressure for extracted H^+ is approximately the same for a similar discharge.¹⁴ The dependence at

lower pressures was not obtained because the discharge was unstable at these pressures.

The translational temperature of the various $H_2(v'',J'')$ states was also obtained. These temperatures are approximately the same as those found previously by the authors.⁷ The temperature measurements have somewhat more scatter than the density measurements. The translational temperature of each rotational state of $v''=1$ was found to increase monotonically with J'' , from 0.041 eV for $J''=0$ to 0.085 eV for $J''=13$. However these increases are not much larger than the uncertainty of the measurement (~10%). There was no difference within the errors bars of the $J''=1$ and $J''=3$ translational temperatures for the higher v'' states.

IV. DISCUSSION

The difference between the results of this experiment and those of the only other experiment which reach the so-called plateau region are much larger than the uncertainties of the measurement, therefore discussion of the differences is warranted. The other experiment to measure up to $v''=5$ during a discharge, performed by Eenshuistra, et al., found results which qualitatively agreed with theory.¹⁰ That experiment was performed in an ion source which has parameters similar to the one used presently, but the measurements were performed using the technique of resonantly enhanced multiphoton ionization on the gas effusing from the source. Tungsten filaments rather than LaB_6 were used in that experiment. The plateau was found to begin roughly at $v''=3$ in that work, and to be strongly dependent on discharge current. The dependence of the $v''=5$ population was almost linear with discharge current, in contrast to the results here for $v''=4$ and 6. In both experiments the population of the lower v'' states decreased mildly with increasing discharge

current. The pressure dependence of $n(5,1)$ was also similar. It is not known if the difference in techniques is responsible for the disagreements found. It is possible that the effusing gas may have a different vibrational distribution than the gas inside the 'hot' zone when the plasma is confined. If the mean free path for all $H_2(v'')$ is long compared to the chamber scale length, then gradients should be small and the two techniques should give similar results. Conversely, if the mean free path for some $H_2(v'')$ are considerably shorter than a scale length, then a disagreement between the two would occur. The scaling behavior of $n(v''=5)$ with discharge current in Ref. 10 is consistent with the dominant loss term being collisions with the wall or $H_2(v''=0)$, whereas in the present work it can be explained by collisions with plasma-produced particles in the volume. If the confinement of charged particles is much better in the ion source used in this experiment, the differences are what is expected qualitatively. The measured thermal electron density in the source used in this work is $\sim 1.6 \times 10^{12} \text{ cm}^{-3}$ compared to $4 \times 10^{11} \text{ cm}^{-3}$ for Ref. 10.

The present results are also in disagreement with the qualitative results of several models, all of which predict a plateau should occur in the vibrational distribution.^{5,6} Specifically, the ratio of $n(v''=4)$ to $n(v''=8)$ is predicted to be in the range of 2-10, but we find a factor of 47. Hiskes and Karo published theoretical results which were done specifically for comparison to the earlier results ($v'' \leq 5$) of the authors.¹⁵ The divergence between the two is increasing rapidly with increasing v'' at $v''=5$, the limit of their projections. It should be noted that the electron density stated above is a factor of two higher than the two different values used by Hiskes and Karo in their calculations, however, their results were fairly insensitive to this parameter for $v'' \geq 4$. Further work is in progress on a similar source with a weak magnetic filter and tungsten filaments to try to clarify this discrepancy. The atom population, vibrationally-excited populations and extracted H^- current will be measured under a range of discharge conditions.

V. CONCLUSIONS

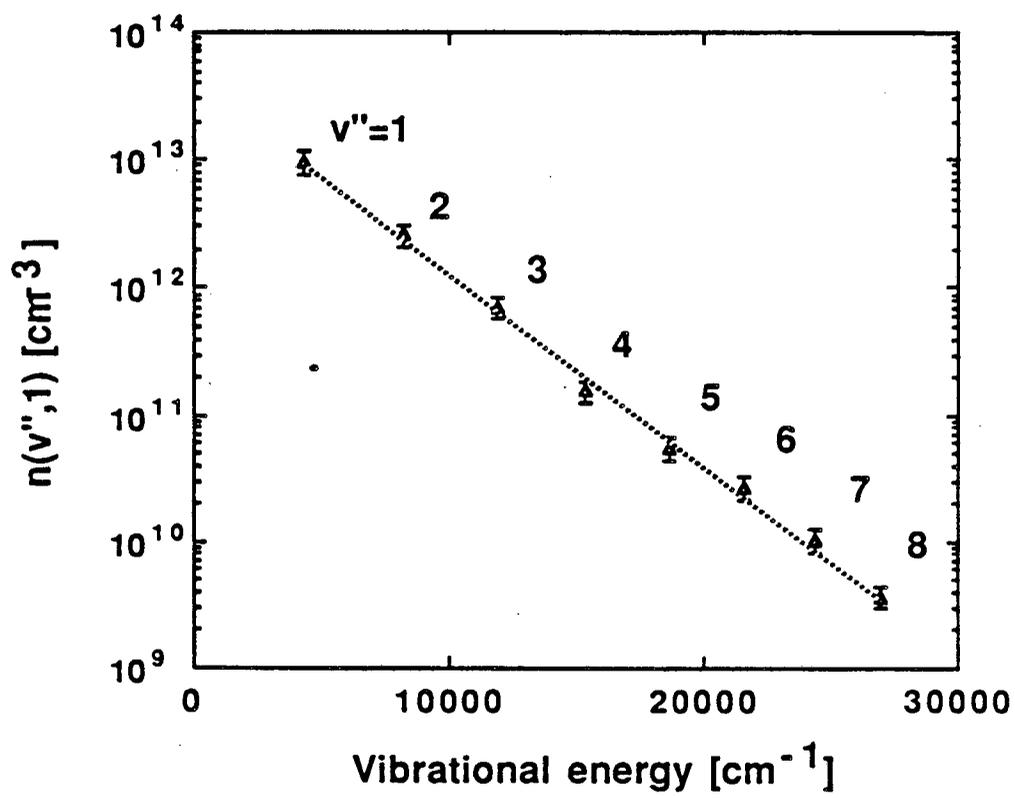
The vibrational distribution has been measured in a medium-power discharge for $v''=1$ to 8. The experimental results are well described by a temperature of 0.36 eV. This is in disagreement with theoretical results which predict a plateau from $v''=5$ to $v''=11$. This is also in disagreement with a recent experiment on a similar discharge which found results which agree qualitatively with theory. The dependence on discharge current of the population of the vibrationally-excited molecules indicates that the mean free path for destruction is short compared to the chamber scale length, ~ 20 cm. The dependence of $n(5,1)$ on filling pressure is almost flat in the range measured.

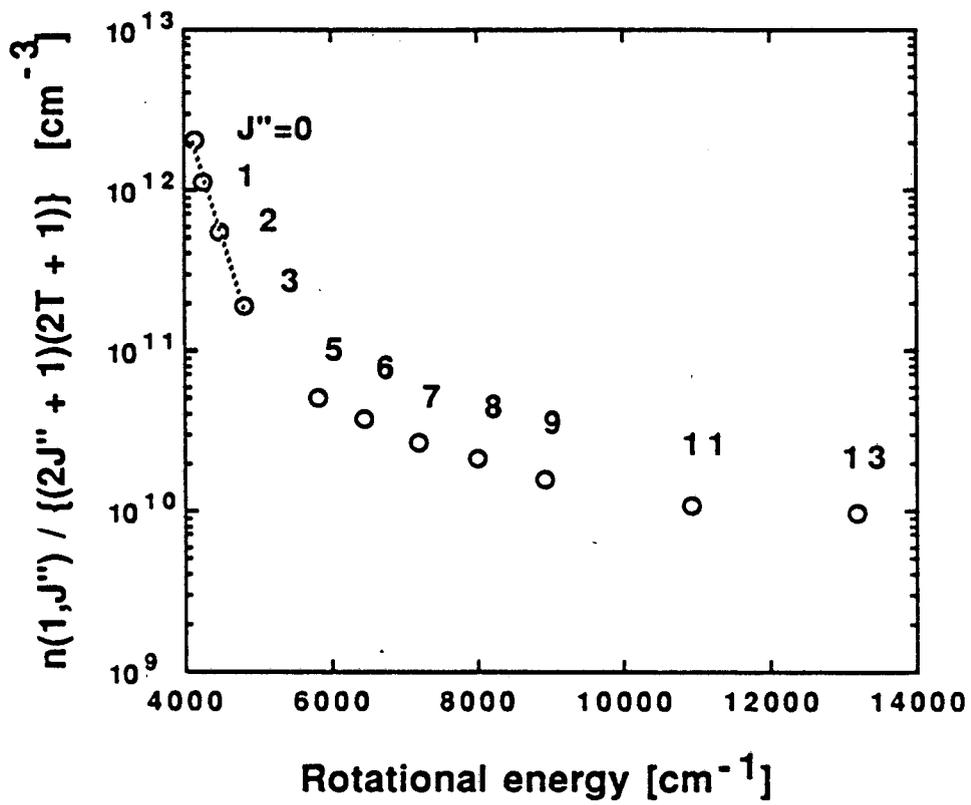
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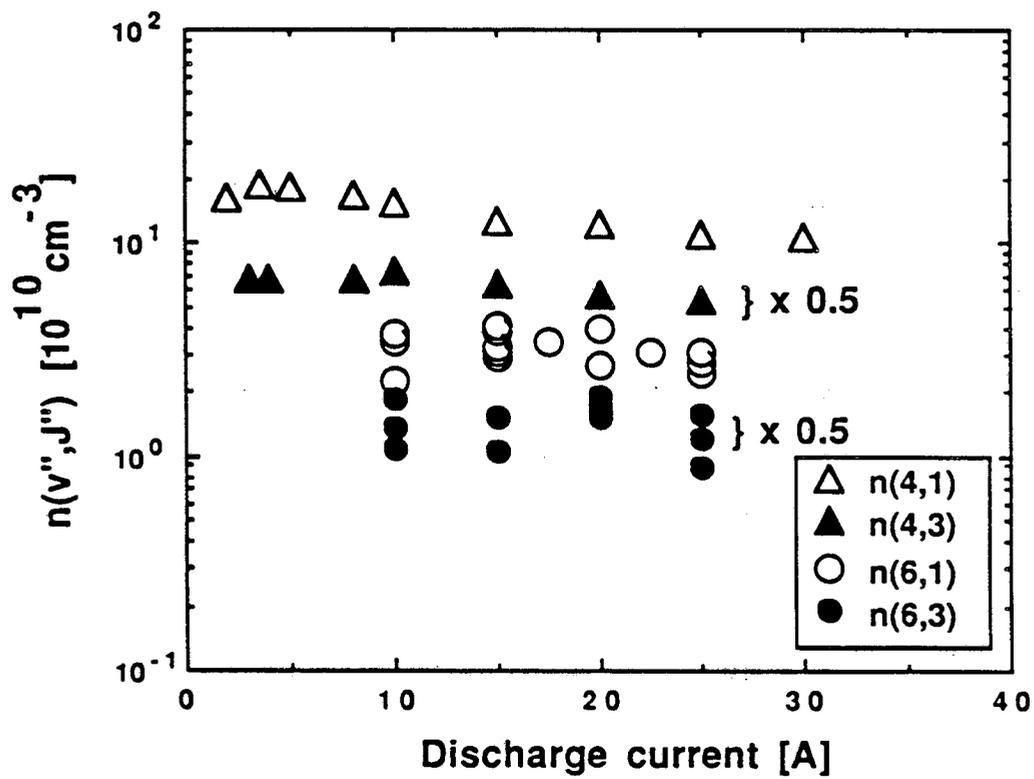
- Fig. 1 Population of $J''=1$ state of each vibrational manifold for $v''=1$ to 8. The dotted line is a least-squares fit and corresponds to a temperature of 0.35 eV. Discharge parameters are 25 A at 120 V with 8 mTorr of filling pressure. The $v''=7$ point was estimated based on $J''=3$ of $v''=7$ (see text).
- Fig. 2 Population of each of the rotational substates of $v''=1$. T is nuclear spin. The distribution is clearly non-Boltzmann, but the first four states have a quasi-rotational temperature of 0.036 eV. Discharge parameters are same as in Fig. 1.
- Fig. 3 Dependence of the population of four rovibrational levels on discharge current. Note that the $J=3$ populations are actually twice those shown on the graph, so that overlap could be avoided. Other discharge parameters are 120 V at 8 mTorr filling pressure.
- Fig. 4 Dependence of the $H_2(5,1)$ population on filling pressure. Discharge parameters are 8 A at 120 V.

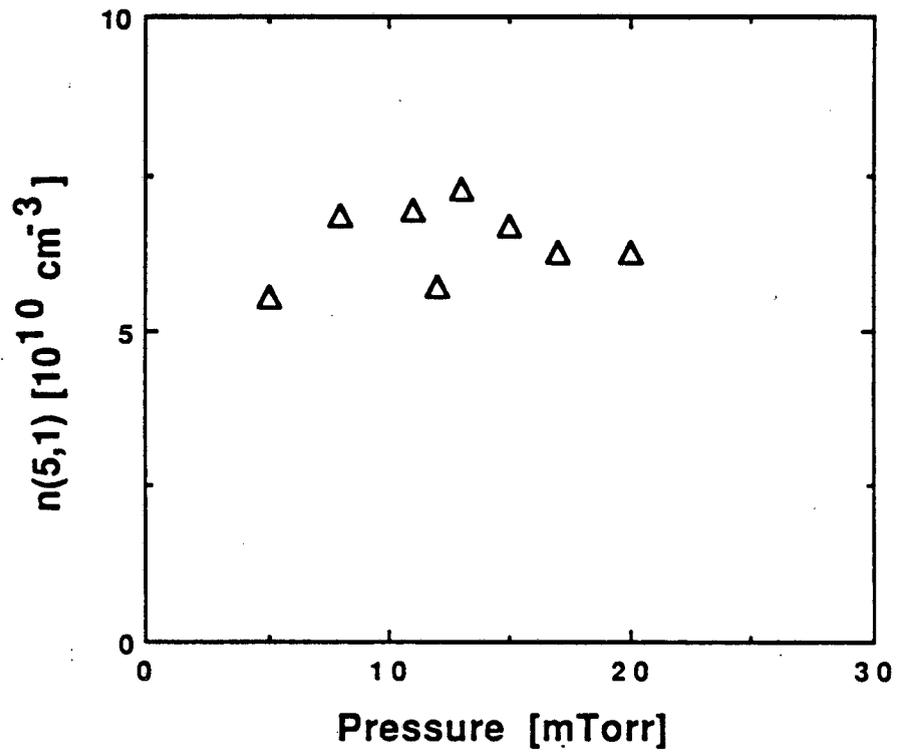
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