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APPENDIX I

Comments on H^- Volume Production in Cs-Seeded Ion Sources

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COMMENTS ON H^- VOLUME PRODUCTION IN CS-SEEDED ION SOURCES

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Considerable interest was generated at the IAEA Negative Ion Beam Workshop in Grenoble, France, in March, 1985, by a report from the Kurchatov Institute on the development of a 2-ampere steady-state H^- ion source, in which the ions were volume-produced in a discharge in H_2, seeded with Cs vapor.[1] The mechanism primarily responsible for this remarkably high current from a volume production source was not yet understood, but it was tentatively presumed to involve the collisional energy transfer from electronically excited Cs 6p atoms into H_2 vibrations. In any case, it was apparently different from the surface-plasma interactions that have been assumed to control the H^- production in the Dudnikov-Dimov type sources.

BACKGROUND

Following the experiments of Allan and Wong, and theoretical developments by Wadehra and Bardsley,[2,3] it has become commonly accepted that vibrationally excited H_2 plays a critical role in the efficient generation of H^- in a hydrogen plasma as first observed by Bacal et al. The "volume production" sources now being developed at LBL and elsewhere based on pure H_2 plasmas have shown promise for high-brightness in tests at LANL, nevertheless the reported high-current results from the Kurchatov source stimulated a consideration of the mechanisms involved in it.

The theoretical calculations[2,3] of dissociative attachment in H_2,

$$e^- + H_2(v) \rightarrow H^- + H,$$

show that the maximum cross section increases by 10^5 between $v = 0$ and $v = 6$, with 10^4 of this occuring by $v = 4$. Because the maximum cross section for attachment via excitation of the $H_2^-\ {}^2\Sigma_u^+$ resonance occurs near the threshold energy,[2,3] which decreases as v increases, the attachment rates will increase even more dramatically for high v's if the bulk of the electrons have kinetic temperatures under a few eV.

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EXCITATION OF H_2 VIBRATIONS BY EXCITED CESIUM ENERGY TRANSFER

Because dissociative attachment with H_2 is the primary H^- production mechanism in H_2 plasmas, we first consider the possibility of increasing the vibrational temperature of the gas to improve the efficiency of the ion source, and in particular for a method of reaching $v = 4$ or higher. Collisional energy transfer from the 6p ^2P resonant state of Cs, which could be "resonantly trapped" in the volume is the most likely mechanism. There have been no measurements of energy transfer from this reaction, but the similar reaction between Na $3p^2P$ and H_2 has undergone fairly extensive study. Reiland, Tittes and Hertel[4] found that Na 3p transfers a about 1 eV (max. probability) of the total 2.2 eV available, to either H_2 or D_2, yielding vibrational distributions of: 60%, 13%, 61%, and 26% and 0%, for $v = 0 \rightarrow 4$, respectively. Cs 6p has about 1.6 eV available, and using a similar relative distribution for E-V transfer in H_2 to that of Na, one can conclude that the distribution from a single collision would peak at $v = 1$.

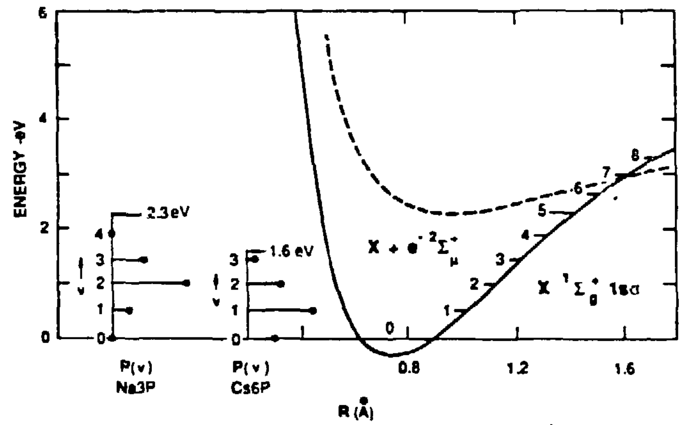

Figure 1 Potential energy diagram of the $H_2(X \, \Sigma_g^+)$ ground state and the location (Ref. 2) of the broad $H_2^- \, \Sigma_\mu^+$ resonance that is responsible for dissociative attachment of low-energy electrons. Bar graphs to the left indicate relative excitation of $H_2(v)$ by thermal collisions with Na 3^2P (Ref. 4) and Cs 6^2P (estimated).

Figure 1 shows the distribution in $H_2(v)$ found by Reiland et al.[4] for Na(3p)-H_2 collisions, and, by analogy, one that could be predicted to be excited by Cs(6p) collisions. These distributions shown at the lower left, indicate the relative excitation of the vibrational levels of the H_2 $X^1\Sigma$ ground state, whose energies are shown on the potential energy diagram. Arrows above the distributions at 2.1 eV and 1.6 eV indicate the energies of the Na and Cs excited states, the maximum that can be transferred in a single collision.

Although the reaction rates (for Na) are reasonably high ($>10^{-10}$ cm^3s^{-1}), it is obvious that single collisions with Cs(6p) cannot excite much above v = 1. Excitation of higher v by multiple collisions with excited Cs is quite unlikely, thus it appears that excitation of $H_2(v \geq 5)$ by cascade radiation induced by the high energy electrons, examined by Hiskes,[5] would be faster than by excited Cs.

Other reactions that could involve excited Cs were considered, but none look promising. However, there are two beneficial effects that ground state Cs could have.

POSSIBLE ENHANCEMENT OF H^- VOLUME PRODUCTION DUE TO GROUND-STATE Cs

1. Electron Cooling.

The introduction of Cs into the volume would have a cooling effect on the electron temperature, due to excitation and ionizing collisions. This effect would

(a) increase the dissociative attachment rates, as well as
(b) reduce the H^- loss by electron collisional detachment.

The magnitude of the effect would depend on the amount of the Cs present and cannot be estimated, but the ionization and excitation cross sections of Cs by electron impact are all large, and because there are no comparable inelastic cross sections in H_2 below about 10 eV, the presence of Cs could be important in cooling the electrons. Other aspects have been considered by Ehlers and Leung.[6]

2. Production of $H_2(v)$ by H_3^+ + Cs Charge Transfer.

We have found[7] that about two-thirds of these reactions,

$$H_3^+ + Cs \rightarrow H_3^* \rightarrow H_2^\ddagger + H + Cs^+,$$

between Cs and the main positive ion, H_3^+, lead directly to 3.6 ± 1.5 eV internal (rovibrational) excitation of H_2, which is equivalent to v = 9 for pure vibration. The remaining 1/3 of the reactions yield H + H + H products.[7] The total cross sections[8] are very large, ≥ 150Å^2 and independent of energy from 1000 eV down to 50 eV, the lowest energy examined (See Figure 2), thus they are

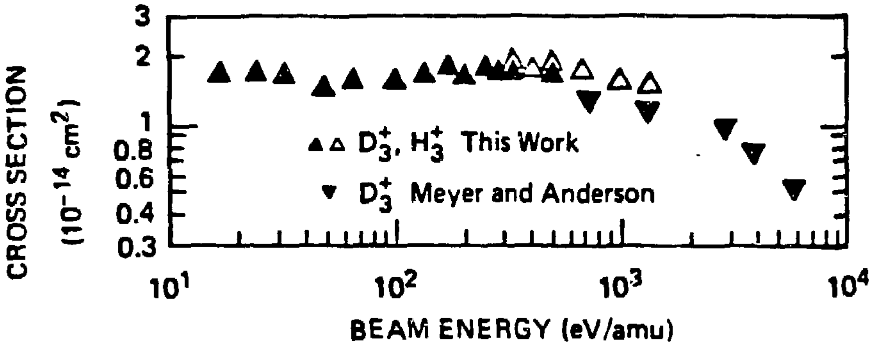

Figure 2 Cross sections for electron capture of H_3^+ in Cs (from Ref. 8).

still large near thermal energies. And because the reaction goes to near-resonant Rydberg states of H_3, (which subsequently predissociate), the final states should be nearly independent of collision energy.

Of course, substantial rotational excitation can also be expected, however Wadehra[9] has shown that combined rovibrational energy is more effective than pure vibration in enhancing dissociative attachment in H_2. In fact, his rates for electron temperatures of 1 eV reach a maximum of about 10^{-8} cm^3/sec for a total internal energy of about 3.8 eV. This reaction with ground-state Cs is fast and effective, and does not require Cs excitation. It thus is, with little doubt, more important than E-V excitation of $H_2(v)$ by Cs 6p. Whether it is as important as vibrational excitation by electron impact is not known.

It is impossible to determine its actual effect without knowing the actual conditions and modeling the plasma. However it can at least be noted that if $n_e \sim 10^{12}$ cm^{-3}, which is reasonable for a negative ion source, and if the dissociative attachment rate coefficient is 10^{-8} cm^3/s (the calculated value for 3.8 eV of rovibrational energy), then an H_2^* will undergo dissociative attachment in 10^{-4} s. Our results[6] also show that about 5 eV is released as kinetic energy in the H_2^* + H products, of which about 1.5 ev would go to the H_2^*. These would have a speed of about 10^6 cm/s, and would leave the ~ 10 cm of the plasma volume in 10^{-5} s, and would thus have about 10% chance of forming H^-. This is a substantial probability, and shows that the mechanism is worth considering.

Finally it should be noted that H_3^+ and D_2^+ ions are vibrationally excited when formed from $H_2^+ + H_2$ and $D_2^+ + D_2$ reactions at thermal energies. From the analysis of the limited energies of the D + D + D products, we concluded that the D_3^+ ions in our beam had internal energies of about 2.2 eV on the average, with half maxima at about 0.5 eV and 3.2 eV. This finding within uncertainties, is consistent with an analysis by Smith and Futrell,[10] who concluded that the D_3^+ is expected to have the excitation energies that at least 69% of of the D_3^+ ions are formed with 0.6 eV or more. The conditions in these experiments are typical of those that exist in any ion source operating at pressures where two-body collisions dominate reactions. The internal excitation in H_3^+ and D_3^+ will also affect the rates and final product states of $e + H_3^+$ dissociative recombination, another source of $H_2(v)$.

SUMMARY

It does not appear likely that energy transfer from excited Cs atoms can excite H_2 vibrationally enough to be responsible for the high efficiency of the ion source. On the other hand, charge transfer with $H_3{}^+$ the neutral Cs is efficient, and leaves the $H_2{}^+$ products highly excited. Cursory considerations indicate that these can have a substantial probability of forming H^- before leaving the plasma volume, and thus the reaction may aid volume production. The presence of Cs in the volume could also have an important cooling effect on the electrons, which would increase the dissociative attachment rate on $H_2(v)$ and also reduce H^- loss by electron collisional detachment.

ACKNOWLEDGEMENTS

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