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METASTABLE NEGATIVE IONS AND ION PAIR FORMATION

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Final Report

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Experimental work was directed primarily toward determining properties (energies, lifetimes, autodetachment and photodetachment) of metastable autodetaching negative ions, including He_2^-, Ca^-, He^-, and vibrationally detaching OH^-. Also explored were the dissociative decay mechanisms and pathways of the lower Rydberg states of HeH, NeH, NeD, H_3, D_3, H_2D, HD_2, OH, and O_2, using two translational spectroscopic methods. In addition, work was devoted to understanding the mechanisms that control the behavior of Cs-seeded H^- ion sources developed for high energy neutral beam production.

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INTRODUCTION AND OBJECTIVES

The main objective of this research was to determine properties of metastable autodetaching negative ions (their energies, photodetachment properties, decay rates, and mechanisms of autodetachment). Most of these ions are electronically excited above the ground state of the neutral species, but they are stabilized against rapid autodetachment by their spin configurations. They offer a unique approach to studying the behavior of highly correlated excited electronic configurations, and they also have potential applications for forming high energy neutral beams, but little experimental research had previously been directed toward them. Secondarily, the research was concerned with the properties and decay mechanisms of Rydberg states in molecular ions. These mechanisms are related to those that occur in dissociative recombination of electrons with molecular ions, a process of great importance because it is the major electron loss mechanism in planetary ionospheres and low density molecular plasmas. In addition, we devoted some effort toward an ongoing objective of clarifying the reactions important in ion sources that produce H^- ions for fast beams. One task we envisioned in the initial proposal has not yet been accomplished, namely, an exploration of long-range ion-pair production at near-thermal energies.

This work was exciting and productive. As often happens in basic research, we encountered unexpected phenomena that led to additional important results and caused some digression from the initial work plan.

ACCOMPLISHMENTS

The research accomplishments under this contract are summarized below. More complete descriptions of some of them can be obtained from past reports and from the publications cited.

OBSERVATION OF METASTABLE AUTODETACHING STATES IN OH^-

Slow autodetachment was observed from OH^- that had been produced from a beam of H_2O^+ traversing a Cs vapor oven. This observation was surprising because OH^- is a stable negative ion. Measurements of the autodetached electron energy spectrum ruled out the autodetachment from excited electronic states, so the possibility of slow vibrational autodetachment was considered. From measurements of the kinetic energy released, we deduced that the OH fragments from dissociative electron capture by H_2O^+ in Cs are internally excited by about 3.5 eV, clearly enough to overcome the electron affinity. The process requires a conversion of vibrational energy to electronic energy, a breakdown of the Born-Oppenheimer approximation. Using a theory first applied to vibrational autoionization of N_2 Rydberg states, we calculated decay rates that were abnormally slow for OH and in agreement with those measured in our experiments. The results were published in *Chemical Physics Letters.*[1]

AUTODETACHMENT He_2^-

We first discovered this unexpected negative ion during our previous contract with AFOSR. We had measured its autodetachment rate and found it to vary with the time delay between formation and observation, indicating more than one autodetaching state. We had made preliminary measurements on the autodetached electron energy spectrum, which we found in disagreement with predictions based on existing knowledge of the He_2 and He_2^- potentials. During the current contract, we set out to explore more details of the autodetachment. The more we examined this process, the more complicated the implications of the results became. We first remeasured the autodetached energy spectrum, confirming and improving our earlier results and confirming the disagreement with predictions based on the existing potentials. We then made an analysis of some earlier published observations on the He_2 Hopfield continuum, which confirmed the potential of the He_2 ground state, thus placing suspicion of the He_2^+ potentials calculated by Michels.[2] That analysis, which was done in collaboration with Michels, appeared in *Chemical Physics Letters*.[3] After fixing the ground state potential. we found that a radial shift of Michels's potential by -0.06 Å, coupled with contributions from He_2^+ vibrational levels $v \leq 3$, gave a good fit for the calculated spectrum. Finally, calculations including the effects of rotational heating, by Hobbs and Michels, showed that a very satisfactory theoretical fit to the data could be achieved using the unshifted potential of Michels, assuming a rotational Boltzmann temperature $T_{rot} = 15000$ K, and still including contributions from $v \leq 2$. Both latter conditions pose interesting questions that we hope will be answered by future work. They are discussed in a paper written in collaboration with Michels and Hobbs and published in *Physical Review A*.[4]

BOUND EXCITED STATES OF HeH

This "excimer" molecule, which is bound only in excited Rydberg states, was first predicted theoretically by Michels and Harris in 1963 was not observed until two optical observations were made in Germany in 1985. We observed that it decayed by both predissociation of the lower $A^2\Sigma^+$ state and dissociation by radiative decay from the lower $B^2\Pi$ state to the repulsive ground state. The excimer states were produced from HeH^+ beams by near-resonant electron capture in Cs. We used a technique developed in our laboratory earlier,[5] applying translational spectroscopy on H^- ions produced at 0° scattering angle following the HeH dissociation while still in the Cs oven. The results were published in *Physical Review A*.[6]

A visit to the FOM Laboratory for Atomic and Molecular Physics in Amsterdam permitted a subsequent collaborative study using a high-resolution position- and time-sensitive detector. The results showed a strong isotope effect between ^4HeH and ^3HeD on the competition between radiative and predissociative decay of the next higher $C^2\Sigma^+$ state. A further collaboration with theorists from the University of Paris at Orsay ensued, and a paper was published in *Chemical Physics Letters*.[7]

BOUND STATES OF NeH AND NeD

An experimental study of the dissociative decay of these isotopic excimers was undertaken similar to that on HeH. This species is of interest because of its possible use in uv lasers. Our particular interest was stimulated by a recent proposal by Selgren et al.[8] that a metastable bound state might exist in the ground state potential, which is not expected on theoretical grounds We found an even stronger isotopic effect than in HeH/HeD and an even more surprising result from our initial experiments: they were in essential agreement with the much less definitive observations of Selgren et al.[8] While NeH was observed to undergo both radiative and predissociative decay, similarly to HeH, the predissociation of the A state of NeD is much slower than its radiation, and almost no predissociation is observed. However, this isotopic effect is straightforward but extreme and leads to information on the predissociative coupling strengths. The confounding results were that the radiative decay of the A state of NeD appeared to be vastly different from that of the B state, leading to the anomalous angular scattering patterns observed by Selgren et al.[8] To understand these results, we performed a number of calculations of the radiative spectrum from all states, including the use of the theoretically predicted strongly R-dependent dipole matrix elements. The results agreed with our observations for NeH but not for NeD. We now believe that our observations included contributions from a D_3O^+ (mass 22) contaminant in the NeD^+ (mass 22) beam, which would have been indistinguishable in the mass spectrum but whose counterpart H_3O^+ (mass 19) would not have contaminated the NeH^+ (mass 21). Recent calculations in our laboratory by Talbi and Saxon[9] have predicted a low-energy well in the ground-state potential of H_3O^+, which would explain our observations. We will examine this possibility shortly (we are using a different experimental configuration for photodetachment experiments at present). If correct, our hypothesis would explain the results of Selgren et al.,[8] and our results would confirm the predictions of Talbi and Saxon[9] and also test the accuracy of their calculated potential.

$^{1,3}\Pi_g$ AND $^{3,5}\Pi_u$ RYDBERG STATES OF O_2.

We had earlier (under DOE support) studied the final states of electron capture by H_2^+, H_3^+, and O_2^+ in Cs when developing the translational spectroscopy technique used in the OH and HeH studies mentioned above.[5] It was of interest to use the high resolution apparatus at the FOM Laboratory in Amsterdam during a 10-week visit in 1986 and to reexamine the work on O_2, which had shown some interesting effects due to avoided crossings between the dissociating final states.[5] We undertook a short experiment, whose results were astounding in their detail. The experiment determines the barycentric kinetic energies released (KER) to the atomic products following dissociation, just as our own method does, but it achieves much greater resolution. The KER spectrum was almost completely structured--direct evidence that electron capture of O_2^+ in Cs leads directly to Rydberg states, which subsequently are predissociated by perturbing valence states. A wealth of information was obtained from the lengthy analysis of the data (including those from secondary measurements) revealing the energies and molecular constants of the Rydberg states, the mechanisms and coupling strengths of their predissociation (both direct electronic configuration mixing and spin-orbit coupling), the final electronic states of the dissociated product atoms (different mixtures of ^3P and ^1D, and the strengths of the couplings between the valence states that prevailed during the separation of the atoms during the "half-collision." The analysis also revealed previously unobservable vibrational excitation in O_2^+ due to autoionization of the $H^3\Pi_u$ Rydberg state, which is present in all ion sources using ionizing electrons of over 40 eV. This work resulted in three papers, a preliminary one in *Chemical Physics Letters,*[10] followed by two longer articles in *The Journal of Chemical Physics*[11] and *Chemical Physics.*[12] One more article is planned. A brief summary was prepared for presentation at the International Symposium on Dissociative Recombination held at Lake Louise, Alberta, Canada, May 28-31, 1988. A written version, to be published by World Scientific Publishing Company,[13] is included as an Appendix.

ANALYSIS OF H⁻ VOLUME PRODUCTION IN Cs-SEEDED ION SOURCES

An analysis of H^- production mechanisms in a high-yield ion source reported by USSR scientists, first written as an informal note, was prepared for presentation at the 1986 Negative Ion Symposium at Brookhaven.[14] It concluded that energy transfer from excited Cs atoms to excite the vibrational population in H_2, which was advanced by the Soviet scientists as a possible explanation for their high H^- yields, was not likely to be effective. Instead, it suggested that the most likely important effects due to Cs were a decreased electron temperature and an increased H_2 ($v > 4$) population resulting from dissociative electron capture by H_3^+ ions from Cs (H_3^+ dominates the ions in the negative ion source discharges). In earlier translational spectroscopy experiments in our laboratory, we had deduced that the reaction $H_3^+ + Cs \rightarrow H_2 + H + Cs^+$ yields highly rovibrationally excited H_2. The internal energy distribution was deduced to peak at 3.6 eV, which corresponds to $v = 9$ for pure vibrations.

This small study gained significance because of some recent work by Leung and coworkers at Lawrence Berkeley National Laboratory, who found a factor of 16 increase in the H^- yield when Cs vapor was admitted to the H_2 discharge in their multicusp ion source.[15]

Because of its potential importance, we reexamined this reaction in further experiments as explained in the next section. We obtained similar results, but with the aid of more recent finding regarding the absolute energies of the several electronic states involved in the formation and decay processes, our conclusions regarding the internal energies of the H_2 products of dissociation were slightly different quantitatively. However, the conclusion that a great deal of internal rovibrational energy in the H_2 products results from the H_3^+ + Cs charge transfer is unchanged. In the course of preparing a paper for the forthcoming SPIE meeting (Jan. 12, 1989), after considering all the processes involved in the plasma volume, we concluded that the production of vibrationally hot H_2 results substantially from the production of H atoms in the discharge, which in turn undergo dissociative recombination at the wall, producing vibrationally hot molecules. A paper regarding volume processes has been prepared for the SPIE conference and will be published in the Proceedings.[16]

PREDISSOCIATION PRODUCT DISTRIBUTIONS FROM H_3^* AND ITS ISOTOPES

The conclusions regarding the internal energy in the H_2 products of the H_3^+ + Cs reaction mentioned above were based on our earlier experiments, which were actually performed on the D_3^+ isotopomer.[1] It was of interest and possible importance, therefore, to study the products of H_3^+ predissociation as well, because of the strong isotopic dependences that we had observed between HeH and NeH and their D-containing isomers. We therefore repeated our original measurements on D_3^+ and H_3^+. The two results were almost identical, and both also were in excellent agreement with our earlier measurements, which had been done without the present automation of our energy analysis.

Thus, we found that the first Rydberg states of H_3 and D_3 decay with a branching ratio of about 2:1 between the H_2 + H and H + H + H channels. This result is significant for the H^- ion source problem. The agreement was not unexpected because we had assumed (without solid justification) that predissociation dominated the decay mechanisms. Furthermore, there was no direct competition between H and D in the separate dissociations.

However, in contrast to the H_3 and D_3 cases, in which the isotopic components are symmetric, we might very well expect to find differences between H and D products ejected from H_2D and D_2H. We examined both cases. We first examined D_2H because its mass 5 is singular among other common isotopic mixtures, whereas beams of H_2D are always contaminated with D_2. We found that the H + D_2 channel predominated over D + HD and that the three-atom channel was quite strong. However, the peak of the kinetic energy released in the two-body dissociation was similar to that in H_3 and D_3. We were only able to examine the H-atom products from H_2D (because of the D_2 contamination), but we found that the H + HD channel was competitive with the three-particle channel. These isotopic effects in the branching are very interesting and have already attracted the interest of theoreticians. Kulander and Light[17] examined the H/D ejection probabilities from the photodissociation of H_2D , and they concluded that H ejection is more probable, primarily because the Franck-Condon factors are more favorable. The same factors occur in dissociative recombination and predissociation, but they are augmented by the dynamic coupling required in most cases, which can also favor the ejection of the lighter atom. A paper on this work is in preparation for publication.[18]

Although our technique[5,6] for applying translational spectroscopy to these dissociative electron capture collisions in Cs does not have the high resolution of the current time- and position-sensitive detectors it has the unique capability of examining the products and energy distributions from three-body decay as well as determining the branching between three-body and two-body decay. We plan further experiments using an ion source that produces rovibrationally cool ions.

PRODUCTION AND DECAY OF OH AND OD RYDBERG STATES.

The final states of charge transfer of OH^+ in Cs were examined. We found that the final states are all Rydberg states and that the decay is dominated by predissociation. Evidence of Rydberg states leading to the $X^3\Sigma^-$ ground state of OH^+ as well as the metastable $a^1\Delta$ and $b^1\Sigma^+$ states in the ion beam are all represented; however, we have not completed our analysis of the Rydberg states themselves because of the uncertainties in the present theory. It should be pointed out that the high-resolution apparatus in Amsterdam that was used for the O_2 studies is incapable of handling OH because of the high asymmetry in the dissociation product masses, so we are exploiting our unique capabilities while recognizing their limited resolution.

SEARCH FOR RESONANT STRUCTURE IN He^- PHOTODETACHMENT NEAR THE 3^3S THRESHOLD

We attempted to excite and identify the states that are expected to exist in this energy region, which occurs at photon wavelengths near 4060 Å. Pumping our dye laser with the uv lines of the Kr ion laser, we were successful in producing about 10 mW of usable laser power, but we were unable to detect the neutral He photodetachment products at this wavelength against the large background resulting from autodetachment. To eliminate this background, we designed a shaped solenoidal magnet similar to one used by Lineberger's group to measure low energy electrons near the photodetachment threshold. It traps the low energy electrons generated along the coaxial superposition of the solenoidal $\vec{B}$ field, laser, and ion beams and directs them out of the merged beams and into an electron multiplier. By properly adjusting its magnetic field, we should be able to discriminate against the 19 eV electrons from He^- autodetachment. The detector is now completed, and we plan to resume photodetachment experiments.

PHOTODETACHMENT OF METASTABLE Ca^-

Ca^- has an interesting history. It has been known for some time that Ca formed a negative ion, but it was generally believed, and calculations showed, that alkaline earths could not bind an electron to the ground state but that bound excited ^4P states existed that should have, for some species at least, slow autodetachment rates.[19] Earlier studies simply found a lower limit to the lifetime, but no observations of autodetachment were made. We had made the first observation of autodetaching Ca^- during our previous AFOSR contract, and we observed metastable decay with an apparent lifetime of about 1 ms. In subsequent experiments at Oak Ridge National Laboratory, however, Pegg et al.[20] were unable to detect autodetached electrons, and calculations[21] then showed that Ca^- does exist stably in a ^2P state below the ^1S ground state of Ca. Unpublished calculations of Beck also found that the autodetachment lifetime of the predicted ^4P state should be too short to observe experimentally, in contradiction to our findings. We had not published our results, and so to repeat our experiments to confirm either them or the theoretical predictions of non-metastability. We did so and again found, rigorously, a metastable autodetaching beam of Ca^-. Measurements of decay rates at several background pressures were extrapolated to zero pressure to eliminate the collisional detachment contribution, and they were found to yield an effective lifetime of $2.9 \pm 1.1 \times 10^{-4}$ s. We initiated photodetachment measurements and were successful in finding an increase near the calculated threshold for the 4s4p $^3D + \varepsilon s$ channel. We have not yet completed these measurements, but they should yield an accurate value for the energy of the metastable negative ion state for comparison with the theoretical calculations of Bunge et al.[19]

CONSTRUCTION OF SLOW-ELECTRON DETECTOR AND NEW ELECTRON-IMPACT ION SOURCE

The new detector for low energy electrons was mentioned above. It is now nearly finished and will be tested before the end of July. We are also constructing a new electron-impact ion source to use for most gases. The colutron ion source that we have been using is good for H_2 and He, but the filament has a short lifetime as a result of sputtering in heavy gases and oxidation or chemical reaction in more corrosive gases. We are constructing a Nier-type source with a filament that is external to the main chamber and with the electrons constrained by a magnetic field that guides them through an entrance slit. The design is similar to one already in use on another experiment.

PUBLICATIONS AND CONFERENCE PRESENTATIONS ON WORK SUPPORTED BY THE PRESENT CONTRACT

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PROFESSIONAL PERSONNEL

Professional contributions were made by SRI staff members J. R. Peterson, Y. K. Bae, D. L. Huestis, P. C. Cosby, and recently P. Devynck.

We have been joined by visiting scientists Prof. P. R. Jones (Univ. of Mass., Amherst) and Prof. W. G. Graham (Univ. of Ulster, Coleraine, N. Ireland), on sabbatical leaves, and by D. Hanstorp, a graduate student from Chalmers University, Goteborg, Sweden. These visitors were all supported by their respective universities.

Two scientific collaborations with other institutions occurred. One began with an invitation to J. R. Peterson to visit the FOM Laboratory for Atomic and Molecular Physics in Amsterdam. He spent 10 weeks there in the summer of 1986. That work formed the basis for the Ph.D. research of W. J. van der Zande, which will have led to at least five publications. We also brought Mr. van der Zande to SRI twice on travel grant support, for the completion of journal articles. He has now received his Ph.D. (cum laude) and is a postdoctoral research and Fulbright Fellow in the group of Prof. R. N. Zare at Stanford University.

In addition, an active collaboration was established with H. Michels of United Technologies Research Center that has greatly aided the interpretation of our He_2^- results. Two joint papers resulted from that collaboration. We anticipate further interaction in the follow-on research on excited molecular states and ion-pair production.

PROFESSIONAL INTERACTIONS

Nineteen presentations on work supported by the contract were made at various professional conferences, including four invited papers. Seven departmental colloquia or seminars were given in Germany, France, the Netherlands, Canada, and the United States. In addition, J. R. Peterson attended two Neutral Particle Beams Technical Interchange Meetings at Los Alamos National Laboratory, and he ave one seminar at the Air Force Office of Scientific Research.

In addition the collaborative work with H. Michels (United Technology Research Laboratory) mentioned above, we have maintained an interactive series of technical interchanges with the ion source group (W. Kunkel and K. Leung) at Lawrence Berkeley Laboratory, whose work has stimulated some of our research. We have also interacted with two theoretical groups at Livermore National Laboratory, which are concerned with H^- ion source mechanisms (J. Hiskes) and dissociative recombination (K. Kulander). We have also interacted with personnel at Jet Propulsion Laboratory regarding problems related to negative ion formation (S. Srivastrava and A. Chutjian) and the Rydberg states of O_2 (S. Trajmar). We attended two Neutral Particle Beams Technical Interchange Meetings at Los Alamos National Laboratory and the Fourth International Symposium on the Production and Neutralization of Negative Ions and Beams at Brookhaven National Laboratory.

DISCOVERIES

This work showed conclusively for the first time that electron capture by molecular ions from Cs leads predominantly to near-resonant Rydberg states when they exist (usually the case) rather than near-resonant valence states in the same region. This finding demonstrates the strong role of Franck-Condon factors (in addition to the near-resonance propensity) in determining the final states of electron capture. We also demonstrated the power of translational spectroscopy in determining the decay mechanisms, pathways, and final product states of such Rydberg states. The work on O_2 led to new knowledge of its four 3s Rydberg states and of the accurate locations of the valance states that perturb them.

The work on HeH gave new information on the coupling strengths between the ground state and the 2s and 2p Rydberg states and on the isotopic difference between them. Similar information was obtained for NeH. In addition, that work led by accident to what is apparently the first observation of the low-lying metastable state in H_3O. This finding is a subject of recent AFOSR-sponsored theoretical work,[9] which we will explore further during our follow-on contract.

We also found the first conclusive evidence of the metastable $4s4p^2\ ^4P$ state of Ca^- and measured its lifetime and its energy. The determination of its energy was not completed during the present contract, but the photodetachment work demonstrated the onset of an excited outgoing state, whose value will permit the energy determination to be made during the current contract.

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APPENDIX

MECHANISM FOR H$^-$ PRODUCTION IN CS-SEEDED ION SOURCES

By

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Mechanism for H^- Production in Cs-seeded Ion Sources

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1. ABSTRACT

Measurements of the rovibrational population of H_2 products following dissociative charge transfer of D_3^+ in Cs vapor are reported in the context of the recent observation that the addition of Cs to an ion source discharge can lead to a significant increase in the H^- ions extracted from the source. Consideration is also given to wall and plasma effects. It is concluded that dissociative charge transfer produces a hot $H_2(v)$ distribution, centered around v=6,with all levels populated into the vibrational continuum. It is also found that the discharge is an efficient producer of H atoms. Through wall collisions these H atoms can contribute both directly and indirectly to H^- production in a Cs seeded discharge. Comparison with results from Xe seeded H_2 discharges suggest that in Cs seeded discharges the effectiveness of the magnetic filter may be reduced, the electron density is significantly increased and the plasma potential will be increased. However, with Xe seeding, there is no conclusive evidence for electron cooling of the bulk electrons.

2. INTRODUCTION

The effective enhancement of H^- production by admitting Cs vapor to volume-production ion source, first reported by Antipov et al.,[1] has recently been confirmed by Walther et al.,[2] who found a 16-fold increase from a small multicusp source. Since this type of ion source had already shown promise as a high-brightness H^- source using pure H_2, this development is dramatic. The mechanisms likely to be effective in cesiated volume sources were considered briefly in an earlier report.[3] The *volume* production of H^- ions results primarily from dissociative attachment,[4] $e + H_2 \rightarrow H^- + H$. This reaction is strongly enhanced if the H_2 is rovibrationally excited[5] (a factor of 10^5 from $v = 0$ to 6) and also if the plasma electron temperature is low (< 1 eV). Energy transfer from electronically excited Cs was first considered as a source of H_2 vibrational excitation but was concluded to be unimportant for $v > 2$.[3] Instead, it was suggested that the rovibrationally excited produced in the dissociative charge transfer of H_3^+ with Cs[6] could be efficient.[3] The Cs could also be effective in cooling the electrons.

The earlier report[3] made use of experimental results of Peterson and Bae, who determined the spectrum of c.m. kinetic energies released to the neutral fragments following dissociative charge-transfer of D_3^+ in Cs vapor.[6] Because of the possibility of isotopic effects in the dissociation channels, we have repeated the measurements not only on H_3^+ and D_3^+, but also on H_2D^+ and HD_2^+ We report some of those results here, and reassess the rovibrational populations in the H_2 products that can be deduced from the results. We also give consideration to wall and plasma effects.

2.1 ENERGY DISTRIBUTIONS OF H_3^+ + Cs PRODUCTS

We first give some background information. Electron capture by molecular ions from Cs atoms (I.P. = 3.89 eV) can be nearly resonant (little change in the total internal energy of the system) if the electron is captured into a Rydberg state of the molecule, the lowest of which are generally about 4 eV below the ion (similar to the 2s electron in H). Population of the Rydberg state in these reactions is preferred over (usually repulsive) valence states of the same energy because (a) no

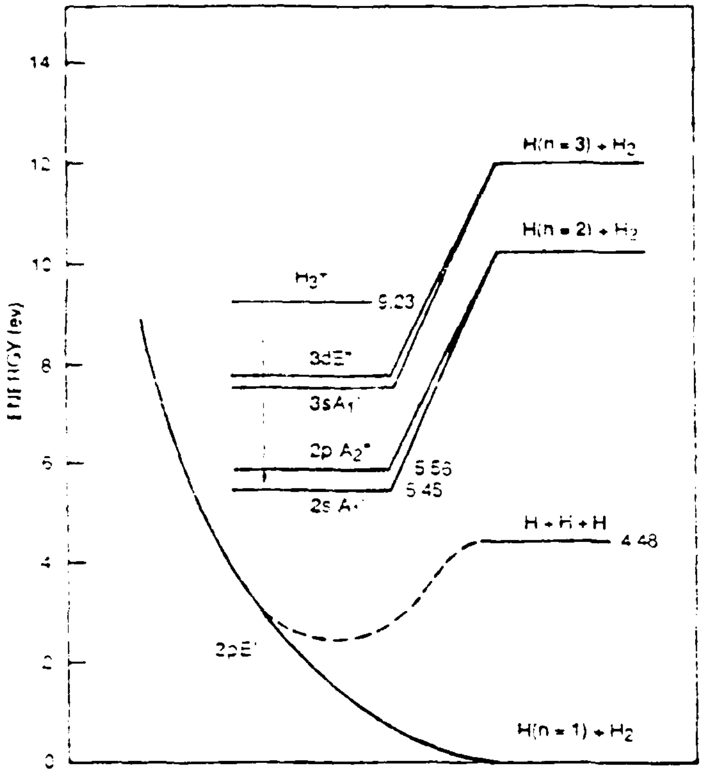

Figure 1. Potential energies of H_3 and H_3^+. Horizontal levels are rovibrationless bound state levels. Abscissa figuratively represents internuclear distance in unspecified geometry. Asymptotic limits are at right.

rearrangement of the valence electrons is required, and (b) the ion-Rydberg Franck-Condon factors are very large (≈1 for $\Delta v = 0$). In the case of H_3^+ these conditions apply to the 2s A_1' and 2p A_2'' states, whose channels are endothermic by 0.11 and 0.22 eV, respectively (see Figure 1). Except for the single metastable rovibrationless state of 2p A_2'', all other levels of both states are predissociated by the repulsive 2p E' state and can yield both H_2 + H and H + H + H products. Our earier study showed that both products result. The total electron capture cross sections are very large (> 150 Å^2),[7] consistent with a long-range capture that leaves the ion core undisturbed. The nearly diagonal ($\Delta v = 0$) Franck-Condon matrix also strongly preserves the vibrational population from the ion to the Rydberg molecule. In ion sources, the H_3^+ ions are formed vibrationally hot by the reaction $H_2^+ + H_2 \rightarrow H_3^+ + H$. This reaction is already 1.83 eV exothermic for H_2^+ (v=0), but in addition, the parent H_2^+ are formed with vibrational energies of about 0.9 eV.[8] Below, we deduce that the H_3 Rydberg molecules produced in our beam were excited by $E_{rovib} \approx 2.1$ eV, and associate this excitation with that of the parent H_3^+ beam.

The technique used to probe the nature of the product states has been described elsewhere.[9] Briefly, we use a fairly simple method to obtain the total center-of-mass (c.m.) kinetic energies released to the dissociation fragments. This quantity, W, is the difference between the total internal energy (electronic plus rovibrational) in the H_3^* before predissociation, and that in final products. We make use of the fact that after the initial two-step reaction

$$H_3^+ + Cs \rightarrow H_3^* + Cs^+, \qquad (1a)$$

$$H_3^* \rightarrow H_2 + H, \text{ or } H + H + H, \qquad (1b)$$

the fast H products (H_3^+ beam energies of several keV) can capture an electron in a subsequent collision to form a negative ion:

$$H + Cs \rightarrow H^- + Cs^+. \qquad (2)$$

We then measure kinetic energies of the H^- ions formed in the direction of the beam, representing 0° and 180° c.m. The data are first corrected to account for the laboratory kinetic energy dependence of the cross section for Reaction (2), and are then transformed[9] to yield relative intensities *vs.* the c.m. kinetic energy release W. The energy scale for this transformation is valid only for the 2-body (H + H_2) case, where the dissociation geometry is known. We consider three geometries for the three-body case. The first is identical to the two-body case with two *unbound* atoms leaving with equal speed in one direction and the third with twice their speed in the opposite direction. The second is the case for one atom at rest (c.m.) and the other two with equal opposing velocities. The W values here are 4/3 the 2-body values. The third is the case for equal angles and speeds; these have W's that are twice the 2-body case. The 2-body and equal angle cases are thus limiting values. Because the equilibrium geometry of H_3 is a symmetric triangle, the equal-angle case should be the most probable for dissociation. To estimate an average W scale for the 3-body products, we place it 75% between the 2-body and equal-angle scales, thus at 1.75 times the 2-body scale.

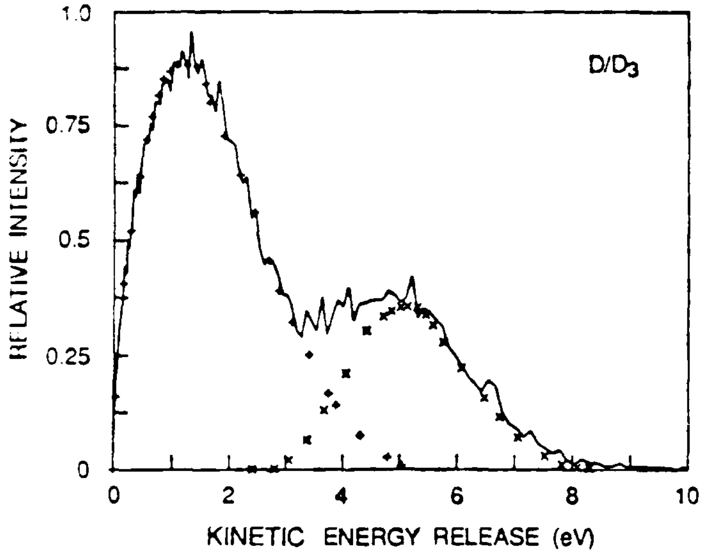

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Figure 2. Transformed data for total c.m. kinetic energies released to D_3 dissociation fragments. The line connects 0° c.m. data from D fragments. Crosses show an approximate fit to the $D + D_2$ fragments at higher energies and the $D + D + D$ distribution at low energies.

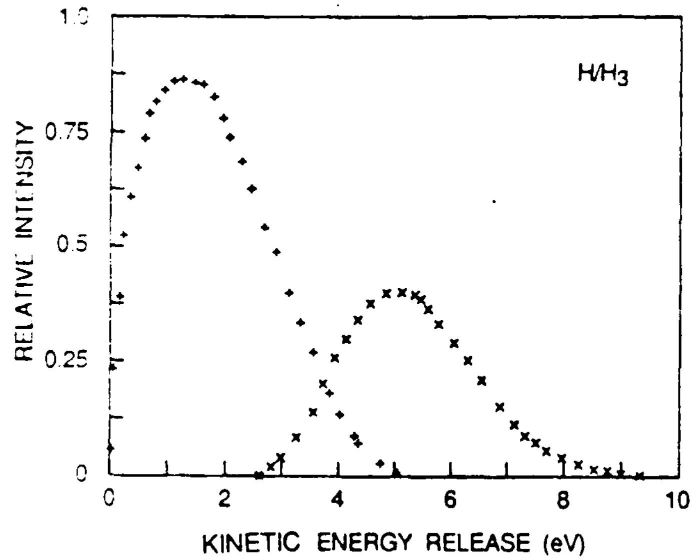

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Figure 3. Kinetic energy release spectrum from H_3 dissociation fragments, analogous to the D_3 apertures in Figure 2.

Figure 2 and 3 show the results for D_3^+ and H_3^+, respectively, with the W values based on the two-body case. The spectra are very similar to each other and also to the one obtained earlier.[5] We find a peak at about 5.05 eV, which we attribute to two-body predissociation, and another lower-energy peak, which we attribute to the 3-body products. This distribution has a long high-energy tail, and has an average value of 3.0 eV on our 3-body W scale.

2.2 ESTIMATION OF THE INTERNAL ENERGY IN THE H_2 PRODUCTS

The rovibrational energy E_{rovib} in the H_2 products is deduced using the relation

$$E_i = W + E_{rovib}, \qquad (2)$$

where E_i is the total energy (electronic plus rovibrational) of the H_3^* products of Reaction (1a), and W is the kinetic energy released in the 2-body channel. The 2s A_1' ($v=J=0$) state is 5.45 eV above the $H_2(v=J=0) + H$ asymptotic limit, and the 2p A_2'' state is at 5.56 eV. Since the 2s state is most nearly resonant, we assume that most of the reactions yield that state. Using the 2p state would yield internal energies 0.11 eV higher.

To assess the amount of rovibrational energy E_{rovib} in H_3^* we note in Figure 1 that the electronic energy of the $2A_1'$ state is 5.45 - 4.48 = 0.97 eV higher than the H + H + H asymptotic limit. The average value of W in the 3-body distribution is 3.0 eV on our estimated scale. We attribute the excess over 0.97 eV results from E_{rovib}, which places E_{rovib} = 2.0 eV in Eqn. (2). Thus the initial total energy in the 2s state is E_i = 5.5 + 2.0 = 7.5 eV.

The $H + H_2$ channel has a W spectrum peaking at 5.0 eV (with 1/2 maxima at 3.7 and 6.5 eV), so we deduce that the H_2 dissociation fragments have an average rovibrational energy of E_{rovib} = 7.5 - 5.0 = 2.5 eV, which is equivalent to v = 6. Thus, the H_2 (v) distribution is very hot, peaking at v = 6, with all levels populated into the vibrational continuum. This is the case for parent H_3^+ ions that are vibrationally excited by an average of 2.0 eV. This result is remarkably similar to the recent results of Cosby and Helm,[10] who photoexcited the rovibrationless 2p $^2A_1'$ to the 3s and 3d Rydberg states about 2 eV higher. These states predissociate to the ground state, and detailed W spectra of the two-body channel showed that the H_2 products are also strongly vibrationally excited, into the (3-body) vibrational continuum. In this case the initial energy E_i was 7.6 eV, similar to ours, but was *entirely* electronic, and was converted to rovibrational energy only as it was dissociated by the repulsive ground state potential. It should be noted that the vibrational excitation in our H_3^+ ions is probably characteristic of most discharge ion sources. We plan to examine the W spectra from a much more vibrationally relaxed ion source.

From the areas under the peaks (after dividing the 3-body area by 3), we find that about 60% of the H_3^+ + Cs electron capture reactions produce rovibrationally excited H_2 for all levels into the continuum, and an H atom, with about 5 eV total kinetic energy, of which the H atom gets about 3.3 eV, and the H_2 receives 1.7 eV. The remaining 40% yield 3 H atoms with an average kinetic energy of about 1 eV per atom.

3. WALL EFFECTS

Although the enhancement effects of introducing Cs into volume-production sources were initially assumed to occur in the gas phase, recent experiments by the LBL group[11] have shown that the introduction of Ba can also strongly enhance the production of H^-. Because the vapor pressure of Ba is negligible compared to that of Cs, it is fairly clear that the enhancing mechanisms are occuring at the surface, whether or not the actual production takes place in the volume. We will examine some effects that can take place at surfaces. Both Cs and Ba form hydrides with bonds of about 1.8 eV, but presumably the surface would bear a mixture of these and the atoms.

3.1 H_3^+ NEUTRALIZATION AT THE WALL

Bulk Cs and Ba have low work functions of 2.1 and 2.7 eV. The work function of metal a surface can be reduced to values below those of bulk materials if Cs or Ba are present on the surface in monolayer or less coverages.[12] It may be expected that the neutralization of H_3^+ as it approaches a Cs covered surface would have similar initial products as with Cs, but with a still higher total internal energy, and would release both $H_2(v)$ and H atoms into the surface and also into the volume. However, H_3^+ ions leaving the plasma will reach the wall with a kinetic energy due to the (positive) plasma potential, which we assume is about 3 eV. The predissociation lifetime for the 2s state is about 10^{-13}/s. During this time a 3-eV H_3^* will travel about 20 Å. The wall interaction is thus not simple to predict.

Mitchell and Graham[13] have considered the dissociative recombination of very low energy (< 1eV) H_3^+ ions with a surface and concluded that there is very little vibrationally excited H_2 formation. Recently, however, Hiskes and Karo[14] have devised a four step model of recombination of H_3^+ ions on metal surfaces. They predict that such recombination will produce vibrationally excited H_2 molecules in o broad spectrum. They find the $H_2(v^*)$ production is relatively insensitive to the energy of the incoming H_3^+ over a range from 1 to 10 eV. The rate of $H_2(v^*)$ produced is such that these molecules make a significant contribution to the total H^- production in the ion source.

3.2 RECOMBINATIVE DESORPTION OF H ATOMS.

From the results of two different but complimentary experiments, Hall et al.[15] and Eenshuistra et al.[16] have recently shown that highly excited H_2 is produced by H atoms from the volume recombining with H atoms physisorbed on the wall, and that these populations survive over a hundred wall collisions.[15] In those experiments, the atoms were produced on hot filaments with no discharge present, and the walls were coated with the filament material. Ta was found to be a more effective filament (or wall) material than W for H_2 vibrational excitation.[15] Similar effects were found by Robie and Bischel (preceding paper), who were able to detect up to v = 11 produced by this method.

Such effects must also occur in discharge ion sources, but the main source of H atoms is in the discharge rather than from the filament. In effect, each ionization event creates about two free atoms. In the direct production by dissociative ionization, the atoms have about 7 eV energy from primary electron ionization of $H_2(v = 0)$. These atoms have sufficient energy to form H^- directly in surface collisions. The H^+ also have 7 eV kinetic energy, and thus will have a good chance of reaching the wall where they are neutralized. However over 90% of the direct ionization yields thermal energy H_2^+, which reacts rapidly (0.5 µs at 30 mtorr) to form vibrationallly excited H_3^+ similar to those in our experiments. These H_3^+ can transfer excitation to H_2 through several proton-exchange collisions, each of which occurs in about 0.5 µs at 30 mtorr. But they will eventually be lost by dissociative recombination or diffusion to the wall. We have already discussed the latter. Dissociative recombination of vibrationally uncooled ions also leads to both 3H (2/3) and H + H_2 (1/3) products.[17] The kinetic and internal energies of these fragments are unknown, but the three-atom channel will yield about 2 eV per atom, and the two-body channel should yield highly excited H_2, similar to the Rydberg state predissociation.

Thus, we see that the discharge is a very efficient producer of H atoms. The less energetic (0-3eV) of these probably recombine dissociatively at the wall to form $H_2(v)$ in the gas, which aids the volume formation of H^-, and the more energetic ones (3-8eV) may form negative ions in surface collisions. The presence of Cs or Ba will decrease the surface work function and thus would help the surface production.

4. PLASMA PARAMETERS AND THE ELECTRON ENERGY DISTRIBUTION

The addition of Cs, or any other material, to the ion source, will alter the basic parameters of the discharge. In particular, changes in the ion species, plasma potential and electron energy distribution function (EEDF) could significantly effect the negative ion current extracted from the ion source. Mullan and Graham[18] have made a preliminary study of the effect on the above plasma parameters, of mixing small percentages of Xenon to a filtered multicusp hydrogen source discharge. Xenon is of interest since it is technically easier to use and has been shown by Walther et al.[2] to produce a factor of 2.7 increase in H^- output when added to a pure H_2 discharge.

Xenon is an additive with some of the features of Cs in that both have large masses and significantly larger ionization cross sections than H_2. However, Walther et al. have shown that Xe and Cs create distinct differences in the discharge chemistry in that Xe leads to a significant increase in the H^+/H_3^+ ratio in the discharge, whereas with Cs this ratio is essentially unaltered. The decrease in the component is probably due to the formation of XeH^+ in collisions of Xe with H_3^+. This has a rate coefficient close to that for H_3^+ formation in H_2^+ - H_2 collisions.

The apparatus used by Mullan and Graham has been described in detail elsewhere.[19] The ion source is similar to that used by Walther et al.[2] in that it is a multicusp, hot-filament driven, ion source which incorporates a virtual filter to prevent energetic primary electrons in the driver region from reaching the extraction region. This ion source is substantially larger, being a box with dimensions 19 x 19 x 24 cm, compared to Walther et al.'s[2] cylindrical source (2.5 cm diameter and 5 cm long). Mullan and Graham[18] measured the plasma parameters and electron energy distribution function using cylindrical Langmuir probes together with a digital data acquisition and analysis system.[19,20]

The operating conditions were chosen to be as close as possible to those used by Walther et al.[2] (i.e., with an arc voltage of 60 V, arc current of 5 A, and gas pressure of 2mtorr). Results were obtained with H_2 alone and with H_2/30%Xe, H_2/10%Xe, and H_2/5%Xe gas mixtures. Measurements were made at various positions axially through the discharge. The results can be taken to indicate the average conditions for that particular region of the plasma. It should be noted that it has been demonstrated that multicusp plasmas are nonhomogenous and measurements at one or two positions should be treated cautiously.[20,21]

In operation, the magnetic filter's role is to prevent fast electrons moving from the driver to the extraction region. In H_2, operating with a filter also results in the electron density in the extraction region being substantially lower than that in the driver. One of the major effects of adding heavy atoms such as Xe or Cs to the discharge should be to increase and change very substantially the momentum transfer rates for electrons in the plasma. This will be particularly important in filtered discharges since the plasma in the extraction region is fed by low energy electrons scattering across the transverse magnetic filter field. In the measurements with Xe, it is found that the change in the electron density across the filter becomes smaller as the percentage of added Xe increases, indicating a more efficient transfer of the low energy electrons compared to H_2. There is also some preliminary evidence from the EEDF measurements in the extraction region that electrons with high energies are also efficiently transferred across the filter. These effects are expected to be even more significant when Cs is added to the discharge and the effectiveness of the filter may be severely restricted as the higher energy electrons scatter from Cs atoms or ions and so cross into the extraction region.

In both the extraction and the driver regions, there is little evidence that significant changes in the bulk electron temperature result from adding Xe. In fact, in the extraction region, a slight increase in the electron temperature is observed. However, a reduction is observed in the fast electron temperature in the driver. The ionization potential of Xe is quite high (12.1 eV) and the addition of Cs with its much lower ionization potential (3.9 eV) may lead to a significant electron cooling effect.

The other significant observation is that the plasma potential increases from about 2 eV to 4 eV when Xe is added to the discharge. This increase is to be expected since the heavy xenon ions will be slower to leave the discharge, and to get charge equality at the walls, the electrons will have to be further contained by increased plasma potential. A similar effect might be expected when Cesium is added. This higher plasma potential will increase the energy of ions striking the walls which in turn may affect $H_2(v^*)$ production from H_3^+ collisions. The energy of H^- ions entering the plasma from the walls will also be increased; such an increase could affect the divergence of the extracted H^- beam depending on the H^- cooling by charge exchange with H atoms in the plasma volume.

5. CONCLUSION

It is apparent that the seeding of multicusp hydrogen ion sources with heavy atoms has a complex effect on the basic atomic, surface, and plasma physics of the discharge. In the gas pressure regime employed by Walther et al., where H_3^+ ions are the dominant hydrogenic species, there will be a significant contribution to the vibrationally excited molecule population through dissociative charge transfer of H_3^+ with Cs. Ths discharge will produce H atoms efficiently. Through collisions with the coated walls of the source these H atoms can contribute both directly and indirectly [through recombination to $H_2(v^*)$] to H^- production in Cs or Ba seeded discharges.

Comparison with results from Xe seeded H_2 discharges suggest that there could be significant changes in the basic plasma parameters of the ion source. The electron density will increase significantly, the effectiveness of the magnetic filter will be reduced and the plasma potential will be increased. From the Xe-seeded work there is no conclusive evidence that significant electron cooling of the bulk electrons occurs.

6. ACKNOWLEDGEMENT

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