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HYDROGEN NEGATIVE IONS AND COLLISIONS OF ATOMIC PARTICLES*

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Abstract

This paper will be an overview presenting some of the basic atomic collisions processes (gas phase) which are fundamental to production and destruction of $H^-(D^-)$. More detailed discussions of the most important processes will be left to other papers at this Symposium, and primarily new results since the 1977 Symposium will be discussed. Recent results provide insight into mechanisms responsible for the high $H^-(D^-)$ ion fractions in hydrogen gas discharges, and the ion-atom collision processes important for "double capture" negative ion sources are better understood than in 1977.

I. Introduction

This paper is a sequel to the paper given at the 1977 Symposium¹ and will concentrate on the new results since that time. Those processes for creation and destruction of H^- or D^- which had been well studied in 1977 left a mystery as to how there could be a large fraction of negative ions in an ion source, since known creation collisions all had cross sections around 10^{-18} cm² or lower while several destruction mechanisms had cross sections exceeding 10^{-14} cm². Various unstudied collision processes were discussed in Ref. 1, and additional ones were suggested at the 1977 Symposium. Various insights have been provided in the interim, principally that vibrationally excited H_2 will have a large rate for production of H^- through dissociative attachment, but the picture is certainly not complete. Nevertheless, the new basic collisions results are exciting for ion source development not only because they provide understanding of what has been observed in discharges but also because they may provide suggestions for improving H^- production in discharges.

Cross sections for conversion of H^+ to H^- by double capture are also better known than they were in 1977. This issue is perhaps of less urgency than direct ion source production since the information was reasonably complete in 1977.² However, the improved collisions data do allow a more reliable assessment of the technique of "double capture" for production of $H^-(D^-)$.

The needs for atomic collisions studies relevant to fusion were recently outlined at a meeting called by the IAEA,³ and the listed needs include needs for ion source development. The atomic collisions needs for modeling and understanding divertor performance in plasma fusion are not yet

*Research sponsored by the Office of Fusion Energy, U.S. Department of Energy, under contract W-7405-eng-26 with Union Carbide Corporation.

well defined, but it is clear that there is considerable overlap of the basic collisions processes operative in ion sources and divertors. This additional area of fusion interest can thus provide further justification for some of the collisions studies discussed here.

II. Electron Collisions

Cross Sections Known in 1977

A number of cross sections of interest were well characterized in the previous paper.¹ Though there were no direct measurements, it was pointed out that three-body recombination for H^- was unlikely in ion sources since for electrons as the third body, a density, n_e , greater than 10^{18} cm⁻³ would be required for three-body attachment to be comparable to radiative attachment (10^{-22} cm²), and a three-body recombination with any atom or molecule as the third body would require a density, n_{atom} , about 10^{16} cm⁻³ to be competitive with radiative recombination. Examples of further specific, well-characterized processes in 1977 are electron detachment, $e^- + H^- \rightarrow H + 2e^-$ (destruction of H^-) with peak cross section near 10 eV of 4×10^{-15} cm² (Ref. 4) and dissociative recombination, $e^- + H_2^+ \rightarrow H + H$ (formation of H^-) with a cross section of 5×10^{-18} cm² at 0.3 eV.⁵

Dissociative attachment, $e^- + H_2 \rightarrow H_2^- \rightarrow H^- + H$, was discussed at some length at the 1977 Symposium in part because it provided a dramatic isotope effect. The resonance state of H_2^- at 3.75 eV above the H_2 ground state can either autodetach (no H^- formed) or dissociate to form H^- . Since dissociation is faster for H_2^- than D_2^- , while detachment is constant, the measured cross sections at 3.75 eV⁶ are 0.9×10^{-24} cm² for D^- formation from D_2 and 1.7×10^{-21} cm² for H^- formation from H_2 - an isotope effect of over three orders of magnitude. While such an isotope effect raised the question of D^- production relative to H^- production in ion sources, the cross sections were so small that no one was alarmed. However, it is this process that has provided greatest interest since 1977.

Dissociative Attachment with Vibrational and Rotational Excitation

In 1978 Allan and Wong⁷ showed that dissociative attachment increased four to five orders of magnitude if the $H_2(D_2)$ were heated to 1400 K with accompanying vibrational excitation up to $v = 4$ and rotational levels up to $j = 7$. Wadehra and Bardsley⁸ explained these results primarily in terms of vibrational excitation. Bardsley will discuss these results in more detail, but they are pointed out here as critical to the understanding of $H^-(D^-)$ production in discharges.

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Fig

These results were missed by previous investigators. In 1967 Chen and Peacher⁹ predicted a strong dependence of dissociative attachment on rotational excitation, and Spence and Schulz¹⁰ subsequently investigated the cross section at 3.75 eV searching for the predicted effect. Figure 1 shows their result that indicated no rotational excitation effect. However, Fig. 2 shows part of the work by Allan and Wong which clearly demonstrates the strong dependence of dissociative attachment on temperature. The results of Spence and Schulz are probably not wrong, but they looked only at electron impact energy of 3.75 eV and hence saw only H⁻ from the ground vibrational state of H₂. The small increase they might have expected in their signal due to rotational heating of ground vibrational state could easily be offset by depletion of the ground vibrational state. The predictions of Chen and Peacher about rotational excitation probably are too high as suggested by the data of Spence and Schulz. When Allan and Wong did the same experiment but with variable energy electrons, an important result emerged. When part of the required 3.75 eV is present as internal vibrational excitation of the initial molecule, the integrated effective cross section (1-5 eV) increased dramatically.

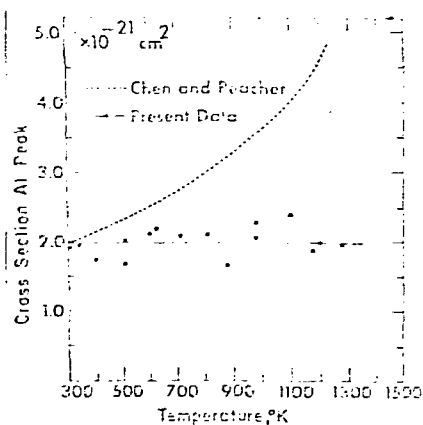


Fig. 1. Temperature dependence of the cross section for H⁻ formation in H₂ at 3.75-eV electron energy (Spence and Schulz, Ref. 10).

There is also good news for ion source production of H⁻(D⁻) on the issue of the isotope effect. When e + H₂(vib) → H₂⁻(vib), dissociation of the negative molecule begins to dominate over autodetachment because of more rapid dissociation when vibrational energy is already present. Thus for D⁻ from D₂(vib), the increase

in cross section is even greater than for H₂ and presumably for sufficiently high vibrational level the H⁻/H₂(vib) and D⁻/D₂(vib) will be equal and autodetachment will be negligible. Thus the effect of vibrational excitation helps to explain not only the high content of negative ions in discharges but also the lack of isotope effect — both recently confirmed in improved measurements by Bacal and Hamilton¹¹ in their low-pressure plasma.

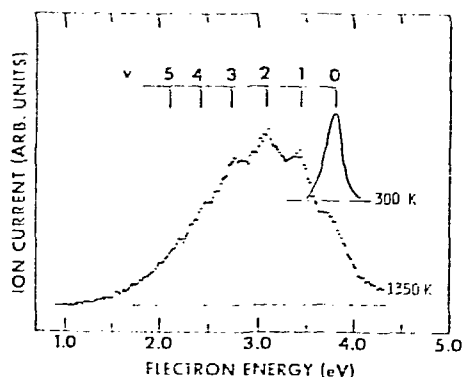


Fig. 2. D⁻ formation in D₂ as a function of electron energy at 1250 K (Allan and Wong, Ref. 7). The inset at 300 K has been added to indicate approximate results at room temperature.

These basic collision studies have provided us with clues to production of negative ions in discharges and could even suggest ways to tune the discharge for higher ion production. However, a number of unanswered questions remain. It is of some interest to ask how general the vibrational excitation mechanism is. For N₂ molecules, recent experimental work¹² shows that for electron dissociative attachment the vibrational heating of the N₂ only increases N⁻ production by about a factor of four rather than the four orders of magnitude in the H₂ case; thus the vibrational heating effects are not universal. In contrast, recent studies by Beterov et al.¹³ (Fig. 3) invoke vibrational excitation to explain an observed increase of negative ion production on surfaces. The negative ion being produced in this case is SF₆⁻, but the authors show that irradiation of the surface with a CO₂ laser greatly enhances negative ion production, and apparently they believe this to be associated with vibrational excitation of adsorbed surface molecules. These two examples are removed from the immediate question of H⁻(D⁻) production but illustrate the broad interest in vibrational excitation.

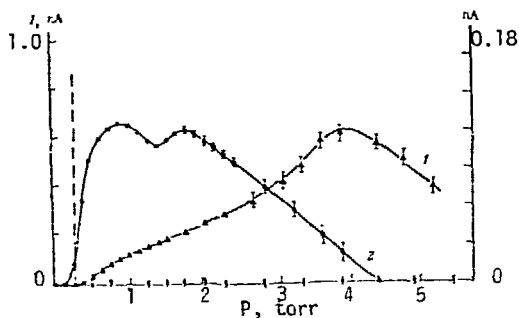


Fig. 3. Negative-ion current produced by SF₆ contact with hot thoriated tungsten.⁶ Curve 1 and scale at left are the normal results while Curve 2 and scale at right show enhancement of negative-ion current when the surface is irradiated with CO₂ laser - 10.6 μ (Beterov et al., Ref. 13).

There are specific unanswered questions about the hydrogen discharge production of H⁻ as well. Hiskes¹⁴ recently reviewed the H⁻ production in light of the vibrational excitation discoveries. For this mechanism to provide the observed H⁻, a significant fraction of H₂ molecules in the discharge needs to be highly excited vibrationally, $v \geq 6$. The question as to where such vibrationally excited molecules would come from has a number of possible answers.

Dissociative Recombination of H₃⁺

Low energy discharges (like the one studied at Ecole Polytechnique^{15,11}) often contain high fractions of H₃⁺. This is at least in part due to the fast reaction of H₂⁺ + H₂ -> H₃⁺ + H which rises rapidly as energy decreases and is above 10⁻¹⁵ cm² at energies below 1.2 eV.^{16,17} At any rate the presence of H₃⁺ in large quantity leads to speculation that the higher than anticipated H⁻ fractions may involve reactions of H₃⁺. The most obvious speculation is that dissociative recombination, e⁻ + H₃⁺ -> H + H₂, could directly produce H⁻. In response to such speculation the cross section was measured by Peart, Forrest, and Dolder.¹⁸ Figure 4 shows their result and demonstrates that the H⁻ production cross section does not exceed 1.8 x 10⁻¹⁸ cm² and is thus an unlikely candidate for the source of H⁻ in plasma. They use the statement of Kulander and Guest¹⁹ that H⁻ production from ground state H₃⁺ would have a threshold near 5.8 eV to infer that their H₃⁺ is vibrationally excited. In addition, Peart et al. reduced ion source pressure to attempt to heat their H₃⁺ vibrationally and found little enhancement of the cross section. (It is noted that Vogler²⁰ infers that the H₃⁺ from such an ion source is likely to be vibrationally excited independent of the source pressure.)

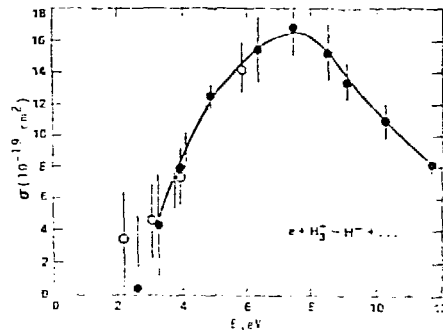


Fig. 4. Direct production of H⁻ from dissociative recombination. Open points are at reduced source pressure to enhance vibrational excitation of H₃⁺ (Peart et al., Ref. 18).

While H₃⁺ apparently does not directly produce H⁻, it might produce the vibrationally excited H₂ needed for dissociative attachment to be effective in H⁻ production. A number of measurements of dissociative recombination, e⁻ + H₃⁺ -> H₂ + H (Fig. 5), demonstrate that this reaction is fast. Statements that the H₂ produced is vibrationally excited are somewhat speculative but Kulander and Guest,¹⁹ theoretically studying the likely state of H₃⁺, suggest that the dissociation will favor vibrational excitation, and the experimental work of Vogler²⁰ on the angle and energy distributions of dissociating H₃⁺ also suggests high vibrational energy in the system.

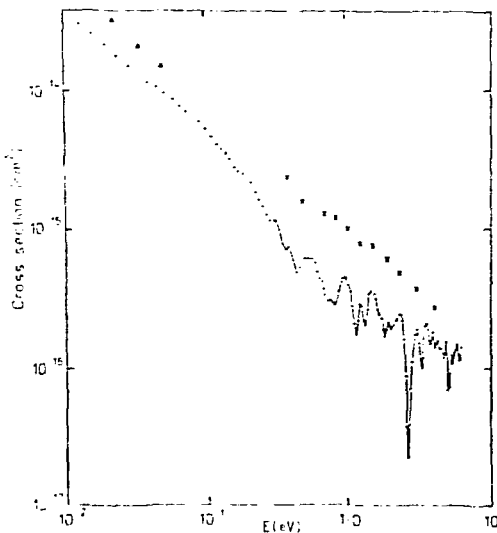


Fig. 5. Total dissociative recombination of H₃⁺. Data are: Δ - Leu et al., Ref. 21; x³ - Peart and Dolder, Ref. 22; • - Auerbach et al., Ref. 23.

Long-Lived States of H_2^- , D_2^- , H_3^-

The question of the possible existence of long-lived (excited) states of H_2^- appears to be still unresolved, as far as theoretical calculations are concerned, although promising candidates²⁵ such as the $4\sigma_g^-(1\sigma_g)(1\pi_u)^2$ state and others²⁶ have been suggested. Experimental results, however, have indicated the existence of both diatomic and even triatomic negative hydrogen (and deuterium) molecular ions that appear to be stable against electron emission with lifetimes exceeding 10^{-5} s.²⁷⁻²⁹ Figure 6 shows the production efficiencies of H^- , D^- , HD^- , D_2^- , D_2H^- , and D_3^- in a hollow cathode duoplasmatron ion source (arc voltage 500 V, arc current 60-100 mA) found by Aberth et al.²⁷ as a function of H_2/D_2 source pressure ratio. Although the relative abundances in ion sources of these molecular negative hydrogen ions compared to H^- (D^-) are quite low (typically 10^{-6} - 10^{-8} at 1 Torr source pressure), insight into their modes of formation and destruction would promote an understanding of the reaction processes occurring in hydrogen plasma sources which produce H^- (D^-).

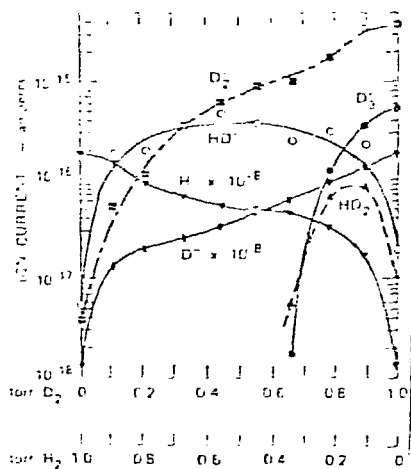


Fig. 6. Observed negative ion currents from a duoplasmatron source with mixture of H_2 and D_2 gas (Aberth et al., Ref. 27).

III. Ion-Atom Collisions

Vibrationally Excited H_2^-

There are at least two known reactions worth mentioning in the context of producing vibrationally excited H_2^- . One, that is also a collisional destruction of H^- , is associative detachment, $H^- + H_2 \rightarrow H_2^- + e_2^-$. This reaction proceeds via the autodetaching $2^1\Sigma_u$ state of H_2^- . Measure-

ment in a flowing afterglow by Fehsenfeld et al.³⁰ gave a room temperature reaction rate of 1.8×10^{-9} cm^3/s implying a cross section near 10^{-14} cm^2 .

Catherinot et al.³¹ performed an experiment to monitor excited states of H ($n = 3, 4, 5$) in a low pressure hydrogen discharge. Their observations are of general interest, but two results are noted here. One is that significant amounts of electronically excited H_2 ($c^3\Sigma_u^-$ and $a^3\Sigma_g^+$) were present in the plasma. The other is that they determine average cross sections in their discharge for excitation transfer H ($n = 3, 4, \text{ or } 5$) + H_2^- - H ($n = 1$ or 2) + H_2 (vibrationally excited). For their mixture of initial H_2 states, they obtain about 1.5×10^{-14} cm^2 for the excitation transfer cross sections which quench excited H but may produce vibrationally excited H_2^- .

There may be invoked a number of such mechanisms for producing vibrationally excited H_2^- , but dissociative recombination, $e^- + H_3^-$, still must rank as a primary candidate.

Ion-Atom Destruction of H^-

As was previously known,^{1,32} mutual neutralization $H^- + H^+ \rightarrow 2H^0$ (Fig. 7); $H^- + H_2^+ \rightarrow$ neutral products (even faster than $H^- + H^+$); and presumably $H^- + H_3^+$ neutrals, are dominant reactions for destroying H^- , or D^- for D^- reactants.

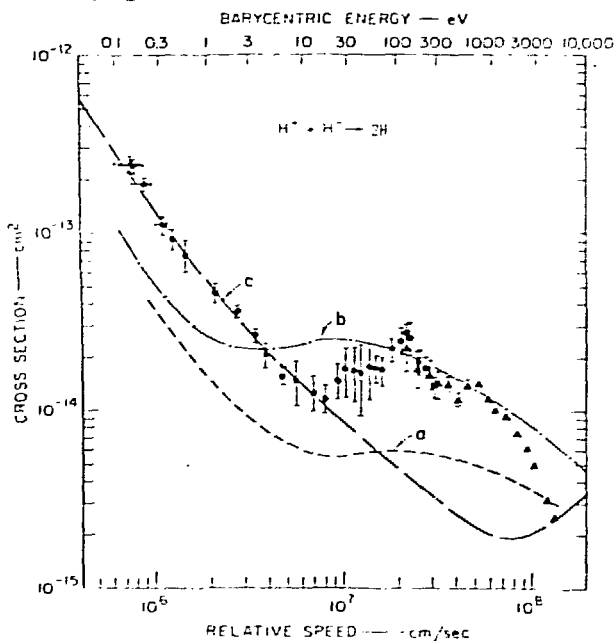


Fig. 7. Cross section for mutual neutralization. Data are: \square - Moseley et al., Ref. 33; \blacktriangle - Rundel et al., Ref. 34. Theoretical curves are: a - Bates and Lewis, Ref. 35; b - Dalgarno et al., Ref. 36; c - Olson et al., Ref. 37. See also Peart et al., Ref. 38. Figure from Ref. 32.

A related reaction which is fast enough to draw interest is associative ionization $H^- + H \rightarrow H_2^+ + e$ shown in Fig. 8 (Ref. 39). The E-1 behavior is familiar by now; it usually signifies high rates at low energy and generally occurs when a negative and positive particle react. The associative ionization probably proceeds through an autoionizing state of H_2 (Ref. 40) and probably results in vibrationally excited H_2^+ .

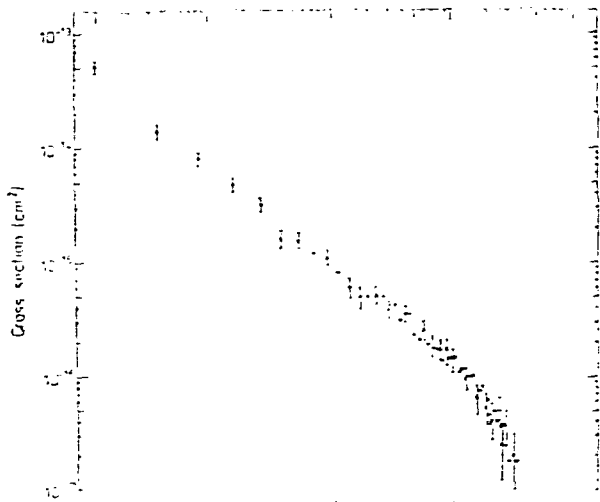


Fig. 8. Cross section for associative ionization; $H^- + H \rightarrow H_2^+ + e$ (Poulaert et al., Ref. 39).

A recent review⁴¹ and an extensive new paper⁴² on negative ion detachment, or stripping, cross sections will direct the interested reader to many results for electron loss or neutralization in $H^-(D^-)$ collisions with atoms and molecules. Other papers at this Symposium will treat negative ion detachment with a view toward employment of this process in neutralization of fast $H^-(D^-)$ beams. Single electron loss resulting in $H^0(D^0)$ has generally large cross sections, typically above 10^{-16} cm² from a few eV to near 1 MeV energies. Between 30 and 200 keV, these cross sections are typically $1-2 \times 10^{-15}$ cm². With respect to neutralization of $H^-(D^-)$ the limit in fractions neutralized at beam injection energies is determined primarily by the ratio of single electron loss to double electron loss. Cross sections for stripping both electrons are usually one order of magnitude smaller than single loss cross sections at beam injection energies which crudely suggests that neutralization with gas targets could be as high as 90%.

Electron loss from H^- in alkali vapors has been of particular interest at low energies (0.1-2 keV) because of interest in production of H^- beams by passage of H^- through these vapors.⁴³ Recent time of flight, as well as differential, cross section measurements on $H^- + Na \rightarrow H^0$ below 1 keV indicate that electron loss occurs by combined autodetachment of the quasi-molecule and charge transfer in proportions roughly 3 to 2. This suggests that in theoretical calculations of electron loss, the interaction of the (alk H^-) and (alk H^0) potential curves at small internuclear separations ($2-4 a_0$), where (auto)detachment occurs, may be just as important to the total loss as the interaction at larger separations ($10-20 a_0$) where the charge transfer occurs.⁴⁴

In the above energy range, experimental electron loss cross sections for $H^- + Cs$ are available⁴⁶⁻⁴⁸ and are included on Fig. 9. Curve 1 on Fig. 9 is the calculated electron loss by Olson and Liu⁴⁴ which contains the long-range coupling. This theoretical result falls below the experimental total loss cross section suggesting that addition of a short-range (auto)detachment component may be appropriate to this case. Qualitatively, the (auto)detachment contribution to the loss cross section should be approximately $\sim R_c^{-2}$ with $R_c \approx 4 a_0$.⁴³ If such a contribution is added to the calculated cross section,⁴⁴ reasonable agreement with the experiment is obtained.

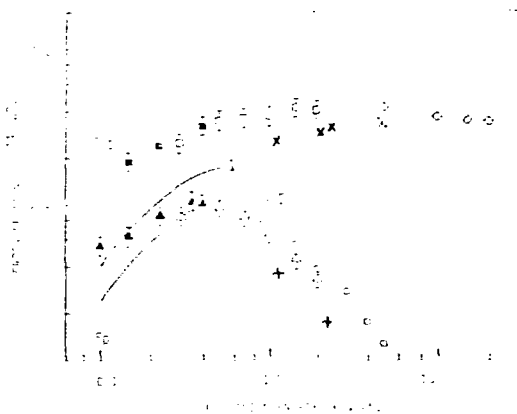


Fig. 9. Electron capture and loss cross sections for hydrogen and deuterium projectiles on Cs. Data are: symbols with error bars (open - hydrogen and closed - deuterium) - Meyer, Ref. 46; \circ - Nagata, Ref. 50; \diamond - Leslie et al., Ref. 48; + and x - Schlachter et al., Ref. 45. Theoretical curves: 1 - σ_{e^-} by Olson and Liu, Ref. 44; 2 - σ_{e^-} by Olson, Ref. 47.

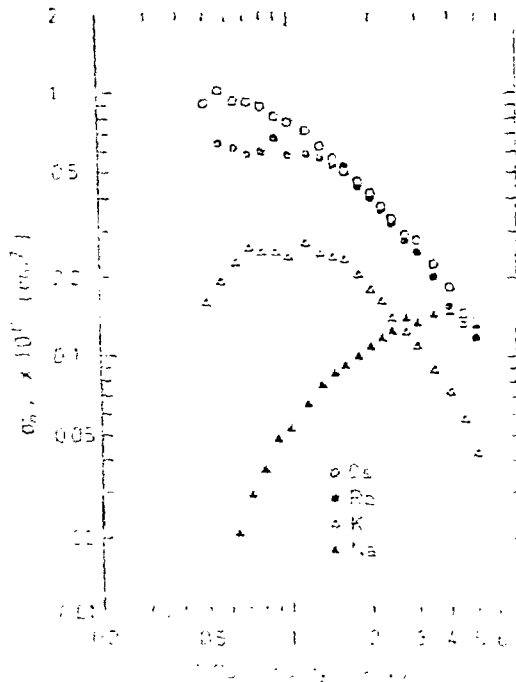


Fig. 10. Cross sections for H^- formation from $H^0 +$ alkalis (Nagata, Ref. 50).

Production of H^- in Ion-Atom Collision

Considerable new data on production of H^- in alkali and alkali earths have been produced since 1977. Refs. 44-51 are samples of the most recent results. Fig. 9 shows some of the cross sections for Cs and Fig. 10 from Nagata⁵⁰ shows measured cross sections for production of H^- from collisions of H^0 with a number of alkalis. Several other papers at this Symposium will discuss these results and their application to H^- beam production. Fig. 11 illustrates the progress in this area since 1977. The original figure is from Schlachter's 1977 paper² but has been changed by removal of results now discounted by the original authors and by addition of recent results by Meyer.⁴⁶ Finally, Fig. 11 shows a reasonably consistent and reliable picture for the equilibrium fraction for H^- in a thick target of Cs.

Of more general interest to the subject of H^- (D^-) formation are recent results for low energy collisions of $H, D + Ar, Kr, \text{ and } Xe$ by Aberle, Grosser, and Krüger.⁵² Figure 12 shows their measurements of ion pair formation $H(D) + Xe \rightarrow H^-(D^-) + Xe^+$ just above threshold (10-100 eV). A remarkable oscillation of the cross section is obvious and is interpreted by the authors with reference to Fig. 13. The oscillation in cross section is interpreted as due to interaction of

the colliding system between molecular curves which have two crossings and which separate to the ion pair observed or to a state of $H(D)$ excitation plus the original rare gas atoms. The isotope effect is an artifact of the energy scale and disappears on a constant velocity scale. In fact, if plotted on a scale proportional to $(\text{velocity})^{-1}$ corresponding to collision time, the oscillations are very regular as has been observed for other scattering events in which such molecular potential curves are appropriate.

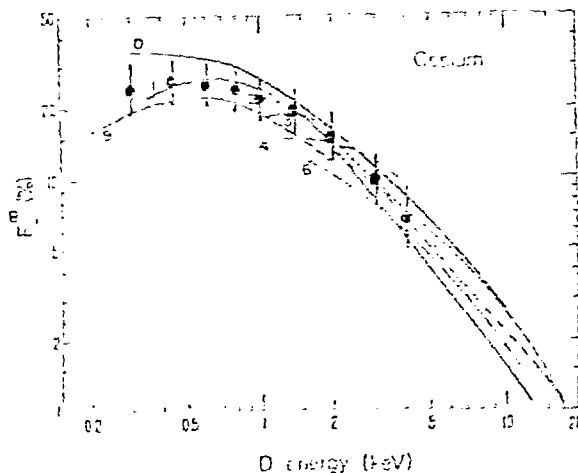


Fig. 11. Equilibrium fraction of D^- for deuterium projectiles in Cs (see text).

Photodetachment of H^-

Photodetachment of H^- received some attention at the 1977 Symposium, and the cross section was given in Ref. 1. The reason for significant interest is that very high neutralization efficiencies are conceivable in a very clean environment employing laser photodetachment.⁵³ The huge resonances in photodetachment cross section near 10 eV (1200 Å)^{34,55} photon energy may even be accessible with new lasers, but the broad maximum in photodetachment in the easily accessible 6000-10 000 Å range is still the obvious choice for a photodetachment neutralizer.

IV. Summary

The atomic collisions data needed to understand double capture production of H^- in beams and for neutralization of H^- beams are reasonably complete. A small portion of these data have been shown here, and more will be presented in other papers. On the other hand, the atomic collisions data for understanding of gas discharge production of H^- seem much less complete in spite of stimulating recent work. To the extent that collisions data can help suggest ways to optimize discharge production of H^- (D^-) and because of parallel application, for example for plasma divertors, it seems highly desirable to extend these data. The roles of vibrational, rotational, and even electronic excitation of molecules and the understanding of molecular negative ions seem quite pertinent to the H^- (D^-) production. The role of H_3 and H_3 molecules in formation of H^- is not well characterized and is important because of large cross sections and high density of H_3 in low temperature discharges.

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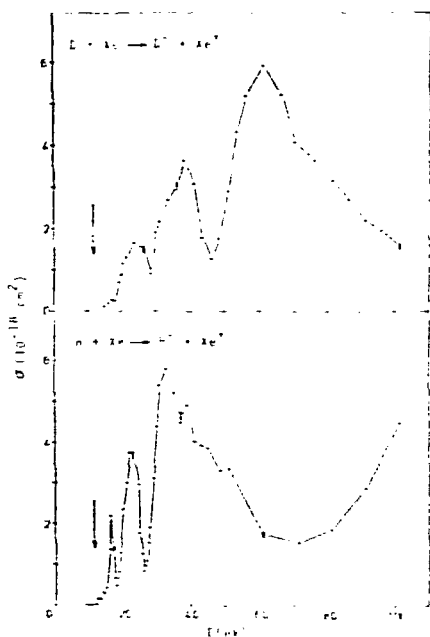


Fig. 12. Formation of H^- (D^-) in Xe (Aberle et al., Ref. 52).

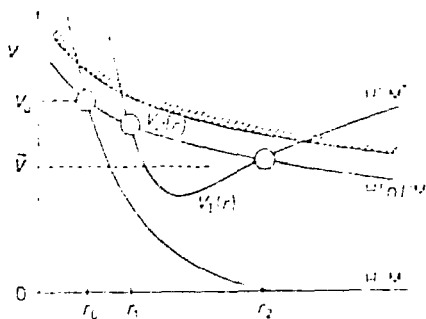


Fig. 13. Schematic potential curves leading to oscillation of cross section for ion pair formation seen in Fig. 12 (Aberle et al., Ref. 52).

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