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EXPERIMENTAL MEASUREMENTS OF NEGATIVE HYDROGEN ION PRODUCTION FROM SURFACES*

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Abstract

Experimental measurements of the production of H^- from surfaces bombarded with hydrogen are reviewed. Some measurements of H^+ and H^0 production from surfaces are also discussed with particular emphasis on work which might be relevant to ion source applications.

Introduction

Recently interest in H^- production from surfaces has been aroused by the need for H^- ion sources for use in injection heating of future MFE plasmas. Studies have suggested that a surface mechanism is creating H^- ions in the present direct extraction ion sources.[1] While there have been many studies of the production of neutral and positively charged hydrogen particles from surfaces, few have investigated the negatively charged component of the backscattered particles, since it was considered to be a small fraction of the total. However, at low impact energies and with particular surface conditions, the negative particles can make a considerable contribution to the backscattered flux.

One of the most pressing problems is to understand the physical processes which create the negative ions at the surface; the theoretical studies of these processes have been outlined earlier.[2]

In this article I propose to review the experimental measurements that have been made on the production of H^- from surfaces bombarded with hydrogen, with particular emphasis on work which might be relevant to ion-source applications. Several experimental measurements of backscattered H^- ions can be applied both to the understanding of these processes and to the construction of ion sources:

1) Total conversion of incident hydrogen particles to H^- as a function of incident energy, angle and surface conditions.

2) The angular distribution of backscattered H^- ions.

3) The energy distribution of backscattered H^- ions.

The backscattering of positive and neutral particles is also important in the overall ion source operation and some relevant measurements are mentioned later.

Experimental Techniques

There have been a number of experimental approaches used to make the measurements outlined above. Since it is not possible to give a detailed account of each apparatus and surface preparation, only the general experimental techniques used will be described.

One of the simplest approaches is to bombard a surface with a known incident flux of particles and measure the total backscattered current. By use of suitably biased grids and magnetic fields, it is possible to suppress all the charged particles backscattered except the negative ions. Particular care must be taken to ensure complete electron suppression. In a similar experimental arrangement it is possible to focus the negative ions emitted from the surface into a mass spectrometer and hence identify the negative-ion species; energy analysis of the negative ions can also be incorporated. The negative ion current is normalized to the incident beam flux. The analysis of these data is complicated by the change in the particle reflection coefficient with energy; at energies below 10 keV this change can be rapid.[3]

A second approach to overcome this problem is to normalize the negative-ion current to the total backscattered flux. However, this entails the detection of neutral particles over the wide range of energies and angles through which they are backscattered. If measurements are taken at a particular scattering angle, an integration over all angles is required to deduce the total conversion efficiency of incident hydrogen particles to H^-.

The greatest problem in interpreting these various measurements is in the reproducibility of surfaces. In general the surface conditions are not well known, with vacuum system pressures in the 10^{-8} Torr range or higher. In most of the work discussed here the surfaces were polycrystalline and had been cleaned with various solvents before installation in the vacuum system. Most

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had then been heated to temperatures where less than a monolayer of adsorbed material was believed to remain on the surface; experiments were performed while the surfaces were still hot or had cooled. However, in other cases, fresh deposits of the particular surface material were laid down prior to the measurements, and pressures in the vacuum system were less than 10^{-9} Torr, enabling clean surfaces to be maintained. (At these pressures an adsorbed monolayer takes about 50 minutes to form.) Studies of positive and neutral backscattering have shown that the influence of adsorbate layers can be very important.[4,5] In the following discussion of results the surfaces have not been shown to be clean, in that they have only been subjected to heating, unless otherwise noted.

Experimental Results

H^- Yield Per Incident Particle as a Function of Energy

Some measurements of H^- production for primary beam energies above 25 keV have been reported. Mitropan and Gumeniuk[6] bombarded Al, Cu, and stainless steel targets with 200 keV to 1000 keV hydrogen and deuterium ions, and measured total negative ion currents of the order of 10^{-4} times the incident current. Using mass analysis they identified H^- as one of the negative ion products and found its abundance was very sensitive to the surface conditions decreasing as the surfaces were heated. Fogel et al.[7] made a more detailed mass spectrometric measurement for H^+ incident on a Mo surface, at an angle of 60° and with energies between 10 and 30 keV. They found the backscattered H^- to total incident beam ratio (K^-) was 1×10^{-5} over their energy range, although it is not clear that they had complete collection of the backscattered H^- ions. Again, heating the target resulted in a reduction in the H^- fraction. In both these experiments the vacuum was of the order of 10^{-6} Torr, so the H^- fractions were determined for "dirty" surfaces, i.e. surfaces with adsorbed gas layers.

Schneider et al.[8] have endeavored to overcome this surface contamination by ensuring good vacuum conditions, in the 10^{-10} Torr range, and by depositing fresh alkali-metal surfaces. The experiment is described in detail in a following paper. Measurements were made of D^- yields from Cs, Rb, K, Na, and Li surfaces bombarded with D_2^+ and D_3^+ in the energy range 0.05 to 3.5 keV/nucleon. A sufficient amount of the alkali metals was deposited so as to ensure that the backscattering occurred in the deposited material and not in the substrate. Some of the measurements are shown in Fig. 1 along with measurements for a "dirty" molybdenum surface. It was found that the yield from D_3^+ was consistently 1.5 times the yield from D_2^+. For convenience the D^- yield is shown divided by the number of incident nuclei.

This shows clearly that the alkali metals produce large negative ion yields, reaching 12% for a cesium surface. Hiskes et al.[9] have suggested that a partial monolayer coverage of a substrate with these alkali metals will result in even larger negative-ion yields. Schneider et al.[8] have made some preliminary measurements which seem to support this theory. This partial coverage phenomenon is thought to be important in surface plasma sources. Dudnikov[10] has made some measurements of the H^- yield from the surfaces in these sources, operating with cesium added to the discharge. The discharge voltage, which determines the energy of the positive ions striking the Mo cathode, could be varied by changing the cathode temperature and cesium injection rate. The ratio of the extracted negative-ion current density to the positive-ion current density at the cathode was then calculated for various values of discharge voltage. The results are also shown in Fig. 1. It should be pointed out that at the optimum cesium injection rate the discharge voltage is around 100 V, while without cesium injection the discharge voltage is around 450 V -- so the results obtained at higher discharge voltages may well be for surfaces with less than optimum cesium coverage. The interesting feature is that at 130 V the H^- yield from the partially coated surface is twice that for a cesium surface.

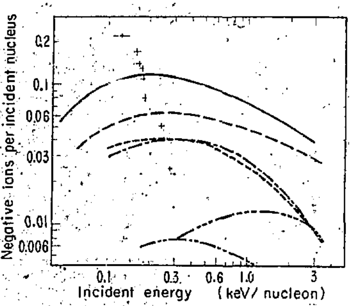

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Fig. 1. Total H^- yields from various surfaces. D_2^+ and D_3^+ incident; ——— Cs surface; — — Rb surface; ---- K surface; —-—- Na surface; —·—·— Li surface; ——-—Mo surface (Ref. 8). +, total H^- yields from H^+ incident on a Mo surface with partial Cs coverage (Ref. 10).

Angular Distribution of Backscattered H^- Ions

At present there are no reported measurements of the angular distribution of backscattered H^- ions. Computer simulations[11,12] predict that the scattering intensity for all backscattered particles at an angle of incidence of 90° will have a cosine distribution about the normal; except at lower incident energies where Oen and Robinson[12] found a slightly more peaked distribution. Some measurements of the neutral and positive hydrogen angular distributions have been made[3,13-14] which confirm a cosine distribution at low emergence energies.

Energy Distribution of Backscattered H^- Ions

Levine and Berry[15] studied the energy distribution of H^- ions produced by H^+ and H_2^+ bombardment of a W surface, with particle energy of the order of a kilovolt and at normal incidence. They positioned the target plate in a mass spectrograph, with the target biased so as to accelerate negative ions produced at the surface. They observed a low energy and high energy peak. The low energy peak had a maximum in the emerging energy spectra at about 3 eV. This was found to be due to adsorbed hydrogen being knocked from the surface; upon heating of the tungsten target to 1100 K, this peak disappeared. The high energy peak was identified as backscattering of the incident particles as H^-. It was noted that H_2^+ behaved like two incident H^+ with one half the incident kinetic energy. At incident energies of the order of 1 keV the peak of the energy distribution of the H^- was found to be 0.6 times the incident H^+ energy.

McCaughan et al.,[16,17] with a similar apparatus, confirmed the existence of these two peaks and also found a medium energy peak in the H^- energy distribution. It was established, by alternating deuterium and hydrogen ion beams on Cu and W targets, that this medium energy peak was due to the re-emission of buried primary ions from the bulk of the target. They noted that the low and medium energy peaks were evident only in the H^- energy spectra and not the H^+ spectra which they also observed. The high energy peak was present in both the H^- and H^+ spectra. Fig. 2 shows the effect of prolonged bombardment on a target which has been extensively cleaned so that the low energy peak was not present. The incident beam was 965 eV H_2^+.

Measurements of H^- energy distributions have been made at higher incident beam energies by Cawthron et al.,[18] and the Garching group.[3,19-21] In both experiments the energy of the backscattered particles was analyzed using electrostatic deflection.

Cawthron et al.[18] bombarded Pt and Ni targets, maintained at red hot temperatures, with various molecular and isotopic species of hydrogen. The particles had incident energies between 2 and 40 keV, were incident to the surface at 45° to the normal, and observed at 45° to the normal, making a scattering angle of 90°. A typical H^- energy spectrum for a Pt target bombarded by protons at various energies is shown in Fig. 3. An H^+ energy spectrum for the same target but bombarded by various molecular hydrogen species is shown in Fig. 4. At low incident beam energies the negative and positive ion backscattering intensities were comparable; however, at the higher energies in the experiment (~ 40 keV) the negative ion intensity was found to be five to ten times less. At the lower incident energies (< 10 keV) the spectra for the H^- ions are similar to, but narrower than, the peaks for the H^+ ions. While the H^+ spectra are found to be forward peaked throughout the energy range, the H^- spectra become flatter as the incident energy increases. It was unclear whether the low energy peak (below about 1 keV) was due to H^- ions or electrons which reach the detector. There is no report of any observed change in the energy spectra with bombardment time. The molecular ions were found to give a higher H^- yield per nucleon than incident protons, but there were insufficient data to observe if there was an isotope effect.

The total H^- yield at 90° was obtained by integrating the energy spectra. A further integration assuming various angular distributions gave values for the fraction of the total beam backscattered as H^-. For incident energies of between 3 keV and 15 keV the total scattered negative fraction was estimated to be around .025 for both Ni and Pt targets; the uncertainty in the exact angular distribution of the particles leads to considerable uncertainty, as large as a factor of two.

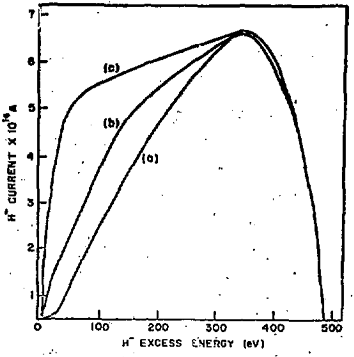

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Fig. 2. Change in H^- energy distribution as a function of bombardment time (primary beam 965 eV H_2^+). Curve (a) immediately after an extensive cleaning operation. Curve (b) after five hours of bombardment at a current density of 0.67 μA/cm² (total dose 7.5 x 10^{16} H_2^+/cm²). Curve (c) after a further 19 hours of bombardment (total dose of 3.6 x 10^{17} H_2^+/cm²). No change was observed past this point. (Refs. 16, 17).

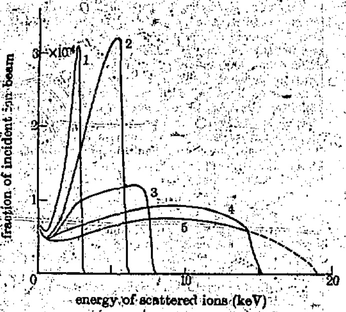

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Fig. 3. Energy spectra of negatively charged particles scattered at 90 ± 3.7° from a platinum target bombarded by protons at various energies: (1) 6.3 keV; (2) 8.3 keV; (3) 11.6 keV; (4) 15.8 keV; (5) 30.5 keV, (Ref. 18).

The Garching group has reported on H^- backscattering from Au,[19,20] Ta,[19] ThO_2,[19] Nb[3,21] and stainless steel[21] targets. The measurements were made with 1 - 15 keV beams of both hydrogen and deuterium. The primary beams were normally incident onto the target and the backscattered particles were observed at an angle of 45° to the normal. The surfaces were cleaned and then heated in the vacuum. The Au target was also sputtered clean with Ne ions and its surface contamination monitored. It was estimated that the Au target had less than one-tenth of a monolayer of adsorbed gas on its surface. The ThO_2 target was a commercially available Ir filament coated with ThO_2. The energy distribution of both H^+ and H^- backscattered from Au are shown in Fig. 5 and from ThO_2 in Fig. 6. The results for the other targets show a similar shape. Energy spectra from a Ta target showed both a molecular and an isotope effect, the molecular effect had also been observed in Nb; yields of H^- were higher per incident proton for H_3^+ and H_2^+ than for H^+ incident. However, neither effect was observed on an Au target. Again there is no report of any observed change in the energy spectra with bombardment time.

It was found that while the peak in the H^- energy spectra shifts to lower energies as the incident energy decreases, the fraction of backscattered H^- to H^+ coincides where the emerging energies overlap, as do measurements obtained with incident D^+ and H^+ beam if plotted versus the energy per nucleon. This is a strong indication that the charge state of the particle leaving the surface is dependent on the emerging velocity.

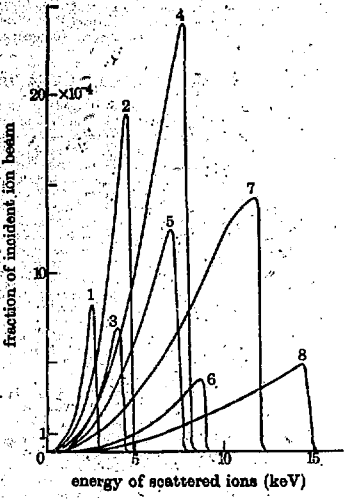

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Fig. 4. Energy spectra of hydrogen particles with positive charge scattered at 90 ± 3.7° from a platinum target at various bombarding energies: (1) H_3^+ at 9.1 keV; (2) H_3^+ at 15.1 keV; (3) H_2^+ at 9.1 keV; (4) H_3^+ at 25.1 keV; (5) H_2^+ at 15.1 keV; (6) H^+ at 9.1 keV; (7) H_2^+ at 25.1 keV; (8) H^+ at 15.1 keV, (Ref. 18).

From earlier measurements of the neutral fraction of hydrogen backscattered from Ta and Au, the negative fraction of all backscattered particles was calculated. For Ta the negative fraction was found to have a maximum of 0.035 at 5 keV, while Au had a maximum of 0.045 at 3 keV. These are consistent with the values calculated by Cawthorn et al. for Ni and Pt, at somewhat higher energies.

The energy spectra obtained by Cawthron et al. *(Figs. 3 and 4)* and *Verbeek et al. (Figs. 5 and 6)* differ in that the former present their data normalized to the total incident beam, whereas the latter present the total measured negative current. However since the incident current is constant throughout an energy sweep the shapes of the curves should be comparable. It is clear from the representative data presented here that there are differences in both the H^+ and H^- spectra. Since the experimental techniques used are quite similar, for want of more information it would appear that these differences may be due to the

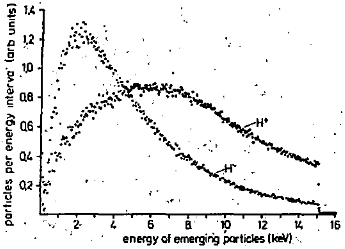

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Fig. 5. Energy distributions of H^+ and H^- ions backscattered from Au bombarded with 15.3-keV protons, (Ref. 19).

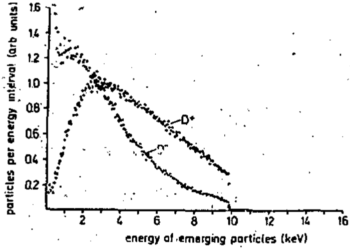

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Fig. 6. Energy distributions of D^+ and D^- ions backscattered from ThO_2 on Ir when bombarded with 10-keV deuterons, (Ref. 19).

different angles of incidence and scattering which are used in the two experiments. As mentioned earlier, no systematic study of these angular dependences has been made.

The charge states of particles emerging from surfaces has also been investigated by passing beams through thin foils. The work of Berkner et al.[22] is of particular interest since they measured the charge state fractions of deuterium beams emerging from freshly deposited surfaces of C, Mg, Nb and Au, in a vacuum system maintained at less than 10^{-8} Torr. The charge states of deuterium particles emerging normal to the surface with energies between 8 and 100 keV were analyzed. In all targets the D^- fraction of the emerging beam was found to increase with decreasing emergence energy. A Mg surface was found to give the highest D^- fraction: 0.12 at an emergence energy of 8 keV.

Other Relevant Measurements

Understanding the overall operation of a negative-ion source requires information on all the processes taking place at the surfaces. Therefore, the backscattering of incident particles in positive and neutral charge states is also of interest. It is not possible in this limited review to go into a detailed discussion of all these processes; however, some measurements should be mentioned.

There has been much work reported on the energy spectra of neutral and positive hydrogen particles backscattered from surfaces.[4,5,18-26] However, due to the difficulty in detecting the backscattered hydrogen atoms, few measurements have been extended to emerging energies of below 5 keV although Behrisch et al.[4] have extended the measurements to as low as 300 eV. In general, it is found that the neutral fraction of the backscattered beam increases and the positive fraction decreases as emerging energy decreases.

There are few measurements of the total fraction of incident hydrogen beams backscattered from surfaces (The reflection coefficient R). The detection of neutral particles with a wide range of energies and scattering angles has been a major problem. Verbeek[3] integrated the measured energy and assumed angular distributions of the neutral and charged particles to calculate the total reflection coefficient for 3-15 keV H^+ incident on Nb. The reflection coefficient increased from 0.02 at 15 keV to 0.34 at 3 keV, although possible oxide layers on the Nb may have resulted in these measurements being low.

Sidenius[27] has used a target mounted in a proportional counter to measure the reflection coefficient for H^+ from Au in the energy range 5 to 50 keV. The reflection coefficient was found to increase with decreasing energy to a value of 0.34 at the lowest incident energy. The high methane pressures in the proportional counter meant that the Au surface was contaminated.

Several authors[28-31] have sought to calculate reflection coefficients by monitoring pressure increases when targets are bombarded with hydrogen. There is considerable disagreement among measurements made at high incident energies.[28-30] The measurements by Baragiola et al.[31] for 300 eV D_2^+ incident on Mo, using a similar technique, give values of $R = 0.31 \pm 0.04$.

Along with these limited measurements there have been several computer simulation calculation of reflection coefficients for hydrogen on metals.[11,12,31]

Discussion

From the results available at present it is clear that H^- production by hydrogen backscattering from surfaces is only significant at energies less than about 10 keV. Surface conditions are very important; and it has been suggested that the work function of the surface is an important factor in determining the backscattered charge state fraction. Experimental measurements would appear to confirm this. This is illustrated in the results of Schneider et al.[8] (Fig. 1) where, as the work function decreases from Li ($\phi \sim 2.4$ eV) to K and Na ($\phi \sim 2.2$ eV) to Rb ($\phi \sim 2.1$ eV) and then to Cs ($\phi \sim 1.9$ eV) surfaces, the negative ion yield increases. The results of Dudnikov[10] are also shown in Fig. 1. At 130 V the cathode has an "optimum" cesium coverage, that is, one in which the work function is minimized at 1.5 eV. As source conditions are altered to change the bombarding particle energy the cesium coverage may be decreasing, hence increasing the surface work function until it reaches the value for a molybdenum target ($\phi \sim 4.3$ eV). This could account for the rapid falloff in the negative *ion yield* with bombarding particle energy in these measurements.

However, in interpreting these data, the reflection coefficient and energy distribution of the backscattered particles are also important. For example, in Fig. 1 the H^- yield from Li ($\phi \sim 2.4$ eV) is lower than for a Mo surface ($\phi \sim 4.3$ eV) at the same incident particle energy. This is because the total number of particles backscattered is less for the Li atoms than for the heavier Mo atoms.

Verbeek et al.[19] have also observed a work function effect in their measurements of the energy distribution of the backscattered H^-. Comparing the energy spectra in Fig. 5 for Au ($\phi \sim 4.8$ eV) and in Fig. 6 for ThO_2 ($\phi \sim 2$ eV) it can be seen *that the maxima in the energy spectra are moving* to lower energies with decreasing work function. For Ta ($\phi = 5.6$ eV) the energy spectrum peak was around 2.5 keV. Similar trends (for the peak in the H^- yield to be at lower energies as the surface work function decreases) can also be seen in Fig. 1.

Interpretation of the available H^- energy distributions is difficult, since they have been made with different angles of incidence and emergence. The low, medium, and high energy H^- peaks observed by Levine and Berry[15] and McCaughan et al.[16,17], who collected the H^- ions coming back with a wide angular distribution, were not observed by Cawthron et al.[18] and Verbeek et al.[19] who collected the H^- ions leaving the surface at specific angles of emergence. Although the fact that Cawthron et al. saw a much broader H^- distribution than H^+ distribution may indicate the presence of the medium energy peak described by McCaughan et al.

The energy spectra obtained by Cawthron et al. and Verbeek et al. at different angles of incidence *and emergence, show little agreement.* It is not yet clear how the energy distribution of H^- ions will change as the angle of incidence and angle of emergence are changed independently of one another. It is also not yet certain whether the molecular and isotope effects described by the backscattered D^- are collected have been found to be independent of the molecular species, if normalized to the number of incident nuclei. However, *they have no comparisons with monatomic incident* beams or different isotopes.

Conclusion

It is possible to produce surfaces that will give sizable yields of negative hydrogen ions when bombarded with hydrogen beams. The measurements available to date indicate that surfaces with low work function and high reflection coefficients will be most desirable for ion source construction.

Before the physical processes which lead to H^- production from surfaces can be understood, it is clear that more experimental information is required.

Several areas would be of particular interest at the moment.

1. Backscattering of particles with incident energies between 1 eV and 1 keV from surfaces, especially those with a partial coverage of alkali metals.

2. The angular dependence *of H^- produced* by surface bombardment.

3. The sputtering of adsorbed or implanted hydrogen from surfaces.

Item 1 is presently under investigation by our group at the Lawrence Berkeley Laboratory.

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