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Negative-Hydrogen-Ion Production by Low Energy Hydrogen Atom Bombardment of Surfaces*

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Abstract
A hydrogen furnace was used to irradiate a molybdenum surface with a flux of low energy hydrogen atoms. If the surface work function was reduced by evaporating Cs onto the surface, a small fraction of the incident hydrogen atoms were observed to be backscattered as negative ions.

The negative ion production dependence on the surface work function and on the atom energy distribution in the incident beam were investigated.

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Introduction

Magnetic fusion experiments will require efficient neutral beam injection systems at energies above 200 keV for plasma heating. The systems will most likely employ intense negative deuterium ion beams. Experiments have shown that it is possible to dramatically increase negative-ion yields from ion sources by adding an alkali metal to the discharge\(^1,2\). In \(H^-\) sources, the addition of Cs to the discharge resulted in increases of more than an order of magnitude in \(H^-\) current density\(^1,3\). Belchenko, Dimov and Dudnikov\(^1,3,4\) and Hiskes, Karo and Gardner\(^5\) have proposed models based upon surface production as the principal mechanism for \(H^-\) formation in these sources.

Belchenko, Dimov and Dudnikov\(^1,3,4\) have proposed that any hydrogen atoms adsorbed on the cathode surface of their ion sources have a high probability of residing as a negative ion and can be desorbed from the surface as a negative ion by an incident energetic particle from the discharge. The addition of Cs to the \(H^-\) source lowers the surface work function and enhances the probability of escape without destruction of the negative ion from the surface, and hence increases the \(H^-\) yield. In this mechanism incident energetic particles are required to liberate the hydrogen from the surface.

Hiskes, Karo and Gardner\(^5\) have hypothesized that \(H^-\) ions are formed in the collisions of 1 to 100 eV hydrogen atoms with adsorbed Cs atoms. As the hydrogen atom approaches the Cs atom the interaction potential is the sum of the image potential and the difference between the CsH and CsH\(^-\) molecular potentials. This interaction potential allows the transfer of an electron from the substrate to the hydrogen atom, which may escape from the surface as \(H^-\). Hiskes and Karo\(^6\) predict, from this mechanism,
that the probability of formation and escape of \( H^- \) ions from surfaces with suitable work function could be very high, even for hydrogen atoms with incident energies of only one electron volt.

There exists very little experimental information on negative ion production from surfaces and if the processes involved in present ion sources are to be understood, or for the ion sources to be improved it is necessary to have experimental measurements of negative hydrogen ion production.

The purpose of the present experiment is to investigate the possible production of \( H^- \) or \( D^- \) ions from surfaces bombarded with low energy (few eV) \( H \) or \( D \) atoms. A tungsten tube furnace, heated to 3000 K can provide a large flux of \( H \) atoms with an energy distribution peaked at about 0.25 eV but extending to higher energies. When single particle detection techniques are used negative ions produced (even by the portion of the beam with energies in excess of 4 eV) should provide measurable fluxes.

Experimental Approach

(i) Apparatus

Figure 1 shows a schematic diagram of the present apparatus. The main vacuum chamber was divided into two differentially pumped sections. In one section the atomic hydrogen source, a tungsten tube furnace was located. This section was pumped with an oil diffusion pump and separated from the other section of the chamber by an aperture (A) which not only acted as a collimator for the atomic hydrogen beam but also as a pumping impedance. The second section of the chamber contained the molybdenum target assembly, the negative ion detector, the cesium dispenser and the work function measuring filament. The two latter devices could be alternatively positioned in front of the target. The target region was pumped by a cryopump and a low magnetic field ion pump.
Base pressures in the target region were typically of the order of $2 \times 10^{-6}$ Pa which could be maintained during cesium evaporation and rose to $3 \times 10^{-6}$ Pa when the tungsten tube furnace was operating.

(ii) H atom Source

The H atom beam was produced by a resistively heated tungsten tube furnace, with a wall thickness of 0.03 cm and an internal diameter of 0.315 cm. A 0.06 cm diameter aperture in the tungsten tube furnace was aligned with the aperture A, diameter = 0.175 cm and the target. These apertures were such that the atomic hydrogen beam had a diameter of 0.2 cm at the target. The hydrogen pressure in the furnace was measured in the gas feed line by a capacitance nanometer. The furnace operating temperature was measured using a calibrated optical pyrometer.

The H atom flux from the tungsten tube furnace was calculated using the equation for the number of particles to strike a surface, derived by Ramsey:

$$I = 1.118 \times 10^{22} \frac{p \cdot A_s \cdot A_t}{l_0^2 \cdot (M \cdot T)^{\frac{3}{2}}} \text{ atoms/sec}$$  \hspace{1cm} (1)

where $p$ is the atomic hydrogen pressure in Torr, $A_s$ is the area of the source aperture, $A_t$ is the area of the target, (in our case the area of the collimating aperture), $l_0$ is the source to target distance, $M$ is the molecular weight and $T$ is the temperature of the source in K.

For typical furnace operating conditions, i.e. 2800 K and 13.3 Pa (0.1 Torr). The calculated flux of H atoms (which comprised more than 90% of the beam) is $10^{14}$ atoms/sec. These calculations were confirmed by placing a U.T.1 quadrupole residual gas analyser equipped with an ionizing head, directly in front of the furnace and measuring the H atom flux as a function of source pressure. The
measured fluxes were in good agreement with the calculated fluxes to within the experimental uncertainties, mainly in the calibration of the electron multiplier and in the efficiency of the ionizing head.

The velocity distribution, and hence the energy distribution, in the H atom beam was calculated using the expression:

\[ F(V^1) = 1 - \int_{0}^{V^1} \frac{2}{\sigma^4} \sigma^3 \exp\left(-\frac{\sigma^2}{\sigma^2}\right) d\sigma \] (2)

where \( F(V^1) \) is the fraction of atoms in the beam with velocities greater than \( V^1 \) and

\[ \sigma = (2kT/M)^{1/2}. \]

Solving this equation for an atomic hydrogen beam from a furnace at 3000 K, indicates that 6% of the atoms have energies greater than 1 eV, 0.1% have energies greater than 2 eV and \( 8 \times 10^{-5}\% \) have energies greater than 4 eV.

(iii) Surface Conditions

The polycrystalline molybdenum targets used in the present experiment underwent no special treatment before installation. Once installed the target was heated to greater than 900 K for 12 hours in a vacuum of about \( 6 \times 10^{-6} \) Pa. It was then allowed to cool to room temperature before Cs was evaporated onto the target area from an S.A.E.S. Cs dispenser which could be positioned immediately in front of the target.

After each successive evaporation the dispenser was withdrawn and the filament assembly positioned in front of the same target area so that the work function change could be determined using the electron beam retarding potential method. The small filament and the target formed a simple diode. Thermal electrons created at the hot filament were accelerated or retarded by the work function
difference between the sample and filament. From the shift in
the I-V characteristic of this simple diode the work function change
due to each successive cesium deposition could be determined.

Wilson\textsuperscript{10} and Swanson and Strayer\textsuperscript{11} have investigated the
change in work function of a molybdenum surface with increasing
cesium coverage. They report the work function decrease from the
clean Mo value of about 4.3 eV to a minimum value of about 1.6 eV
before rising to a value of about 1.8 eV which remains unaltered
with further cesium coverage.

The present measurements are of work function change after
successive cesium evaporations onto uncleaned polycrystalline Mo. Typica
results for Cs evaporation onto a Mo surface which had been heated to 900 K and then cooled to room temperature are
shown in figure 2. The work function is shown to increase with
successive evaporations until a saturated value is reached. There
is no indication of a minimum in the work function. This could
be due to contaminants on the Mo surface or the fact that the Mo
in this case is polystralline since there is evidence, for example,
for Ta\textsuperscript{9}, that for a polycrystalline surface the work function minimum
effect is reduced.

Over a series of measurements it was noted that, as expected,
more Cs evaporations were required to reach the saturation value
as the Cs output per evaporation was reduced. An average saturation
work function change of 2.6 eV ± 0.2 eV was measured, the large
uncertainty is due to differing work functions at the beginning
of the evaporation sequences. Assuming an initial work function
of about 4.3 eV, the final work function value of about 1.7 eV is
consistent with that to be expected for saturated Cs coverage on
Mo.
When the surface was subjected simultaneously to the hydrogen flux from the tungsten tube furnace the work function dependence on Cs coverage was unchanged.

(iv) Negative Ion Detection

The Mo target was biased at a potential of -100 V during hydrogen bombardment. Any negative ions created on the surface would therefore be repelled. The two aperture lens system, designed using the EGUNS computer program\textsuperscript{12}, ensured that a high fraction of these negative ions were focused to form an acceptable beam at the entrance of the negative ion mass analyser. The EGUNS program predicted that, with appropriate biases, 20% of the negative ions produced on the target with energies of 0.5 eV would enter and be transmitted through the quadrupole used for mass analysis. This fraction was essentially independent of whether the initial ion distribution was assumed to be uniform, Gaussian or Cosine. Higher collection efficiencies could be obtained by using larger biasing potentials, however, the quadrupole resolution was better at lower incident ion energies.

Experimental tests of the general features of the focusing system were in accord with the computer program predictions. Typical operating voltages for electrodes I and II were -30 V and -90 V respectively.

The quadrupole mass filter used for mass analysis was a U.T.I 100c residual gas analyser with the ionising head removed and modified for negative ion detection. The ions, having passed through the quadrupole, were deflected off axis and detected using an electron multiplier.

The signal from the electron multiplier was amplified before being counted on a scaler or displayed on a ratemeter. The ratemeter
output could be plotted as a function of the voltage applied across the quadrupole to produce a mass spectrum.

Results

When H atoms were incident on an untreated Mo surface, i.e. one onto which no Cs had been deposited, a background count rate of less than 100 counts/sec was observed, independent of quadrupole voltage. However when Cs had been evaporated onto the surface a sizeable signal was obtained. The dependence of this signal on the quadrupole mass scan is shown in figure 3A and indicates the production of negative hydrogen ions.

The initial count rate prior to the appearance of the pronounced peak is a feature of the quadrupole operation, in that all ions initially are transmitted prior to the quadrupole voltage reaching a high enough value to deflect them. The mass scan shown in figure 3B shows the signal dependence when operating with incident deuterium atoms. This shows clear evidence of negative deuterium ion production and also the differing response of the quadrupole with mass.

Several tests were made to ensure that these peaks represented negative ions produced by hydrogen or deuterium atoms. In order to confirm that the signal was not due to electrons the target was replaced by a filament which was heated to emit electrons. Under the same biasing arrangement as for normal operation a signal was detected, this was found to appear, on a mass scan well to the low mass side of the mass one peak position, again, due to the transmission of electrons during the initial phase of the mass scan. It was noted that the application of a small permanent magnetic field of the order of 10 G in the vicinity of the filament removed this electron signal. This magnetic field was applied during the subsequent negative ion observations.
In normal operation when the mass analysis was set so as to accept only ions of mass one (or mass two for deuterium operation), the count rate remained unchanged when the quadrupole fields were switched off, implying very few electrons from the surface reached the quadrupole.

The negative ion signal was investigated as the furnace temperature and pressure were varied. In figure 4 it can be seen that for a fixed furnace temperature the negative ion signal was linear with hydrogen or deuterium gas pressure. This is to be expected from equation 1 as the total number of hydrogen atoms in the beams varies linearly with pressure, while the energy distribution is independent of pressure. The negative ion signal was found to decrease rapidly with furnace temperature for a given gas pressure. The behaviour of the hydrogen atom flux with temperature is complex, since both the number of hydrogen atoms in the beam and their energy distribution is dependent on the temperature. However, from equation 2 a rapid decrease with temperature would be expected, this temperature dependence is discussed in more detail below.

The possibility that positive hydrogen ions created in the tungsten tube furnace were contributing to the production of negative ions was eliminated by observing that the negative ion signal showed no change as the furnace was biased to -100 V with respect to the target.

As a result of these observations it was concluded that negative hydrogen ions were being produced from targets, onto which Cs had been evaporated, as a result of hydrogen atom bombardment.

Having confirmed that negative hydrogen ions were being produced from the target surfaces the dependence of this negative ion production on work function and incident hydrogen beam conditions was investigated.
The general procedure was to evaporate Cs, measure the work function of the surface followed by a measurement of the negative ion yield from that surface then evaporate more Cs onto the surface. The measurements obtained in this fashion were relative to the particular furnace operating conditions, since the position of the furnace aperture would vary under different operating conditions, due to thermal expansion of the tube. This would effect the hydrogen atom beam transmission through the collimation. For comparison of measurements made at differing temperatures, as described below, it was necessary to optimize the negative ion signal at each temperature by adjusting the position of the furnace.

Typical data, obtained in this way with incident deuterium atoms, is shown in figure 5. The dependence of the negative ion yield with successive Cs evaporations is characteristic of all measurements for both incident deuterium and hydrogen atoms. Typical data for incident hydrogen atoms with different Cs evaporation rates and various furnace pressures is shown in figure 6. In all cases the negative ion signal initially rises rapidly with work function reaches a maximum value and then starts to decrease, even though the work function remains constant. From equation 1 the total number of atoms striking the surface under the same furnace pressure and temperature conditions would be expected to vary as \( M^{-\frac{3}{2}} \). In the case of hydrogen and deuterium, this corresponds to a 30% difference, however, the uncertainties in the present measurements are too large to adequately distinguish an isotope effect.

The work function measurement shown in figure 2 illustrates the general features of work function change with cesium coverage. In figure 5 the surface had not been recycled hence the initial work function is lower than for example in figure 2.
The reason for the pronounced decrease in the negative ion yield at higher Cs coverages is unclear, but could be due to a change in the reflection coefficient for the incident atoms on the surface as the Cs coverage increases. However there exists little information at present on reflection coefficient effects at these low energies.

For the same furnace pressure and temperature conditions the maximum negative ion yield was reproducible from one sequence of evaporations to another to within 40%. For furnace pressure and temperature conditions which would produce a calculated incident hydrogen atom flux of the order of $10^{14}$ atoms/sec, the maximum flux of negative ions was about $8 \times 10^6$ per second. Considering the calculated collection efficiency of the detection system, the conversion rate to negative ions is clearly extremely small. However, this could be interpreted as only the more energetic atoms producing negative ions. Clearly to fully investigate this phenomena requires a velocity selected low energy atomic hydrogen beam. However, as a preliminary investigation, the number of negative ions produced under varying furnace temperatures can prove interesting.

As the furnace temperature is varied, while the pressure is kept constant, then as noted previously both the total number of H atoms and the energy distribution of those atoms change. However by a combination of pressure and temperature changes it is possible to maintain the same total flux of hydrogen atoms while varying the energy distribution, hence changing the number of atoms in the beam which exceed particular energies. The calculated relative dependence of the number of atoms in a beam produced under varying furnace conditions with energies exceeding 1 eV, 2 eV, 3 eV and 4 eV using equations 1 and 2, are shown in figure 7. Also shown is the relative maximum yields of negative hydrogen and deuterium ions produced with the furnace operating under these
conditions. Clearly the negative ion yield is strongly dependent on the energy distribution in the atomic beam, and apparently most of the negative ions are produced by atoms in the beam with energies of greater than at least 3 eV.

Conclusions

It has been demonstrated that negative hydrogen ions can be created by reflection of low energy hydrogen atoms from a surface with a low work function. The fraction of negative ions produced by bombardment of atoms from a tungsten furnace is low, but there are indications that the negative ions are produced by atoms with energies in the high energy tail of the energy distribution. These observations suggest that a backscattering model, such as that proposed by Hiskes et al\textsuperscript{5}, may play an important role in the formation of negative hydrogen ions in ion sources. Further more detailed investigations of negative ion production from low energy bombardment are clearly desirable.
Acknowledgements

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References


Figure Captions

Figure 1  Schematic diagram of experimental arrangement.
Figure 2  Change in molybdenum target work function with successive cesium evaporation.
Figure 3  A typical mass dependence of the negative ion signal (A) with incident H atoms (B) with incident D atoms. The lower trace in (A) is the signal with the furnace operating with residual gas.
Figure 4  Variation of the negative ion signal with furnace gas pressure. Operating with O, H$_2$ and D$_2$.
Figure 5  Negative ion signal and work function variation with successive cesium evaporation at a furnace temperature of 2500 K.
Figure 6  Negative ion signal variation with successive cesium evaporation with incident H atoms at a furnace operating temperature of 2700 K and pressures of 0.14 Torr, 0, 0.1 Torr, 0.05 Torr. The open symbol data points were obtained with a larger cesium flux per evaporation.
Figure 7  The lines show the calculated variation of the energy of the atoms in the beam with different furnace operating conditions. (The furnace operating conditions being such that the total hydrogen atom flux is constant while the energy distribution of the atoms in the beam varies.) The symbols show the variation of the measured negative ion signal under these operating conditions during several runs. Closed symbols are for D atoms, open symbols are for H operation.
RELATIVE YIELD

NO. OF CESIUM EVAPORATIONS

CHANGES IN WORK FUNCTION (\(\psi\))

FIGURES
Figure 6
\[ D^+ \text{ FRACTION, RELATIVE TO YIELD AT 3000 K} \]

\[ >1\text{eV} \]
\[ >2\text{eV} \]
\[ >3\text{eV} \]
\[ >4\text{eV} \]

\[ \text{FURNACE TEMPERATURE AND PRESSURE} \]

**Figure 7**
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