AN INVESTIGATION OF CHARGE STATE EFFECTS IN ATOM-SURFACE SCATTERING

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Abstract

The dynamics of low energy atomic collision and ionization processes at surfaces are under investigation for satellite applications. Atomic scattering experiments have been performed under high vacuum conditions (~$10^{-8}$ Torr) similar to those encountered in space environments. Experimental results indicate that energy loss in fact depends on the charge state of the incident atom. Measurements have been accomplished for all charge states of atomic hydrogen ($H^+$, $H^-$, H) impacting a copper surface and scattering as $H^-$. Energy and angular distributions of scattered $H^-$ are presented here for collision energies ranging from 25-200 eV. All scattered $H^-$ energy distributions exhibit a broadened peak associated with inelastic collisions followed by charge exchange. However, for collision energies less than ~100 eV, the charge state of the incident atom has measurable effects on the energy distributions of resulting $H^-$. These $H^-$ energy distributions resulting from incident $H^+$ and H are similar, however the apparent threshold energies differ by 4.7 +/- 0.5 eV. In addition, the energy distributions of scattered $H^-$ resulting from incident $H^+$ exhibit a secondary peak attributed to ion survival.

1. Introduction

The interactions of ions and beams with materials have long been a subject of investigation within various scientific communities. Of particular interest are those collisions involving charge exchange at surfaces, i.e. electron transfer between the scattering particle and the target media. This work investigates the effects of charge state in such collisions of atomic hydrogen with a copper surface.

Historically, the production and containment of hydrogen plasmas in nuclear fusion reactors has motivated investigation of the physics and chemistry involved in low energy (less than ~1 keV) collisions of atomic and ionic hydrogen with surfaces.1-4 The development of intense negative ion sources for fusion research has likewise stimulated a large number of investigations into these interactions.1-4

More recent applications of these charge exchange processes are emerging in the space sciences. For the many Earth-orbiting satellites, these processes can contribute to the buildup of excess charge as the spacecraft drags through the upper atmosphere, possibly resulting in damaging electrical discharges among sensitive electrical equipment.9,10 A characterization of these collision processes is therefore an important factor in spacecraft design and engineering. In recent magnetospheric studies, surface charge exchange has been used to analyze fast neutral atoms emerging from space plasmas. This use of a "converter surface" has been successfully applied as the basis of the LENA (Low Energy Neutral Atom) instrument on the IMAGE satellite.11 In this instrument, the scattering of low energy H and O atoms from a tungsten surface yields the charge-exchanged ions $H^-$ and $O^-$, which are analyzed with electrostatic and time of flight techniques. This analysis is an important part of understanding the outer atmospheric interactions occurring in the solar system.

Given the variety of current and potential applications of charge exchange collisions, the details of energy loss and charge transfer mechanisms need to be understood. Due to the level of complexity of the atom-surface system, especially for "real" or applicable surface conditions, theoretical development has been somewhat limited, and studies are largely experimental.

Laboratory studies of neutral-ion charge exchange at surfaces have long been performed using incident positive ions,12-16 mainly due to the difficulties associated with producing a well-characterized beam of variable energy ground state atoms. These studies are performed under the assumption that an incident ion is neutralized upon entering the surface, thus all dynamics and charge exchange processes associated with the final ion state are equivalent for incident ions and atoms. Very few studies comparing the effects of incident charge state have been performed,17,18 and no experimental comparisons of atomic ions and neutrals have been performed at collision energies less than 100 eV. This report presents the results of measurements of energy and angular distributions of $H^-$ produced in 25-200 eV collisions of $H^+$, H, and $H^-$ with a copper surface.
2. Experimental

a. Atomic Beam Instrument

The atomic beam apparatus incorporated in these measurements has been described in detail previously so only a brief overview is given here. The instrument consists of two differentially pumped high-vacuum chambers forming the ion beam and target regions. Mounted on the first chamber is a duoplasmatron ion source, from which either \( \text{H}^- \) or \( \text{H}^+ \) ions are extracted and focused into a beam. Ions are mass analyzed with a magnetic sector, and focused through a differential pumping aperture into the second chamber. Pressure in this first chamber is typically \( \sim 10^{-5} \) Torr during operation due to the gas load from the ion source. After entering the second vacuum chamber the ions are passed through a 9° electrostatic bend, eliminating from the beam any fast neutrals produced via charge exchange or charge stripping collisions along the ion beam path. For the case of H atom beam formation, the H beam intersects an extended cavity Nd-Yag (1064 nm) laser beam prior to entering an electrostatic ion deflection and measurement region. The photodetachment process \( (\text{H}^- + \text{hv} \rightarrow \text{H} + e^-) \) neutralizes a fraction (5-20%) of these ions, resulting in a beam of purely ground state H atoms which impact the target surface. The difference in measured ion fluxes with the laser on and off is an absolute measure of the neutral atom flux. For incident ion measurements, the laser is off and the \( \text{H}^- \) or \( \text{H}^+ \) beam is directed to the surface. Typical ion and atom flux levels in these experiments were on the order of \( \sim 10^{-12} / (\text{cm}^2 \text{s}) \).

b. Scattering Measurements

Measurements of negative ion energy distributions were performed using standard retardation techniques, and the analyzer has been described in detail previously. Briefly, an electrostatic retarding potential analyzer aligned in the plane of incidence was rotated 45 degrees around the perpendicular axis passing through the target surface, as shown in Figure 1.

The ion or atom beam impacted the scattering surface at 72° from the surface normal, and negative ions were energy analyzed at various scattering angles within the forward quadrant.

Prior to insertion in vacuum, the copper surface was wet polished and rinsed in ultrasonic baths of methanol and deionized water. The entire vacuum chamber was heated to 100°C for ~36 hours during pumpdown, after which a base pressure of \( 5 \times 10^{-9} \) Torr was achieved. No further in vacuo surface cleaning was performed. Residual gas analysis performed in this system has shown the vacuum background to consist mainly of hydrogen and water vapor. During operation the pressure was \( \sim 10^{-8} \) Torr due to \( \text{H}_2 \) from the ion source. The results presented here therefore pertain to a "gassy" surface containing at minimum one monolayer of adsorbed gases.

The total ionization efficiency is defined here as

\[
\gamma_{\text{tot}} = \frac{N_{\text{tot}}}{N_{\text{inc}}} \int \int D(E, \Omega) dE d\Omega
\]  

where \( N_{\text{tot}} \) denotes the total number of negative ions leaving the surface at all energies, \( N_{\text{inc}} \) denotes the total number of incident ion or atoms impacting the surface, and \( D(E, \Omega) \) is probability distribution for negative ion production. The quantity measured in these experiments is referred to as \( \Delta \gamma^- \), and is related to the ionization efficiency through the integrand in (1) by

\[
\Delta \gamma^- \propto \int \int \int \frac{\partial}{\partial E} \frac{\partial}{\partial \Omega} D(E, \Omega) dE d\Omega
\]

where the spatial integral is carried out over the detector solid angle, \( \Delta \Omega_{\text{det}} \). Equation (2) is simply the projection of the negative ion yield onto the detector aperture.

The definitions above refer to all negative ions produced at the surface, and the electrostatic analyzer has no mass selection capabilities. However, the only electronegative elements present in large quantities at the surface are hydrogen and oxygen, both present in these high vacuum conditions. The low mass of hydrogen results in very inefficient sputtering of higher mass particles, and it is therefore assumed that all negative ions measured here are \( \text{H}^- \). Additionally, secondary electrons are produced at the surface with energies less than \( \sim 5 \) eV. The secondary electrons are discriminated by keeping the lower limit of the energy integral in (2) at 20 percent the collision energy.
3. Results and Discussion

a. H\(^+\) and H

Angular distributions of scattered H\(^+\) resulting from incident H\(^+\) and H at 200 eV are shown in Figure 2. The horizontal uncertainty bars denote the 6° aperture of the detector. For any given measurement, the value of \(\Delta \gamma\) is uncertain by less than 2%. However, due to long term evolution of vacuum and surface conditions, these values have varied by as much as a factor of 2. Although this variability masked any systematic trend in \(\Delta \gamma\) with collision energy, the shapes of the negative ion angular distributions were essentially identical at all collision energies for both H\(^+\) and H incident. The steep decline of \(\Delta \gamma\) towards small angles is expected as the detector begins to pass into the shadowed region at 18°. The peak near the specular direction (36°) is a common feature observed in many ion-surface scattering experiments.\(^{12,13,27,29}\) The sharpness of this peak is expected to depend on the surface roughness.

The energy spectra of H\(^+\) resulting from 200 eV H\(^+\) and H are shown in Figure 3. All data have been normalized to the height of the peak in the energy spectra. As expected from collisional kinematics, the peak progresses to lower energies, and the low energy tail becomes more pronounced as the scattering angle increases. The shape of these distributions should depend at least on the energy loss of the incident particle due to scattering and electronic stopping, and to a lesser extent the charge exchange transition rates for negative ion formation. At this energy, these distributions exhibit no measurable dependence on the charge state of the incident particle, suggesting incident H\(^+\) is neutralized by the surface and loses energy in the surface in the same manner as an incident H atom.

Measurements of H\(^+\) energy distributions at lower collision energies however do reveal a noticeable effect of incident charge state. Figure 4 shows the energy distributions of H\(^+\) resulting from 50 eV incident H\(^+\) and H. The forward direction (\(\theta=20°\)) is shown, as small angle scattering does not significantly broaden the peak. The H\(^+\) which result from incident H clearly lose more energy than do those from incident H\(^+\), a feature which has not been reported in the relevant literature. Despite the apparent shift in energy, the H\(^+\) energy distributions are very similar in shape, again suggesting that the kinetic energy losses associated with scattering and

![Fig. 2. The negative ion yield (\(\Delta \gamma\)) as a function of scattering angle (\(\theta\)) for 200 eV H\(^+\)(---) and H(----). No correction has been applied for the transmission of the analyzer.](image)

![Fig. 3. Energy distributions of negative ions formed by E\(_{in}=200\) eV H\(^+\)(---) and H(----). The vertical line marks the peak for (\(\theta=20°\)).](image)

![Fig. 4. Energy distributions of negative ions formed by 50 eV H\(^+\)(---) and H(----). Scattering Angle (\(\theta=20°\)).](image)
electronic stopping in the surface are the same for both incident H' and H. The occurrence of the shift at the high-energy limit, which is dominated by single collisions with surface atoms, verifies that this energy shift must be effectively decoupled from the kinetic losses. This implies that the shift in energy between the two incident charge states occurs prior to, or is a consequence of the neutralization process.

The location of the elastic scattering limit in the energy spectra has been plotted as a function of incident particle energy in Figure 5 for a 20° scattering angle. Also shown are nonlinear least squares fits of the data to elastic scattering behavior including an appearance potential of the form

$$\frac{E_{\text{ion}}}{E_{\text{inc}}} = R_E - \frac{E_{\text{app}}}{E_{\text{inc}}}$$

where $R_E$ and $E_{\text{app}}$ are the energy reflection coefficient and appearance potential respectively. Since the data are for elastic scattering by surface atoms, the reflection coefficient is nearly unity due to the small mass of hydrogen. The resulting shift in energy is apparently due to a difference in appearance potentials for incident H' and H. Data collected at other scattering angles exhibit this same threshold behavior. The difference is apparently constant at all energies, and measured to be $\Delta E = 4.7 \pm 0.5$ eV.

Due to the limited data both in these measurements and in the relevant literature, as well as the inherent complexity of atom-surface interactions, a detailed analysis of the effect of charge state is beyond the scope of this report. However, a simple mechanism may be the pre-acceleration of incident ions by the induced image charge. Classical calculations of the image attraction between an ion and conducting surface yield an increase in kinetic energy of

$$\Delta KE = \frac{17.4}{z(a_o)} eV$$

where $z$ denotes the distance from the surface in atomic units (a.u.). In order to reproduce the measured energy shift an incident H' must be neutralized ~ 3.7 +/- 0.4 a.u. from the surface. This simplification obviously fails for values of $z$ which are equal to atomic dimensions at the surface, so the applicability of (4) at this length scale is debatable. It is interesting however that the generally accepted mechanism for H' neutralization at a metallic surface is resonant electron transfer into an excited state, followed by Auger or radiative de-excitation. This neutralization takes place a few a.u. from the surface, in qualitative agreement with the neutralization distance estimated by ion acceleration in (4).

b. H' surface collisions

The angular distributions of scattered H' resulting from incident H' are similar to those previously shown for incident H' and H, the main difference being a consistently larger yield ($\Delta \gamma$) by approximately a factor of 2 for incident H'. Maximum yields of H' were observed to increase with decreasing collision energy, suggesting a contribution of reflected primary H' ions to the measured yield. Significant differences are
observed in the negative ion energy distributions resulting from incident H− as compared to those from H+ or H, as shown in Figure 6. In addition to the inelastic charge-exchanged peak, the appearance of a second high-energy peak (indicated by arrows in the figure) is evident at all energies investigated here. The contribution of this component to the measured negative ion yields increases significantly as collision energy decreases, and constitutes the majority of the negative ion signal at the lowest collision energies. The width of this peak is quite narrow compared to that of the inelastic peak, suggesting energy losses are due to only one or two grazing collisions. These characteristics leave little doubt that this high-energy peak is due to elastically scattered H−, which retains its initial charge throughout the surface collision(s).

Measurements of this type have previously been performed on atomically clean surfaces,12,26 and there have been no observations of H+ surviving such collisions. The relatively large contributions to the negative ion yields measured here suggest the surviving H+ is scattered by extended adsorbate layers, enabling the retention of an extra electron rather than neutralization via empty states in the conduction band. If image acceleration mechanisms indeed participate in ion-surface interactions, that effect cannot be resolved in the observed H+ scattering.

Summary

First observations of charge state effects in low energy atom-surface scattering have been reported for hydrogen impinging a gas covered copper surface. Both incident H+ and H are charge exchanged at the surface to form H+. These scattering processes both have apparent threshold energies that differ by 4.7 +/− 0.5 eV. The scattering of incident H+ shows surviving primary ions at energies up to 200 eV.

This work is supported by NASA through the Office of Space Sciences.

References

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