Electron removal from $H^{0}(n)$ in fast collisions with multiply charged ions

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The cross sections for electron removal from highly excited (n=9-24) hydrogen atoms in fast collisions with multiply charged (q=1-5) N, O, and Ar ions were investigated in an ion-atom crossed-beams experiment. The ion-atom collisions occurred inside a deflector where a moderate electrostatic field of up to 1.8 kV/cm was applied. The range of collision velocity (v_c) investigated is $v_c=1.0v_1-2.0v_1$, where $v_1=2.2\times10^8$ cm/s is the Bohr velocity. The electron-removal cross section was found to be independent of ion species for a given q and v_c , to increase as q^2 for a given v_c , and to decrease as v_c^{-2} for a given q. These q and v_c dependences of the experimental cross section are in accord with classical Coulomb ionization theories. The experimental n dependence of the cross section differs significantly from the theoretically predicted dependence, but the difference can be accounted for if we assume the presence of the external electric field in the collision volume reduces the ionization energy.

I. INTRODUCTION

Present-day plasma-fusion devices rely on injection of intense neutral hydrogen beams to increase the plasma temperature. Neutral beams, however, may cause harmful plasma instabilities if the electron loss of the beams is too large at the plasma boundary.¹ In this regard, collisions between the injected atoms and multiply charged plasma impurity ions are important because the probability for electron removal from fast H atoms increases steeply with increasing charge of the collision partner.

The two relevant processes leading to electron removal in ion-atom collisions are

 $\mathrm{H}^{0}(n) + X^{q} \rightarrow \mathrm{H}^{+} + X^{(q-1)+}$

(charge exchange) and

$$\mathbf{H}^{0}(n) + X^{q} + \rightarrow \mathbf{H}^{+} + X^{q} + e$$

(impact ionization). Here a hydrogen atom in the *n*th principal quantum state is designated as $H^{0}(n)$ and an impurity ion having charge q as X^{q+} . The charge-exchange process dominates electron removal from $H^{0}(n)$ if the collision velocity v_{c} is slower than the electron orbital velocity v_{n} , while ionization dominates if $v_{c} \gg v_{n}$; both processes contribute significantly if $v_{c} \simeq v_{n}$.² The injection energy of interest is 20-60 keV/amu, and the corresponding collision velocity range is

$$0.9v_1 \le v_c \le 1.6v_1$$

(where $v_1 = 2.2 \times 10^8$ cm/s is the Bohr velocity).

We have experimentally investigated electron removal from hydrogen atoms during fast collisions with a variety of multiply charged ions at velocities relevant to fusion devices using a colliding-beams apparatus. While our primary motivation for this investigation was to obtain information useful to fusion technology, the results obtained are of general and fundamental interest.

II. EXPERIMENTAL CONSIDERATIONS

A. General information

The measurements were performed with an ionatom crossed-beams apparatus. Because the apparatus used is the same as in our earlier report³ and a detailed description will be given elsewhere,⁴ only a brief description essential for this paper will be given.

Neutral beams used in fusion devices are obtained from energetic protons by electron-capture collisions in a neutralizing gaseous medium. The atoms thus neutralized, while predominantly in the ground state (n = 1), are also produced in various excited states. The distribution of excited-state population of a typical neutral beam is shown in Fig. 1. A beam transit time of $\sim 10^{-6}$ s is sufficient to radiatively quench the atoms formed in lower *n* states leaving only the atoms in $n \sim 8$ and higher states.⁵ The usual stray fields quench the H(2s) metastables while long-lived higher *n* states (say n > 24) are attenuated from the beam because electrons from such highly excited states are readily

1310



FIG. 1. A sketch of the *n*-state population distribution of a typical atomic hydrogen beam for fusion devices. Note that the vast majority of the atoms are in the ground state.

stripped off in the fringe magnetic field of the fusion device. In our experiment we used atomic hydrogen beams having similarly distributed excited states.

B. Neutral hydrogen beam and signal detection

A schematic layout of our apparatus is shown in Fig. 2. The atomic hydrogen beam was obtained by neutralizing momentum-analyzed energetic protons in a water-vapor neutralizer. The neutralization pressure was set for the optimum yields. Charged particles, which remained in the beam after the neutralizer, were swept out electrostatically by a pair of condenser plates placed downstream. The distribution of excited atoms in the beam was measured to be of the form n^{-3} for $n \ge 9$ and agrees with previous results.⁶⁻⁸ After collimation by a pair of slits the beam was transmitted through the intense electric field of a field ionizer. The ionizing field (E)was applied transverse to the beam direction. In this field, highly excited atoms of the beam were field ionized and attenuated from the beam. The excited-state population of the beam could be readily truncated by changing the ionizer settings. According to the semiempirical relation of Ref. 7, atoms in n > N states are ionized in the field $E(kV/cm) = 6.25 \times 10^5 N^{-4}$. We were able to apply up to E = 100 kV/cm. The N value corresponding to this maximum ionizer setting is 9.

As shown in Fig. 2, the two beams collided inside a deflector to which suitable deflection voltages were applied. These voltages were chosen so that only the H^+ originating from a small region along the neutral-beam path centered around the beam crossing were accepted by the cylindrical analyzer



FIG. 2. Schematic layout of experimental apparatus. Symbol S represents the location of beam-limiting apertures. Field applied to the H⁺ sweepers is ~ 300 V/cm. Deflector modulator consists of three parallel-plate electrodes with a rectangular hole for the ion beams. Upper electrode is split, and the proton trajectory is determined by the combined dc fields provided by the deflection voltages applied on the downstream portion of the upper electrode and the central electrode. With this arrangement, a desired deflection can be obtained for a range of suitable voltage combination. In addition to a dc deflection voltage, a sawtooth modulating voltage was applied to the deflector modulator. Proton counts were recorded in a multichannel analyzer as a function of the sawtooth voltage for a given dc deflector voltage.

and counted using a channel electron multiplier. The neutral-beam transit time across this small region was $\leq 10^{-9}$ s; consequently, only those crossed-beams events leading to electron removal with a rate commensurate with this transit time were accepted. This technique of discriminating real events from background events is a novel feature in the present arrangement of the usual crossed-beams method. The strength of the deflection field (E_d) varied linearly with the energy of the neutrals $E_{\rm H^0}$ as $E_d(\rm kV/cm) = 0.044 \ E_{\rm H^0}$ (keV).

C. Absolute cross-section determination

In this experiment we measured the number of H⁺ detected per incident atom in unit area of the neutral beam per ion encountered in the beam crossing. This number, for the simpler case of all atoms being in a single n state, is the electron-removal cross section σ_n . However, because of the distribution of excited atoms in the beam, the measured number represents a sum of electron-removal cross sections weighted by the population of relevant excited states (i.e., $Y_N = \sum_{n=1}^{N} P_n \sigma_n$, where P_n is the excited-state population). The population for $n \ge 9$ measured in an earlier experiment⁹ is $P_n = a / n^3$, where a is an experimentally determined constant which depends on incident proton energy, neutralizing medium, etc. Measurements of P_n for lower states were not accessible to the experiment, but most of the atoms which leave the neutralizer in n < 7 are expected to have decayed to the ground state by the time they reach the ion beam (drift time is $\sim 2 \times 10^{-6}$ s). Therefore, the population near n=9 is also distributed in the manner sketched in Fig. 1, and

$$Y_N \simeq (1-\beta)\sigma_1 + a \sum_{n=9}^N \sigma_{n/n^3}, \qquad (1)$$

where $\beta = a \sum_{n=9}^{N} n^{-3}$ and σ_1 is the electronremoval cross section for ground-state atoms.

III. RESULTS AND DISCUSSION

A. General considerations

Our experimental results consisted of the measurement of absolute value of Y_N and the observation of changes in Y_N effected by varying the experimentally accessible parameters N, q, v_c , and ion species. These results provide a comprehensive experimental basis for understanding the electronremoval process; however, because the quantity Y_N is a population-weighted sum of substate averaged total cross sections, the experimental results can only provide insight into those features of the electron-removal process that persist after the averaging and summing inherent in the present experiment. In interpreting our results, we used the simplest applicable theory and relied particularly on the predictions based on the classical treatment of ion-atom collisions by Bohr¹⁰ and Bohr and Lindhard.¹¹ As we shall see, the classical treatment is especially amenable to our interpretation.

According to Bohr and Lindhard, impact ionization is the dominant electron-removal mechanism if

$$(v_c/v_n)^2 \ge 2\sqrt{q}$$

This condition is satisfied by all but the groundstate (n = 1) atoms of the beam in the present collisions and, accordingly, we shall ignore the electron-capture contribution to the electron removal for $n \ge 9$. In the high-collision-velocity regime, the ionization cross section σ_{nI} is

$$\sigma_{nI} = k \left(\frac{q}{v_c}\right)^2 \left(\frac{1}{\Delta E_n}\right), \qquad (2)$$

where k is a constant and ΔE_n is the minimum energy that must be transferred impulsively during the collision for ionization to occur. For those fast collisions in which the disruption of electron binding (due to, e.g., polarization of the atom) during the collision is negligible, ΔE_n is the ionization energy (I_n) of the isolated, free atom. $I_n = R_y/n^2$ for the hydrogen atom, where R_y is 13.6 eV. Quantal theories give essentially the same q and ΔE_n dependences for ionization, although the velocity dependence is slightly different¹²:

$$\sim \left[\frac{1}{v_c}\right]^2 \ln(v_c).$$

B. n dependence

The population-weighted cross sections Y_N for the H⁰(n) + N³⁺ collisions measured at $v_c = 1.0 v_1$ and 1.3 v_1 as a function of N are shown in Fig. 3. The $v_c = 1.3 v_1$ results are from an earlier report.³ The error bars shown are our estimate of random errors. The absolute value are subject to $\pm 20\%$ additional error. Excited atom with $n \ge 9$ for both cases contribute less than 0.5% of the beam intensity, yet they give rise to a significant fraction of the H⁺ detected. The average electron-removal cross



FIG. 3. Population-weighted electron-removal cross sections for the $H^0(n) + N^{3+}$ system at $v_c = 1.0 v_1$ and 1.3 v_1 . Error bars accompanying the data represent the random uncertainty. Curves are explained in the text.

sections for the excited atoms

$$\overline{\sigma}_n = \sum_{n=9}^{24} P_n \sigma_n / \sum_{n=9}^{24} P_n$$
(3)

are enormous:

$$(7.3\pm1.5)\times10^{-12}$$
 cm²

at $v_c = 1.3 v_1$ and

$$(1.8+0.4)\times 10^{-12}$$
 cm²

at $v_c = 1.0 v_1$.

The theoretical shape of the weighted cross section (*N* dependence) can be readily obtained by using Eq. (2) and the empirical relation $P_n = an^{-3}$. Letting y_N stand for the theoretical counterpart of Y_N ,

$$y_N = y_8 + ka \left[\frac{q}{v_c}\right]^2 \sum_{n=9}^N \left[n^3 \Delta E_n\right]^{-1},$$
 (4)

where y_8 includes contributions up to n=8. If $\Delta E_n = R_y / n^2$ (the ionization energy of an isolated atom), then Eq. (4) gives a particularly simple shape: N dependence is given by a simple sum $\sum_{n=9}^{N} (1/n)$. However, the experimentally observed shapes may be more complex due to the moderate

electrostatic deflection field (~1.8 kV/cm) that was needed for the selective detection of signals. The collisions thus occurred in the presence of an external field, and $\Delta E_n = R_y/n^2$ may not be valid. All atomic levels acquire an ionization probability when placed in an electric field, and the higher the level of excitation the larger the probability becomes. For sufficiently high-lying levels, their ionization width exceeds the level spacing and the levels merge into a continuum.¹³ The onset of this continuum occurs below the zero-energy field-free level that marks the ionization limit for an isolated atom, as demonstrated by the recent photoionization experiment of Rb atoms in high Rydberg states ($n \sim 20$) placed in moderate electrostatic fields.¹⁴

Using this line of reasoning, we analyzed the experimental results using Eq. (4) and taking $\Delta E_n = I_n - \delta$, where δ is the amount by which the continuum energy is assumed lowered, in analogy with the photoionization results. The solid curves shown in Fig. 3 represent best fits to the observed Ndependence, based on the formulation of Eq. (4) and the above modification in ΔE_n with δ as the fitting parameter. The values of n_{δ} , where $\delta = R_{\nu}/n_{\delta}^{-2}$, giving these solid curves are 24.7 ($v_c = 1.3 v_1$) and 28.0 ($v_c = 1.0 v_1$). A small deviation (±5%) from these values of n_{δ} gave visibly poorer fits, especially for the $v_c = 1.3 v_1$ case for which Eq. (4) has a singularity at $n_{\delta} = 24.0$. The calculated values y_{24} y_9 were normalized to the corresponding experimental results for convenience. Similarly normalized theoretical curves for $\delta = 0$ are also shown by the broken curves for comparison. As can be seen, they represent the data poorly.

As shown in Table I, the amounts of energy lowering needed to fit the data are rather close to the minimum binding energies calculated using the semiempirical formula for field ionization. This suggests a possible close connection between the field ionization and impact ionization in an applied field. In this regard Olson's recent suggestion¹⁵ provides an interesting and informative link: He regards the electron-removal cross section in the presence of the applied field to consist of the ionization

TABLE I. Energy lowering and minimum binding energies for collision velocities.

Collision velocity v_c (units of v_1)	Deflector strength E_d (kV/cm)	$n_{\delta}(E_d)$	$n_f(E_d)^{\mathrm{a}}$
1.0	1.10	28.0	27.3
1.3	1.76	24.7	24.3

^aValues calculated using the semiempirical relation given in Ref. 7. cross section (σ_{nI}) for free atoms and excitation cross sections to all high-lying bound states $(\sigma_{nn'})$ which subsequently field ionize, i.e.,

$$\sigma_n = \sigma_{nI} + \sum_{n'=n_f}^{\infty} \sigma_{nn'} \; .$$

As shown below,¹⁶ Olson's way of viewing the presently studied collisions can lead to Eq. (2) with

$$\Delta E_n = R_y \left[\frac{1}{n^2} - \frac{1}{n_f^2} \right]$$

according to the classical theory.

C. q dependence

An assumption implicit in the theory that gives a $(\Delta E_n)^{-1}$ dependence of the ionization cross section is that the needed energy transfers occur impulsively through the Coulomb field during collisions. As such, the cross section must scale as the square of electric charge of the ion increases [see Eq. (2)]. To the extent that the multiply charged ions can be regarded structureless, we naturally expect the electron-removal cross sections to scale as q^2 for excited atoms in the beam for which $v_c \gg v_n$. In order to experimentally check the validity of this simple but very basic scaling law, we examined the qdependence of the weighted cross-section difference

$$\sigma_{NN'} = Y_N - Y_{N'} ,$$

where $N > N' \ge 9$. By taking differences, selected bands of excited states could be isolated for which the condition $v_c \gg v_n$ is satisfied. Figure 4 shows $\sigma_{NN'}$ as a function of q for the $H^0(n) + N^{q+}$ system for two bands of excited states. As is evident in Fig. 4 the q^2 -scaling prediction agrees with the data very well.

The observed values of $\sigma_{NN'}$ of highly excited hydrogen atoms in a given *n* band in collision with different ion species cluster together for a given *q* and v_c . This is shown in Fig. 5, where the experimental values of $\sigma_{NN'} = Y_{24} - Y_{17}$ for N^{q+} , O^{q+} , and Ar^{q+} ions in common arbitrary units are plotted versus *q* on a log-log scale (N^{q+} results from Fig. 4 are replotted here). The error bars associated with these $\sigma_{NN'}$ are $\leq 40\%$. This clustering indicates that the cross section for electron removal from a highly excited *n* band is independent (or nearly independent) of ion species for a given *q* and v_c . Furthermore, the excellent agreement between the data and the theory, illustrated in Fig. 5, indi-



cates that the q^2 -scaling law represent O- and Arion results very well also.

D. v_c dependence

The velocity dependence of the cross sections averaged over the population of a narrow band of excited states can be inferred from the Y_N measured at different velocities. The $\langle \sigma_n \rangle$ for the n=10-14band for the $H^0(n) + N^{3+}$ system at three velocities, deduced from the measured values of Y_{14} and Y_{10} , is shown in Fig. 6. The rather large error bars shown reflect the fact that errors from the population (P_n) , in addition to the Y_n errors, are propagated onto

$$\langle \sigma_n \rangle = (Y_{14} - Y_{10}) / \sum_{10}^{14} P_n$$
.

Also shown in this figure are the v_c^{-2} and v_c^{-2} ln (v_c) dependences predicted by the classical and quantal theories, respectively. The present results are consistent with either of these high-velocity collision regime predictions.





FIG. 5. Electron-removal cross sections for hydrogen atoms in the (n=17-24) band in collision with multiply charged ions. N^{*q*+} results of Fig. 4 are replotted here. Error bars ($\leq \pm 40\%$) are omitted from this figure to avoid cluttering. Solid line shown gives the q^2 dependence of $\sigma_{NN'}$.

E. Ground-state cross section

The absolute cross sections (Y_N) measured by crossing an $H^{0}(n)$ beam obtained by neutralizing H⁻ ions in a thick water-vapor neutralizer with a N^{3+} beam at $v_c = 1.0 v_1$ are shown in Fig. 7. For comparison, the yields for the $H^{0}(n)$ beam obtained from H^+ ions at the same v_c are also shown. Although the fraction of excited atoms in the $H^{0}(n)$ beam obtained from the neutralization of H⁻ ions was measured to be about half that obtained from H⁺ ions, the population of excited atoms was observed to have similar distributions in both beams (i.e., $P_n = 0.16/n^3$ for the H⁻ case and $P_n = 0.28/n^3$ for the H⁺ case). Given these populationmeasurement results, the convergence of the two population-weighted cross sections, shown in Fig. 7, suggests that at the investigated velocity, at least for N < 9, the ground-state contribution dominates the sum $Y_N = \sum_{1}^{N} P_n \sigma_n$. The electron-removal cross section σ_1 for the H⁰(1) + N³⁺ system at $v_c = 1.0 v_1$



FIG. 6. Electron-removal cross sections for hydrogen atoms in the (n=10-14) band in collision with N³⁺ ions at different v_c . Error bars shown represent random error (see text for detail). Theoretical curves shown indicate the v_c dependence given by the classical theories (solid line) and quantal theories (broken line).

obtained¹⁷ from the two sets of absolute cross sections Y(n) shown in Fig. 7 is

$$\sigma_1 = (3.0 \pm 1.4) \times 10^{-15} \text{ cm}^2$$
.



FIG. 7. Population-weighted electron-removal cross sections from hydrogen beams prepared by two different methods: Electron capture by energetic protons (open circles) and electron stripping from energetic H^- ions (closed circles). Y_N given by the open circles are the same ones shown on Fig. 3. See text for explanation of the broken lines.

The rather large uncertainty for the σ_1 reflects the reality that a number of independent errors are propagated onto the value of σ_1 , but does not include, e.g., the possibility that the two Rydberg populations have different (l,m) substate distributions, and consequently different radiative lifetimes and field ionization thresholds for the same principle quantum number n. The inferred value for the ground-state electron-removal cross section is somewhat larger than the sum of cross sections for charge exchange and impact ionization

$$[(1.7+0.25)\times 10^{-15} \text{ cm}^2]$$

measured at similar velocities for the same collision system.^{18,19}

IV. CONCLUSION

Assuming allowance is made for the fact that the collisions occur in the presence of an externally applied electric field, the present results are consistent with the $(q/v_c)^2 (1/\Delta E)$ scaling. This indicates that the dominant cause for electron removal from excited hydrogen atoms colliding with heavy ions is Coulomb ionization for $v_c \ge v_1$. The universal factor (σ_0) of the Coulomb ionization cross section (σ_{CI}) , where

$$\sigma_{\rm CI} = \sigma_0 q^2 \left[\frac{v_1}{v_c} \right]^2 \left[\frac{I_1}{\Delta E} \right]$$
(5)

[deduced from the present $H^0(n) + N^{3+}$ collision study] is $(13\pm4)\pi a_0^2$, where πa_0^2 is the geometric cross section of the ground-state hydrogen atom. Theoretical values of $(20/3)\pi a_0^2$ and $6\pi a_0^2$ given by Richards and Percival²⁰ and Olson,²¹ respectively, are smaller than the experimental value.

Fusion plasmas are confined in strong magnetic fields, and, as the injected beam reaches the plasma, the atoms experience an electric field (motionally induced) as well as a magnetic field. We studied the effect of an external electric field on the cross section for electron removal from hydrogen atoms in fast collision with a variety of multiply charged heavy ions, but not the effect of a magnetic field. Because the state of an atom (especially a Rydberg atom) in such a strong magnetic field undergoes severe mixing with an attendant energy change²² (which is quite different from the mixing in an electric field), the relevant electron-removal process may be quite different from the case presently studied. A similarly comprehensive study of the electron-removal process in strong magnetic fields would provide valuable complementary information.

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$$\int_{I_n-I_n}^{I_n} \frac{d\sigma_n}{dn'} \rho_{n'} dE ,$$

where $\rho_{n'}$ is the level density and $I_{n_f} = R_y/n_f^2$. Adding this contribution to the classical ionization cross section one obtains

$$\int_{I_n}^{E_m} \frac{d\sigma_n}{dE} dE + \int_{I_n-I_n}^{I_n} \frac{d\sigma_n}{dn'} \rho_n dE = \int_{I_n-I_n}^{E_m} \frac{d\sigma_n}{dE} dE ,$$

which shows an apparent lowering of the ionization en-

ergy from I_n to I_n - I_{n_f} . The quantity E_m in the above expression is the maximum value of energy transferred in the collision.

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