Electron Loss from Highly Excited States of H^0 in Collisions with N^{3+}

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Cross sections for electron loss from excited hydrogen atoms in collision with a N^{3+} beam at 40-keV/amu collision energy were determined by use of a crossed-beams experiment. The value of the principal quantum number, n, of the excited atoms investigated ranged from 9 to 24. In this range the electron-loss cross section increases as $\sim n^3$, in contrast to the n^2 increase predicted by a number of theories.

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In this Letter we present the first experimental observation of the n dependence of the cross section for electron loss from highly excited hydrogen atoms ($9 \leq n \leq 24$) in fast collisions with multiply charged ions. The collision velocity of the present experiment was 2.8×10^8 cm/s, substantially higher than the range of velocities covered in previously reported experiments[1, 2] involving Rydberg collisions. The study of ionization of Rydberg states of atomic hydrogen by heavy particles is not only of basic interest, but it is also of use in applied research, such as the study of neutral-beam–injection heating and refueling of magnetically confined fusion plasmas.[3]

The measurements were performed with an ion-atom crossed-beams apparatus. Since a detailed description will appear in a forthcoming paper, only a brief account of the experiment will be given. Shown in Fig. 1 is a schematic diagram of the experimental arrangement. We use a N^{3+} beam (typically $\sim 6\times10^{12}$ particles/s) from the Oak Ridge National Laboratory Multicharged Ion Facility and a fast H^0 beam ($\sim 2\times10^{12}$ particles/s) produced in a H_2O neutralizer cell from 40-keV protons. The two beams are crossed at 90° in a deflector-modulator where two separate dc electric fields are applied transverse to the H^0 beam. Each proton resulting from a collision between a H^0 atom and either a residual gas atom or a N^{3+} ion is thus labeled by a unique deflection (trajectory) determined by its point of formation, permitting selective detection of only those protons produced near the interaction region. An application of between 1- and 1.5-kV/cm field at the beam-crossing region (and correspondingly adjusted field immediately downstream) provided necessary deflection of 40-keV protons (see Fig. 1). The yield of proton was independent of the ap-

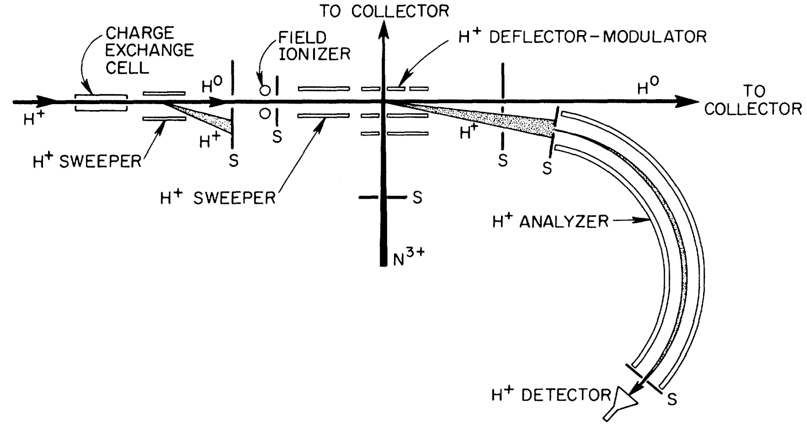

FIG. 1. Schematic layout of experimental apparatus. The symbol S represents the location of beam-limiting apertures. The field applied to the H^+ sweepers is ~ 300 V/cm. The deflector-modulator consists of three parallel-plate electrodes with a rectangular hole for the N^{3+} beam. The upper electrode 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.

plied field strength in this range. The N^{3+} beam was chopped to further facilitate separation of signal and background events.

Because the electron-capture process results in a H^0 beam with a distribution of excited n states, the observed proton counts are related to the population-weighted sum of electron-loss cross sections $\gamma_N = \sum_1^N P_n \sigma_n$, where P_n and σ_n are, respectively, the population and electron-loss cross section of H^0 (principal quantum number = $=\eta$), and N is the largest value of n in the H^0 beam, as selected by an electrostatic field ionizer upstream of the beam crossing region. With use of the empirical relation[4] $\mathcal{E}$ (kV/cm) $= 6.25 \times 10^5 N^{-4}$, values of N in the range $9 \leqslant N \leqslant 24$ could be selected by appropriate electric field settings of the ionizer. Two additional data points were obtained with use of $\vec{v} \times \vec{B}$ equivalent electric fields. The observed proton counts were converted to weighted cross sections via standard crossed-beams analysis.[5]

In Fig. 2 the measured weighted electron-loss cross sections for collisions of 40-keV $H^0(n)$ with 30-keV N^{3+} are shown as a function of N; also shown are the corresponding field strengths of the ionizer necessary to attain the given N values. The error bars indicate statistical uncertainties at one standard deviation. The quantity Y_N increases (on a log-log plot) nearly linearly with N, suggesting an N dependence of the form $Y_N = CN^{\alpha}$. A least-squares fit of the data in the range $9 \leqslant N \leqslant 24$ yields

$$Y_N = 0.41 \times 10^{-15} N^{1.12} \text{ cm}^2. \tag{1}$$

In an auxiliary experiment,[6] we have determined the population of excited states P_n in the 40-keV H^0 beam (in the range $9 \leqslant n \leqslant 24$) to be

$$P_n = 0.41 n^{-3}, \tag{2}$$

which agrees with similar previous measurements.[4,7] In obtaining this result [Eq. (2)], we assumed, following Barnett, Ray, and Russek[7] and others,[8] that n was a continuous variable in view of the Stark broadening of the levels. Since both Y_N and P_n are well described by power-law dependences, it is reasonable to expect a similar dependence of σ_n on n. With use of the definition of Y_N together with Eqs. (1) and (2), we find

$$\sigma_n = 1.19 \times 10^{-15} n^{3.12} \text{ cm}^2 \quad (9 \leqslant n \leqslant 24). \tag{3}$$

The cross section σ_n is nearly as large as the "geometric" cross section $\pi a_n^2 \simeq 10^{-16} n^4$ cm^2 for the range of investigated excited states $9 \leqslant n \leqslant 24$. We estimate the propagated uncertainty in the magnitude of σ_n to be ~20%.

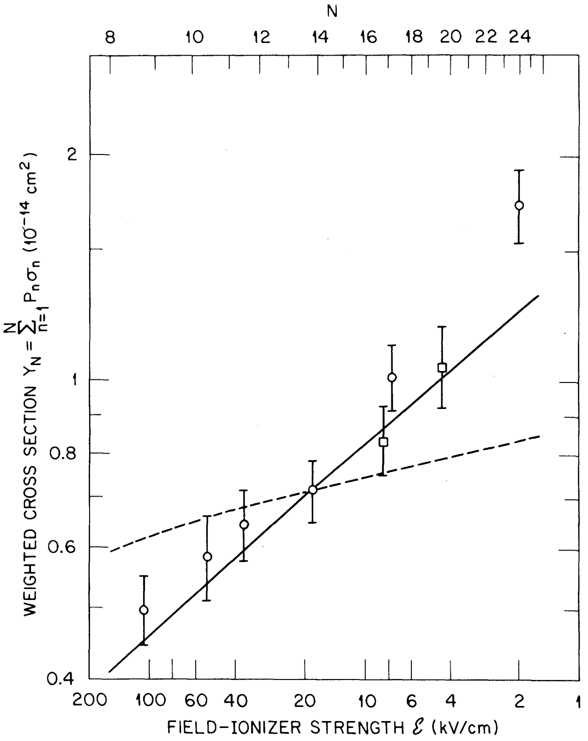

FIG. 2. The population-weighted cross section $Y_N = \Sigma_1^N P_n \sigma_n$ as a function of N, the upper cutoff of P_n, selected by field ionizer. The round points were obtained by electrostatic field ionization and the square points by magnetic field ionization. The theoretical results based on n^2 scaling, normalized to the data at $N = 14$, are shown by the dashed line. The solid line shows similarly normalized least-squares results. (See text for details.)

The collision velocity in the present investigation exceeds relevant electron orbital velocities by at least an order of magnitude. For such fast collisions, electron loss is dominated by impact ionization,[9] and in the following comparison with theory, we take the observed electron-loss cross section as being due entirely to impact ionization and neglect the contribution due to electron transfer collisions.

For collision velocity v_c exceeding the electron orbital velocities v_e, classical theories of ionization become applicable. Consequently, the present collisions should be describable by classical theories since $v_c > 10 v_e$. The binary-collision theory of Garcia, Gerjuoy, and Welker[10] predicts ionization cross sections to increase as n^2. Using arguments based on the law of (classical) mechanical similarity, Percival and Richards[11] also give an n^2 scaling, as do recent numerical results[12] employing the classical-trajectory Monte Carlo method. A similarly general n-scaling law based on quantum mechanical theories (e.g., the Born approximation) applicable for the present cases is not available in the literature, but the numerical results of Matsuzawa's first-order Born calculation[13] for the hydrogen atom ($n = 10$ and 20) in collision with a fast electron show the ionization cross section increasing as n^2. Because the projectile dependence in first-order Born theory[14] appears as an external scaling factor $(q/v_c)^2$, where q is the ionic charge of the projectile, we infer that this theory predicts an n^2 dependence for the present cases also. In Fig. 2 the population-weighted cross section Y_N predicted by this n^2 dependence after normalization at $N = 14$ (dashed line) are compared with the measured values. As can be seen, the theory represents the data poorly. This poor representation may, in part, arise from the perturbation of excited atoms by the trajectory-defining electrostatic field applied in the collision region. However, our earlier stated observation that the proton yield did not change with the applied field strength suggests the observed n dependence is not significantly affected by the applied field.

Besides the condition that the collision be fast, implicit in these fast-collision theories giving n^2 dependence, and particularly in the Born theory, is the assumption that the electron wave function for the initial or final state of the target atom is undistorted by the passage of the projectile ion. A very important and tangible consequence of this assumption is that in the first-order Born theory of ionization, an ionized electron is in the continuum state of the resulting H^+; The theory cannot accommodate the possibility of the electron's being "dragged" away by the incident N^{3+}, or, alternatively, being captured into the continuum state of $N^{3+} + e^-$. Thus if the electron is dragged away in a significant fraction of the collisions, the Born theory is not applicable. Existing atom-ion collision data[15] show that the capture into continuum is a significant, if not dominant, mode of ionization for fast heavy-ion collisions. Based on these data we expect the continuum capture to be significant for the present collisions also. In fact, according to Shakeshaft's calculations[16] nearly 30% of ionization for the $H^0(n = 1) + H^+$ collision at our collision velocity results from the continuum capture.

The assumption of being free of distortion can be fulfilled if the electrostatic projectile-electron potential energy is small compared with the electron binding energy. For the present case this assumption is satisfied for those collisions having impact parameter $b > 2qa_n$, or $b = 6$ atomic radii. For smaller-impact-parameter collisions the distortion may be too severe for the Born theory, and the classical theories, to be valid. Earlier, Basbas *et al.*[17] and others[18] ascribed similar distortion and subsequent polarization of target atoms as the cause for the anomalous projectile-charge dependence of inner-shell ionization cross sections measured for a variety of targets by fast ions.

Recently, Chibisov[19] and Grozdanov and Janev[20] advanced a quite different view of the electron-loss process. In this view the target electron leaks out of the atom by tunneling through the barrier formed by the charges of target nucleus and projectile during the collision. Since this is an adiabatic charge-exchange theory, the importance of tunneling contribution to the present (fast) cases is difficult to assess. If applicable, the tunneling theory gives cross sections that agree well: The magnitude as well as the n dependence agrees substantially with the experiment.

In summary, experimental electron-loss cross sections σ_n of Rydberg states of hydrogen atom in fast collision with N^{3+} ions, as deduced from a $H^0(n)$-N^{3+} crossed-beams experiment, show σ_n to increase as $\sim n^3$. This is in marked contradiction to an n^2 dependence predicted by a number of theories. A plausible reason for this contradiction is the inability of these theories to account for the distortion of electron wave functions in the field of ionizing N^{3+} ion during the collision. We conclude that the classical as well as the first-order Born theory of ionization may not be adequate to describe ionization of loosely bound electrons in Rydberg states by multiplying charged ion impact.

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Direct Detection of the Product Vibrational-State Distribution in the Associative Detachment Reaction $Cl^- + H \rightarrow HCl(v) + e$

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The initial product vibrational-state distribution is obtained for the thermal associative-detachment reaction, $Cl^- + H \rightarrow HCl(v = 0, 1, 2) + e$, produced by infrared chemiluminescence in a flowing afterglow. The ratio of HCl($v = 2$) to HCl($v = 1$) population formed in the reaction is $N_{v=2}/N_{v=1} = 0.60 \pm 0.03$. Comparison of the total emission intensity to that from a reaction of similar exothermicity suggests that HCl($v = 0$) formation may be small.

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There is tremendous interest at present[1] in electron-scattering,[2,3] dissociative-attachment,[2,4-6] and associative-detachment[7,8] processes in HCl. The interpretation of the HCl$^-$ negative-ion states, through which all of these processes must pass, is of major importance.[1,9] A remaining problem associated with these HCl$^-$ states is the product vibrational-state distribution of the associative-detachment process: $Cl^- + H \rightarrow HCl(v) + e$, $\Delta H = -0.82$ eV. Previous experimental studies of this associative-detachment process were carried out in a flowing-afterglow[7] and flow-drift apparatus.[8] A near-Langevin rate constant ($k = 9.6 \times 10^{-10}$ cm^3 molecule^{-1} sec^{-1}) is obtained at thermal energies.[7] Preliminary drift-velocity studies show only a slight decrease in the rate coefficient up to 0.2 eV kinetic energy, indicating an attractive HCl$^-$ potential in the autodetaching region.[8] There are no investigations of the product vibrational-state distribution.

The most important results for vibrational distribution in the products of other associative-detachment reactions have been obtained by energy analysis of the detached electrons.[10] Here