Electron Loss in Low-Energy H^+-H(high n) Merged-Beam Collisions*

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We have measured the electron-loss cross section for collisions between protons and highly excited hydrogen atoms having principal quantum numbers in the nominal n band $44 \lesssim n \lesssim 50$. The c.m. collision energy range was $0.4\ \text{eV} \leq W \leq 61$ eV. Theoretical ionization-cross-section results are compared with the data and are then used to unfold from the data the approximate electron-transfer cross section. Classical scaling calculations are found to be in reasonable agreement with the unfolded data while quantal predictions are not.

The comparison of experiment and theory for H^+-H(n) collisions leading to electron loss, the sum of electron transfer and ionization, has been limited to the one restricted case of principal quantum number $n=1$.[1] Although theory exists, the n dependence of the electron-loss cross section has never been measured in any collision energy range, either in this most fundamental ion-atom system or in any other.

The electron-transfer and ionization cross sections for low-energy (eV energy range) H^+-H(high n) collisions are among those required for evaluation of the important role that electron-capture and -loss collisions involving excited states may have in affecting the transport properties of plasmas.[2] Astrophysically these collisions are believed to be important in the ionized-hydrogen (H II) regions in interstellar space.[3]

It is the purpose of this Letter to report results of the first experimental study of H^+-H(high n) collisions. The merged-beam electron-loss cross-section data cover the center-of-mass collision-energy range $0.4\ \text{eV} \leq W \leq 61$ eV; the H(high n) target atoms had quantum numbers n in the nominal n band $44 \lesssim n \lesssim 50$.

The transition from "low"- to "high"-energy behavior of electron-transfer and ionization cross sections in ion-atom collisions occurs when the velocity v_r of relative motion of the nuclei is comparable with the orbital velocity v_n of the electron taking part in the collision. For H^+-H(n) collisions, $v_r \simeq v_n$ when $W \simeq 1.3\times 10^4 n^{-2}$ eV. For $n=1$, this occurs at $W \simeq 13$ keV; for $n=47$, at $W \simeq 6$ eV. The cross-section data reported in this Letter cover both the low- and high-energy regions.

The measurements were performed with a new, single-ion-source, merged-beam apparatus incorporating significant technological advances. The method is based on that introduced by Belyaev, Brezhnev, and Erastov.[4] Part of the apparatus is shown in Fig. 1. A pure H^+ beam with energy E_B near 11 keV and energy spread measured to be $\lesssim \pm 20$ eV around E_B was generated with an rf ion source, accelerator, and mass analyzer system not shown in Fig. 1. The neu-

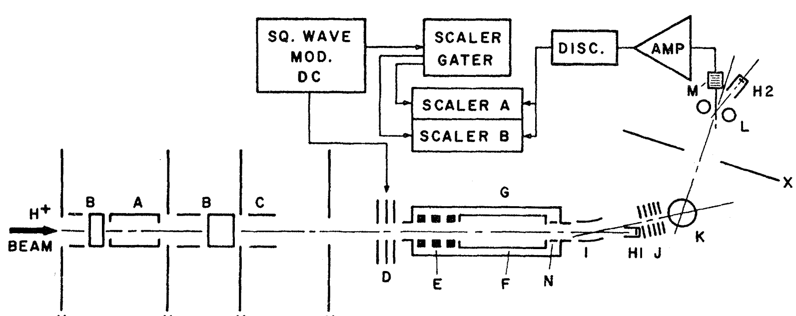

FIG. 1. The merged-beam apparatus: A, electron-transfer gas cell; B, I, L, electrostatic deflection; C, ion-beam gating plates; D, n-band–defining electric field; E, N, electric-potential change rings; F, voltage-labeled interaction region; G, liquid-He cryopump; $H1$, $H2$, Faraday cups; J, electrostatic filter lens [H. D. Zeman, K. Jost, and S. Gilad, Rev. Sci. Instrum. 42, 485 (1971)]; K, deflection magnet; M, particle multiplier; X, differential pumping apertures.

tral H(n) beam was produced by electron-transfer collisions $H^+ + Xe \rightarrow H(n) + Xe^+$ in scattering cell A.

Signals caused by atoms in a fairly narrow band of n values were defined[5] by passing both the H^+ and H(n) beams through the axial electric field modulated at about 1 kHz in lens D. Electric fields $F = 105$ and 171 V/cm defined the nominal n band $44 \lesssim n \lesssim 50$ in the present experiment. In the remainder of this Letter we characterize the n band by its approximate central n value, $n = 47$. The spread in n values affects the comparison of theory with experiment only fractionally.

The fraction of the total neutral-atom flux at energy E_B that was in the present n band was known to about ±15% from the results of measurements similar to those described in Ref. 5. Those measurements were directly applicable to the present experiment since the same apparatus was used in each case.

The H^+ and H(n) beams, both essentially at energy E_B, entered a liquid-helium–cooled cryopump (G) in which the background pressure was measured to be $\lesssim 10^{-11}$ Torr. The H^+ beam energy was decreased in a nearly uniform (to 20%) axial electric field (E) which was too weak to ionize H(high n) in the n band. Merged-beam collisions then took place between H^+ at lab energy $E_B - eV_{\rm int}$ and H(high n) at lab energy E_B inside F, a 50-cm-long interaction region kept at an electric potential $+V_{\rm int}$ with respect to ground. Any hydrogen atoms at energy E_B that were ionized inside the interaction region by merged-beam electron-loss collisions (or by ionization collisions with background gas molecules) were accelerated at the exit end (N) of the interaction region to a higher "labeled" energy $E_L = E_B + eV_{\rm int}$. Charge, energy, and momentum analyses at I, J, and K, respectively, selected for detection at M the energy-labeled H^+ flux about 10^{-9} as intense as the primary H^+ beam.

The c.m. collision energy W was varied by changing *both* E_B and $V_{\rm int}$ such that E_L was kept constant. Thus the analyzers did not have to be retuned at each W. The transmission of the combined analyzer apparatus was measured to be typically 0.93.

Both the H^+ and H(n) fluxes were collimated to beams with 3.0-mrad pencil angles by apertures located *after* the scattering cell and before the interaction region. This ensured that both beams occupied very nearly the same phase space independent of scattering angles at A. The overlap integral[6] for the merged beams was calculated with use of this assumption.

Spatially varying electric fields, such as those caused by stray, motional, or fringe fields inside the apparatus, were seen by the H(high n) in their rest frames as time-varying electric fields. Calculations based on matrix elements presented by Hiskes and Tarter[7] indicate that these fields induced Stark substate transitions that may have caused all n^2 substates (ignoring spin) of each n level in the present n band to be about equally populated.

The n-band–dependent merged-beam signal was registered as the difference in count rates recorded by scalars A and B in Fig. 1. Each was gated on for opposite half-cycles of the n-band modulation wave form while both were gated off during switching transients when conditions were undefined. The measured sum of all n-band–dependent backgrounds and noise was typically only 5% to 15% of the merged-beam signal, mainly because of the very low background gas density inside the cryopump.

The results of the present electron-loss cross-section measurements are shown in Fig. 2. The absolute cross-section scale is uncertain by a factor of about 2, chiefly because of uncertainty in the determination of the overlap integral. The error bars show the estimated relative error in the cross-section data at different values of W. The gargantuan size of the atoms, an electron orbital radius $\langle r \rangle \simeq 1200$ Å for $n \simeq 47$, is reflected in the magnitude of the cross-section scale, 10^{-9} cm^2.

Abrines and Percival[9] have indicated that with-

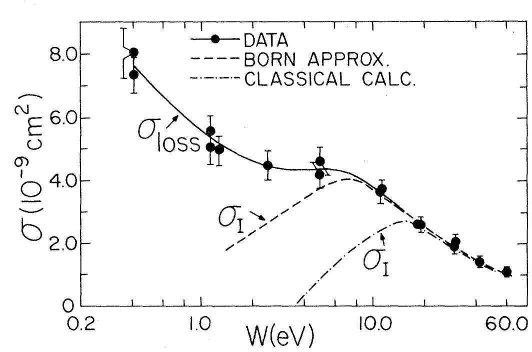

FIG. 2. The measured electron-loss cross section $\sigma_{\rm loss}$ for collisions between H^+ and an atom H(high n) in the nominal n band $44 \lesssim n \lesssim 50$. A smooth curve has been drawn through the data points. The cross-section scale is uncertain by about a factor of 2. The theoretical $n = 47$ Born (Ref. 8) and classical Monte Carlo (Refs. 9 and 10) ionization cross sections σ_I that are shown have been classically scaled from $n = 1$ and multiplied by an additional factor 3.48.

in the classical approximation, classical mechanics scaling rules may be used to express electron-transfer and ionization cross sections for H^+-H(n') collisions in terms of those for H^+-H(n) collisions: The cross section is scaled as $(n'/n)^4$ if the collision energy is scaled as $(n'/n)^{-2}$. The validity of these rules has not been experimentally tested.

In Fig. 2 we show two different theoretical ionization curves for $n=1$ classically scaled to $n=47$. They have been multiplied by an additional factor 3.48. For $W \gtrsim 8$ eV, the shape of the scaled Born-approximation curve[8] agrees well with the data, as does the classical Monte Carlo curve[9,10] for $W \gtrsim 20$ eV.

We point out that the H^+-H(high n) electron-transfer cross section furnishes a sensitive test of the n dependence of theory. It has been experimentally and theoretically established that low-energy resonant-electron-transfer cross sections σ_{10} for tightly bound atomic targets satisfy the relation $\sigma_{10}^{1/2} = A - B \ln W$, where A and B are constants dependent on the particular ion-atom pair *and* electronic state involved.[11] For H^+-H(n) collisions, the approximate quantal calculations of Rapp and Francis[11] (all n) and Bates and Reid[12] ($n \leq 5$) indicate that the slope B scales with the first power of n. Smirnov, however, has criticized the two-state approach used in these calculations when applied to weakly bound, excited target atoms and has emphasized the importance of barrierless electron transitions.[13] Furthermore, we note that classical scaling predicts a slope B proportional to n^2. For $n \gg 1$ there is obviously a dramatic difference between classical and quantal predictions.

It is important, therefore, to attempt to unfold the electron-transfer cross section as a function of W by subtracting the ionization contribution from the present electron-loss data. In Fig. 3 we show the results for $\sigma_{10}^{1/2}$ for $W \lesssim 20$ eV obtained by using for subtraction both the classically scaled Born and Monte Carlo curves shown in Fig. 1. Each curve, EX1 and EX2, respectively, yields an approximately constant slope for the low-energy range and a shoulder in the region $W = 7$ to 10 eV. It is reasonable to believe that the actual electron-transfer slope is bounded by these two curves.

For comparison we show some theoretical electron-transfer predictions for $n=47$: SM, the classical results of Smirnov[13]; MC, the classical Monte Carlo results of Abrines and Percival[9] and Banks[10]; and RF, the quantal results of Rapp and Francis.[11] We also show SD, the results of classically scaling experimental $n=1$ data to $n=47$.[1]

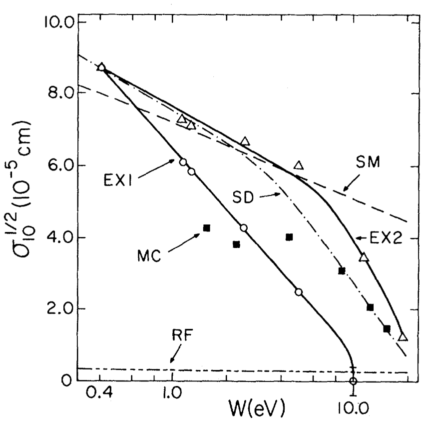

FIG. 3. The electron-transfer cross section σ_{10} obtained by subtracting from the measured $\sigma_{1\mathrm{oss}}$ data the σ_I values shown in Fig. 2: EX1 (circles), Born σ_I subtraction; EX2 (triangles), Monte Carlo σ_I subtraction. Smooth curves have been drawn to aid the eye. Some theoretical σ_{10} results for $n=47$: SM, Ref. 13; MC (squares) Refs. 9 and 10; RF, Ref. 11. Experimental $n=1$ σ_{10} data of Ref. 1, classically scaled to $n=47$: curve SD.

Curves SM, MC, and SD are clearly consistent in magnitude and in slope with the present unfolded electron-transfer data, although curve SM has a different high-energy behavior. Curve RF yields a cross section with a slope B a factor of about 100 smaller. We are aware of no quantal electron-transfer calculations for $n=47$ in agreement with the present results. In fact, we have found that the quantal results of Smirnov[14] yield an equation for σ_{10} that breaks down in our energy range (yields no solutions) for many substates of $n=47$. We conclude that electron transfer in low-energy H^+-H(high n) collisions is not well understood quantum mechanically.

In future experiments we hope to be able experimentally to separate electron-transfer signals from ionization signals in order to test more precisely the n dependence of the slope B. The present results indicate that low-energy e^--H(high n) merged-beam collision studies should also be feasible.

In conclusion we emphasize that in the present experiment the keV-energy-range H(high n)-production cross section scaled as n^{-3} whereas the eV-energy-range electron-loss cross section scaled as n^4. This produced signals which scaled as n. Therefore the neglect of the effects of highly excited atoms in some experiments, because of presumed inefficiency of production, could lead to serious error.

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Nature of the Molecular Alignment in a Smectic-*H* Phase

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Quasielastic neutron scattering experiments on terephthal-*bis*-butylaniline (TBBA) show that the phenyl rings do not have a strongly preferred orientation in the smectic-*H* (tilted *B*) phase. This suggests that the Meyer-McMillan model is not quite adequate for this particular system.

Recently, Meyer and McMillan[1] extended to smectic-*B* and -*H* phases a molecular theory[2] of smectic-*C* and -*A* phases, by including a softcore repulsive interaction in addition to the dipole-dipole interaction used previously. In this theory, this dipole-dipole interaction plays an essential role and is responsible for the tilted character of the smectic-*C* and -*H* phases by orientational ordering of the dipoles. An attempt to confirm the orientational order experimentally was recently made by NMR[3] on a series of smectic compounds. It was found that, while some smectic-*H* phases [in particular that of terephthal-*bis*-butylaniline (TBBA)] are consistent with the idea of a partial freezing out of rotation about the long axis, no freezing of this rotation could be observed in smectic-*C* phases.[4] For similar reasons we have recently studied[5] the smectic-*H* phase of TBBA (in Ref. 5 this was simply called the smectic-*B* phase; other names have been used[6]) using high-resolution quasielastic neutron scattering. The data were found to be consistent with the idea of rapid, uniaxial rotational diffusion of the molecules about their long axis, implying a lack of orientational order. In this Letter, we extend our interpretation of the same