Electron capture in $H^+ + N_2$ collisions

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Electron capture in $H^+ + N_2$ collisions is studied at small scattering angles and energies in the range from 0.5 to 3.0 keV. The important collision processes are identified using time-of-flight techniques for energy analysis. Our results show that the quasiresonant $H^+ + N_2 \rightarrow H(1s) + N_2^+(X)$ channel dominates the electron capture only at the smallest angles. As an example, at 1.0 keV, capture to this channel occurs with a probability less than 0.5 for scattering angles beyond 0.7°. The reduced cross section for excitation of the quasiresonant channel shows a maximum which moves to larger reduced scattering angles with increasing projectile energy. A second important process, populating $H(1s) + N_2^+(C)$ is found. Although the excitation of this channel involves a multielectron rearrangement it is found to dominate over the "one-electron" $H^*(n=2) + N_2^+(X)$ channel which lies close in energy. We also find that the electron capture, even at small scattering angles, can generate highly excited N_2^+ states.

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I. INTRODUCTION

A major fraction of the collisions which occur between ions and neutral atoms or molecules result in the capture of an electron which neutralizes the ion. These capture processes are important and found under varied conditions in plasmas, in beam lines of accelerators, and in the atmosphere. They generally result in energy losses and in addition can complicate any modeling calculations. As an example, in the upper atmosphere, kilo-electron-volt energy solar protons can capture electrons and thereby generate keV energy H atoms which may then undergo additional collisions. The modeling must therefore consider the initially occurring H^+ collisions as well as processes involving $H^0(1s)$, and possibly $H^{0*}(2s)$ and H^-. The H^- can result from two-electron capture by H^+ or from electron capture by H^0. There have been numerous studies of electron capture in ion-atom collisions. In the ion-molecule case the number of studies is limited even for relatively simple collisions involving diatomic molecular targets.

The $H^+ + N_2$ collision has been the subject of a number of theoretical and experimental studies. As examples we cite the studies of the vibrational excitation of the N_2 in the direct scattering by Moore [1] and studies of the excitation of $N_2^+(B)$ in $H^+ + N_2$ electron capture by Birely [2] and Lavrov *et al.* [3]. The studies of the $N_2^+(B)$ state were motivated by possible applications to atmospheric processes involving low-keV-energy solar protons. Coincidence studies were performed on $H^+ + N_2 \rightarrow H(n=3) + N_2^+(B)$ by Young, Murray, and Sheridan [4]. The direct scattering and summed electron capture were also studied and addressed within a quasimolecular framework by Dhuicq and Sidis [5]. In a more recent publication Gao *et al.* [6] reported on high-resolution measurements of the summed cross sections for electron capture in $H^+ + N_2$. To data there has been only limited work on electron capture to $H(1s) + N_2^+(C)$ or to higher-lying levels.

In addition to the importance of understanding $H^+ + N_2$ charge-exchange collisions for possible applications, this system is sufficiently simple to guide the development of ion-molecule collision theory. However, there have been no previous studies of the electron capture in $H^+ + N_2$ that directly identify and probe the dominant processes involved. The large cross section for generating H atoms has generally been attributed to the availability of the $H^+ + N_2 \rightarrow H(1s) + N_2^+(X)$ quasiresonant electron-capture channel. We find that although this channel is indeed dominant at very small scattering angles the probability for $H(1s) + N_2^+(X)$ following electron capture falls to less than 0.5 (at an energy $E = 1.0$ keV and scattering angle $\theta = 0.7°$).

II. EXPERIMENTAL TECHNIQUES

Several types of experimental studies, providing complementary information, are generally made on charge-exchange collisions. These include the energy dependence of the total cross section for charge exchange into all final states, the "summed" (into all final states) differential cross sections as a function of scattering angle, and optical studies (which may include coincidence measurements) on selected channels. In this work, using time-of-flight (TOF) techniques, we present results on the electron capture which are differential in both angle and energy loss.

The experimental techniques have been previously described [7] and are only briefly outlined here. The incident H^+ beam is generated in a Colutron ion source. For the present measurements a mixture of H_2 and Ar gases is supplied to the ion source to increase the long-term H^+ beam stability. Although the gas mixture does increase the total H^+ yield in the source, the final H^+ beam intensity (after tuning to minimize the energy spread) is only slightly increased when compared to using H_2 source gas alone. The beam is extracted from the ion source and focused by an Einzel lens. The H^+ beam

passes through a set of shim fields and a collimating aperture into a beam chopping region consisting of two plates (about 1 cm long and separated by 0.5 cm) where it is "pulsed" at 0.3 MHz for the time-of-flight measurements. It is then mass analyzed by a Wien filter and directed into a scattering cell containing the N_2 target gas. After scattering through an angle θ the H^0 traverses a 4.2-m-long flight tube to the detector. The incident H^+ beam typically has a full width at half maximum (FWHM) energy spread of 0.5 eV per keV and an angular FWHM of 0.1°. All measurements are taken under single-collision conditions.

In direct scattering measurements ($H^+ + X \rightarrow H^+$) the incident beam provides a convenient energy reference from which energy losses in the scattered beam can be determined. In electron-capture collisions the detected beam is in a different charge state and a known collision process must be used to supply the energy reference. In this work $H^+ + H_2 \rightarrow H^0$ provides the reference. Figure 1 shows energy-loss spectra for $H^+ + H_2 \rightarrow H^0$ and $H^+ + N_2 \rightarrow H^0$ collisions at an energy $E = 1.0$ keV and scattering angle $\theta = 0.05°$. The data are obtained by acquiring a spectrum for N_2, removing the target gas, and replacing it with H_2. The scattered H^0 beam has a FWHM of 2.5 eV, and within the resolution of the apparatus the maxima of the two peaks are seen to reasonably coincide. The peak shown for the electron capture in the H_2 collision involves a single electronic state $H(1s) + H_2^+(X)$ [7]. A comparison of the shapes of the N_2 and H_2 peaks is consistent with the electron capture from N_2 at this angle primarily involving the single $H(1s) + N_2^+(X)$ channel.

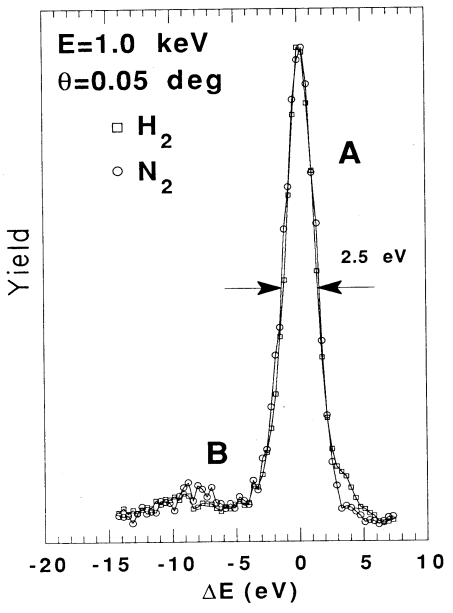

FIG. 1. Energy-loss spectra for $E = 1.0$ keV, $\theta = 0.05°$ $H^+ + H_2 \rightarrow H(1s) + H_2^+(X)$, and $H^+ + N_2$ electron-capture collisions. The "common" positions of the maxima show that the dominant capture from N_2 at this angle occurs to $H(1s) + N_2^+(X)$. The two peaks have the same basic shape indicating that the main peak (A) is due primarily to a single N_2^+ electronic state.

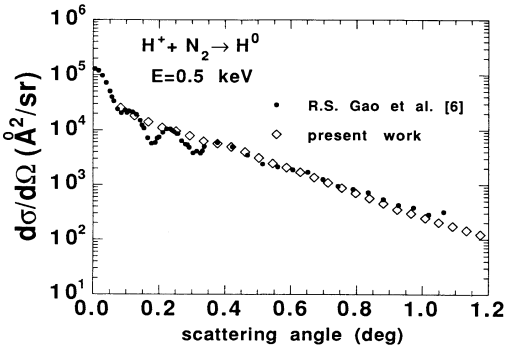

FIG. 2. The summed differential cross section for $H^+ + N_2 \rightarrow H^0$ at $E = 0.5$ keV. The present results are normalized to those in Ref. [6] at $\theta = 0.4°$. "Chopping" in TOF experiments causes losses in beam intensity which require compromises in the apparatus angular resolution. Our results therefore do not show the structure, at the very small angles, reported in Ref. [6]. The two cross sections, however, are seen to be in reasonably good agreement over most of the angular range.

Figure 2 compares our "summed differential cross section" at $E = 0.5$ keV for $H^+ + N_2 \rightarrow H^0$ with Ref. [6]. This cross section, which involves the total neutral flux (independent of state) scattered at each angle, is normalized to Ref. [6] at $\theta = 0.4°$. The very-small-angle structures reported in Ref. [6] is not resolvable since our apparatus is set up for TOF measurements which require larger collimating apertures to compensate for the severe losses in beam intensity due to the required pulsing. The two cross sections do, however, show reasonably good agreement. The cross sections that we report for capture to the $H(1s) + N_2^+(X)$ ground state are determined by

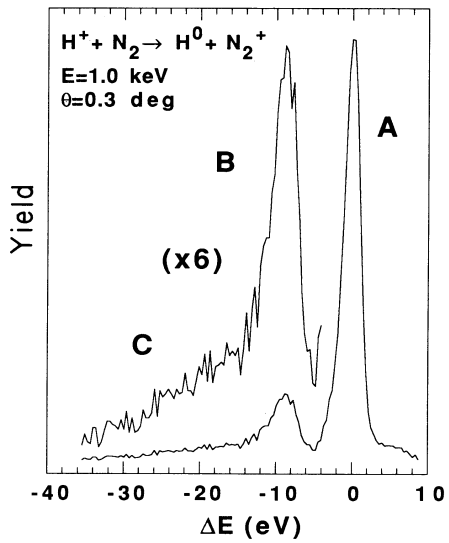

FIG. 3. An energy-loss spectrum for electron capture in $E = 1.0$ keV, $\theta = 0.3°$ $H^+ + N_2$ collisions. The two main peaks are attributed primarily to (A) $H(1s) + N_2^+(X)$ and (B) $H(1s) + N_2^+(C)$. The tail (C) is attributed to capture to $H(1s)$ with the resulting N_2^+ in a highly excited state.

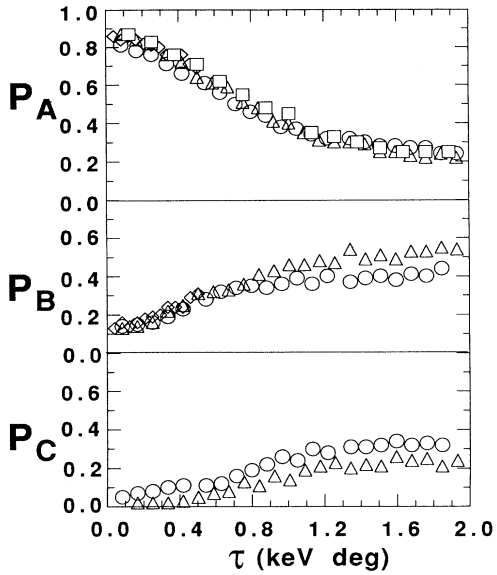

FIG. 4. The probabilities of electron capture as a function of τ, the reduced scattering angle, at energies of 0.5 (◇), 1.0 (○), 2.0 (△), and 3.0 (□) keV. Over the angular range investigated, the plot of P_A corresponds primarily to $H^+ + N_2 \rightarrow H(1s) + N_2^+(X)$. Any contributions from $H(1s) + N_2^+(A\ ^2\Pi_u$ and $B\ ^2\Sigma_u^+)$ are estimated to be less than 10%. Plot P_B results from electron capture primarily to $H(1s) + N_2^+(C)$ with some contributions from $H^*(n=2) + N_2^+(X)$. P_c is attributed to capture resulting in highly excited N_2^+ states. The uncertainties in the reported values are at most: P_A, ±0.03; P_B and P_C, ±0.08.

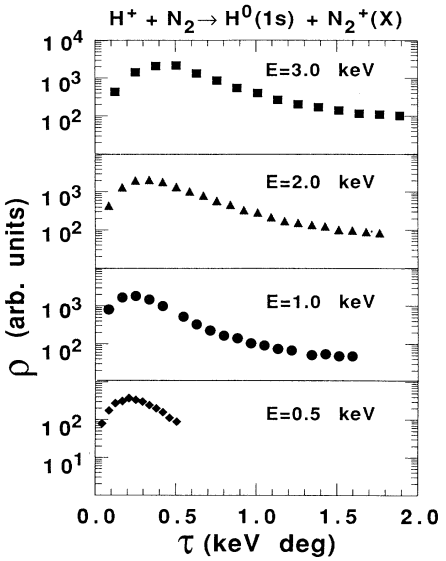

FIG. 5. The reduced differential cross sections as a function of reduced scattering angle for peak A. The curves are plotted to different arbitrary units but clearly show that the maxima move to larger τ values with increasing energy.

multiplying our summed cross sections by the fraction of the scattered H^0 found in peak A of the energy-loss spectra.

III. EXPERIMENTAL RESULTS AND CONCLUSIONS

A typical energy-loss spectrum for $H^+ + N_2 \rightarrow H^0$ at $E = 1.0$ keV, $\theta = 0.3°$ is shown in Fig. 3. Two major peaks are seen. Using Gilmore's [8] potential energy curves the peak maxima are attributed to the $H^+ + N_2 \rightarrow A$ $[H(1s) + N_2^+(X\ ^2\Sigma_g^+)]$ and B $[H(1s) + N_2^+(C\ ^2\Sigma_u^+)]$ electron-capture channels. Over the angular range the shape of peak A is generally consistent with the electron capture dominated by a single electronic state (in agreement with the comparison of the $H^+ + H_2$ and $H^+ + N_2$ spectra presented in Fig. 1). Weak contributions from $H(1s) + N_2^+(A\ ^2\Pi_u$ and $B\ ^2\Sigma_u^+)$ cannot be ruled out, however. Although the shape and position of the maximum in peak B depend somewhat on the scattering angle, electron capture to $H(1s) + N_2^+(C)$ is found to dominate the peak over the angular region studied. This peak (B) may, however, contain some contributions from other states including $H^*(n=2) + N_2^+(X)$. The spectrum also shows a low-energy tail labeled C. The shape of the tail structure is characteristic of the excitation of an N_2^+ state having a potential curve that rises significantly over the Franck-Condon region or to contributions from a number of highly excited N_2^+ states such as reported by Cartwright and Dunning [9]. The excitation of these states would involve "two-electron processes" in the capture.

P_A, P_B, and P_C, the probabilities of electron capture to the processes discussed above, are shown as a function of the reduced scattering angle, $\tau = E\theta$, in Fig. 4. The probabilities are shown only at energies where the peaks are resolvable (P_C is negligible at 0.5 keV).

Figure 5 shows ρ $[=\theta^2\sigma(\theta)]$, the reduced differential cross sections as a function of reduced scattering angle at energies of 0.5, 1.0, 2.0, and 3.0 keV for peak A. In the angular region investigated this peak corresponds primarily to $H^+ + N_2 \rightarrow H(1s) + N_2^+(X)$ with contributions from other states estimated at less than 10%. The maxima in the cross sections are seen to shift to larger τ values with increasing energy. In most cases the maxima occur at a common τ value or move to smaller τ with increasing energy. We would also like to point out that at 1.0 keV the position of the maximum of our "summed cross section" is found at an angle $\theta = 0.2°$. This does not match its position, near 1°, as reported in Ref. [5]. We attribute this disagreement to the higher angular resolution in the present work.

The $H(1s) + N_2^+(X)$ ground-state channel is excited via a Demkov [10] type coupling with the incident $H^+ + N_2$ channel. This excitation, which involves a "one-electron" process, results from the transition to $H(1s)$ of a $3\sigma_g$ electron from the initial $N_2\ (1\sigma_g)^2(1\sigma_u)^2(2\sigma_g)^2(2\sigma_u)^2(1\pi_u)^4(3\sigma_g)^2$ molecular-orbital (MO) configuration. The excitation of $N_2^+(C)$ generates N_2^+ described by a $\ldots(1\pi_u)^3(3\sigma_g)^1(1\pi_g)^1$ and $\ldots(2\sigma_u)^1(1\pi_u)^4(3\sigma_g)^2$ MO configuration [8] following the transition of an electron to $H(1s)$. It is interesting to

note that this more complex process with an excitation energy near 9 eV is favored over the 10.2-eV "one-electron" process resulting in $H^*(n=2)+N_2^+(X)$. This is particularly significant for modeling calculations since capture to $H^*(n=2)$ could populate $H^*(2s)$ which would result in a long-lived state with a large cross section for additional collisions.

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