Formation of Metastable Hydrogen Atoms by Charge Transfer*

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Measurements are presented of the total cross sections for formation of metastable hydrogen atoms by charge transfer as protons traverse targets of helium, argon, nitrogen, and oxygen. Projectile energies range from 4 to 26 keV. An H^+ projectile beam was directed into a cell containing the target gas and emerged into an evacuated region where the metastable-state content was determined. The H(2*s*) flux was monitored by electric field mixing of this state with the 2*p* level and detection of the resulting Lyman-α photon. The relative variation of cross section with projectile energy for targets of He and Ar is in agreement with previous work. For oxygen the cross section increases with energy from 4 to 10 keV and remains constant from 10 to 26 keV; for nitrogen the cross section increases monotonically with increasing energy.

I. INTRODUCTION

The objective was to study the formation of metastable H by the process of charge transfer as a beam of H^+ ions traverses a gaseous target. The reaction is given by

$$H^+ + X \rightarrow H(2s) + [X^+] . \quad (1)$$

The experiment detects the formation of the metastable hydrogen and gives no information on the state of ionization or excitation of the postcollision target system shown within the square brackets. There has been much previous work on processes of the type described by Eq. (1), and the techniques are well established. Jaecks *et al.*,[1] Andreev *et al.*,[2] and Bayfield[3] have made detailed studies of this type of process but confined their work primarily to targets of the rare gases. The present work was undertaken with the primary aim of studying the charge-transfer process on targets of O_2 and N_2, mechanisms that have obvious importance to the understanding of auroral phenomena.

II. APPARATUS

The apparatus for this work was of conventional design and is shown diagramatically in Fig. 1. Hydrogen ions were produced in a rf source, accelerated to energies of between 4 and 26 keV, and mass analyzed to produce an H^+ beam. Two circular apertures of 2.54- and 1.02-mm diam separated by a distance of 36 cm were used to collimate the beam. The target was contained in a cell of 6.7-cm length, the beam entered through a circular aperture of 2.54-mm diam and exited through a circular aperture of 3.2-mm diam. After traversing the cell, the beam emerged into an evacuated region where the excited-state fraction and beam current were monitored. An electric field was applied transverse to the beam to induce mixing between the 2*s* and 2*p* states, causing emission of a Lyman-α photon. The photons were detected by a funneled electron multiplier (Mullard type B 419 BL) which was operated in a counting mode; it was arranged to view perpendicularly to the particle trajectory and to the electric field. A LiF plate was placed over the cathode of the detector so that it was sensitive to photons of wavelengths from the LiF transmission cutoff at 1100 Å to the sensitivity cutoff of the channel multiplier at 2000 Å. The metastable detector was placed 14.2 cm from the exit of the gas cell; at this point spontaneous emission from the 2*p* state had decayed to negligible proportions. Beyond the detector was a Faraday cup which monitored the total flux of ions in the projectile beam. Suitable potentials were provided to inhibit the loss of secondary electrons.

Projectile energy was determined directly by a precision 90° cylindrical electrostatic analyzer, located between the collimating apertures on the path of the incoming H^+ beam. Energies were determined to an accuracy of $\pm 1\%$.

The target gases, stated by the manufacturer to be 99.99% pure, were supplied from high-pressure cylinders and leaked into the scattering chamber through a needle valve. A dry-ice and acetone cold trap was used to remove condensable impurities. Target pressures were generally maintained at or below 10^{-4} Torr; it was demonstrated that at these pressures the measured cross sections were independent of target density. Target pressures were monitored with a capacitance manometer whose response was independent of the nature of the gas. Linearity of response was checked against a trapped McLeod gauge, using H_2 as the test gas, and shown to be linear within $\pm 4\%$. Since the data from this experiment were in the form of relative cross sections, it was unnecessary to determine an absolute value of target pressure.

The beam preparation system, collimator, energy analyzer, and detection systems were located in a

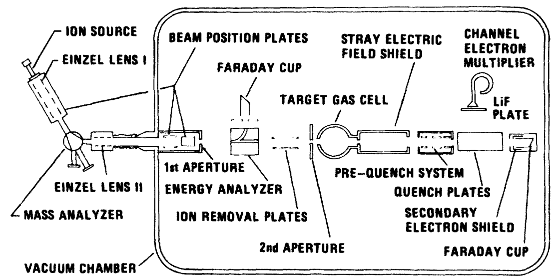

FIG. 1. Schematic diagram of the apparatus.

large tank evacuated with trapped oil diffusion pumps to a base pressure of better than 10^{-7} Torr. The target gas occupied a small cell within the main tank; flow of gas from the target to the main tank was minimized by the small apertures through which the beam entered and exited. The pressure differential between the cell and tank was a factor of 100 or more.

III. EFFICIENCY OF METASTABLE DETECTOR

The objective of the experiment was to determine relative cross sections as a function of impact energy. No attempt was made to directly determine the absolute sensitivity of the system. Particular attention was directed to ensuring that detection sensitivity remained invariant with projectile energy; an empirical test was devised to test this invariance.

The quenching electric field was provided by a parallel plate assembly consisting of two rectangular plates 3.2 cm high by 7.6 cm long and spaced 2.54 cm apart. Two grounded shields were incorporated to reduce the spatial extent of the fringe field; the U-shaped shields wrap around the front of each quench plate and protrude into the space between the plates approximately 2.6 cm leaving 5 cm of the quench plate exposed. Between the quench plates, the shields were spaced 0.508 cm apart and were centered on the axis of the particle beam. Care was taken to ensure that no appreciable fraction of the metastables were quenched outside the detector's field of view by fringe fields from the quench plates.

The intensity of the electric field along the beam axis was calculated to an accuracy of $\pm 10\%$ by the use of a mapping function. The intensity rises from 0.1 to 90% of its asymptotic value in a distance of 1 cm; it is approximately 50% at the edge of the grounded shield. Thus the fringe field was negligible at distances greater than 0.5 cm from the edge of the grounded shield. The detector viewed a region extending from 2 cm in front of this edge to 7 cm past this edge; consequently, the small fraction of the metastables quenched by the fringe field (0.5%) did contribute to the measured signal. It was concluded that there was no significant loss of signal due to quenching by fringe fields.

It was demonstrated that the signal saturated as the applied field was increased and the effective lifetime of the 2*s* state became essentially twice that of the 2*p* state. The difference between the signal with the quench field on and with it off should represent the field-quenched emission from the 2*s* state. It was observed that when the quench field was turned off there was an appreciable background signal. The sources were not completely identified, but the signal included components caused by impact of the beam on surfaces and excitation of background gas. There was concern that when the quench field was turned on this background might change, particularly due to the alteration in trajectory of the projectile ions and acceleration of stray electrons onto metal surfaces. This problem was obviated by placing before the detection region a "prequench" electric field parallel to the beam axis. The prequench field had the function of removing the metastables from the beam before it entered the observation region; tests indicated that removal was 97% efficient. Background signals observed by the photon detector will be unaffected by whether the prequench field is on or off. The quench field in the detection region was maintained slightly above the value which produced saturation in the signal; the difference between the signal with the prequench off and the signal with the prequench on was taken as the true signal from quenching of metastables.

Under high-quenching fields sufficient to ensure complete mixing of the 2*s* and 2*p* levels, the effective lifetime of the metastable state is 3×10^{-9} sec, twice that of the 2*p* level. The decay length (product of impact velocity and lifetime) in the quenching electric field is comparable with the field of view of the detector. Consequently as velocity changes, the distribution of emitters in the quench region will also be altered. In the event that the efficiency of the photon detector varies with the angle of incidence of the photon, the detector sensitivity might exhibit a dependence on impact velocity. Tests are required to assess the magnitude of this effect. The lifetime of the H(2*s*) state is dependent on the strength of the quenching electric field and may be predicted theoretically, following the work of Bethe.[4] It would be possible to guarantee velocity-independent detection efficiency by varying quenching field with velocity to maintain a constant decay length. This does have the disadvantage, however, that quenching fields will be less than that for an optimum efficiency; and therefore signal-to-noise ratios are degraded. In the present experiment the detection efficiency was maintained at its optimum value using saturation fields. A test of the velocity independence of this efficiency was carried out by measuring the ratio of the signal under optimum quench field to the signal at a lower field where decay length was kept constant; the variation of this ratio with impact velocity reflects changes in detection sensitivity. This test indicated that the magnitude of the optimum detection efficiency varied by no more than 10% over the impact energy range 4–26 keV. Rather than make a correction for this change, we choose to regard it as a contributing factor to the limitation of accuracy with which the cross sections were determined.

It has recently been shown[5] that the field-induced Lyman-α emission will be polarized by an amount which varies with field strength. Polarization is related to anisotropy of emission. An experiment which detects photons emitted into a limited solid angle will therefore experience a change in effective detection sensitivity with electric field. The test described above depends upon the detection efficiency at a series of selected low fields being independent of impact velocity. Using the theoretical prediction of polarization of Sellin *et al.*[5] it is readily shown that over the range of low fields used to maintain constant decay length (30–40 V/cm) the detection efficiency of the system varied by a negligible amount[6] (less than $\frac{1}{4}$%). The high-quench field used during cross-section measurements was not maintained constant; various values were employed between 300 and 600 V/cm, depending on projectile energy; the changing polarization contributes to the variation of detection efficiency with projectile energy that is identified by the tests described above. In conclusion, the existence of the field-dependent polarization does not invalidate the result that detection efficiency changes by less than 10% over the impact energy range of this experiment.

Care was taken to ensure that no appreciable fraction of the flux of metastable atoms was lost before the projectile beam entered the detection region. Loss due to interception by the exit aperture of the gas cell was assessed from a study of the angular distribution of the metastables to be less than 1%. The possibility that metastables were destroyed by collisions with background gas was shown to be negligible by demonstrating independence of signal from background pressure. Destruction due to stray-field quenching was prevented by complete shielding of the beam from high-voltage leads and insulating surfaces; the fringing of fields from the quenching regions was minimized by grounded shielding. It was estimated that with the precautions described above, the loss of metastables did not amount to more than 1% of the flux.

There is a possibility of a spurious contribution to the signal from metastable atoms formed by neutralization of protons traversing the region outside the target cell. Formation of metastables by charge transfer on background gas was evaluated by measuring the metastable signal with the target cell evacuated; this was subtracted from the signal observed with gas in the cell, so arriving at a true signal due to charge transfer on the target medium. In most cases the correction was negligible. The true signal included a contribution due to metastables formed by neutralization on target gas that leaked from the target cell into the main vacuum chamber. This contribution is estimated to be less than 10% of the metastable flux produced in traversing the target cell and has the effect of introducing an uncertainty in the effective thickness of the target. Its presence does not, however, materially influence the present experiment since the data are relative and no attempt was made to determine the true target thickness.

IV. NORMALIZATION OF DATA

It was not the purpose of this present work to measure absolute cross sections. However, recognizing the utility of absolute values in practical situations, the data were assigned absolute values by normalization to previous experiments.

Bayfield[3] noted that there are three independent determinations[1–3] of the cross section for the process

$$H^+ + Ar \rightarrow H(2s) + Ar^+ . \qquad (2)$$

He suggested that since the data agree within 12%, one might normalize future measurements of metastable production to these previous determinations. The apparent agreement of the three independent determinations is somewhat illusory. All three experiments assumed that the field-induced emission was isotropic; Sellin *et al.*[5] showed this assumption to be incorrect; the emission is polarized with respect to the direction of the quenching field. Moreover the degree of polarization is dependent upon the strength of the electric field. Sellin *et al.*[5] calculated the relationship of polarization to the quenching field and confirmed these predictions by experimental measurement. It is not clear whether the polarizations predicted and measured by Sellin *et al.*[5] may legitimately be employed to correct for the polarization-related anisotropy in the published experiments. None of the previous experiments utilize conditions that are similar to those employed by Sellin *et al.*[5] In two of the experiments[1,3] the quenching field is nonuniform; for two of the experiments[1,2] the excited atoms are formed within the quenching field itself. We therefore conclude that all the existing data may be in error due to neglect of anisotropy; moreover, the magnitude of the error cannot be reliably estimated in retrospect. Consequently, there are no reliable data to which the present experiment may

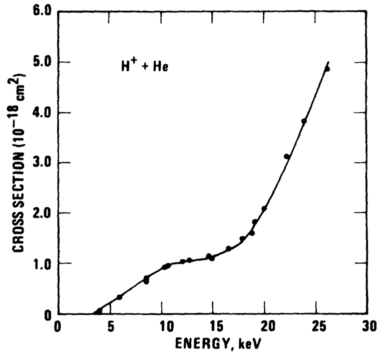

FIG. 2. Cross sections for the formation of H(2*s*) by impact of H^+ on He.

be normalized.

In order to provide absolute magnitudes we choose to normalize the present results to a cross section of 2.90×10^{-17} cm^2 for the formation of metastable hydrogen by impact of 20-keV protons or argon; this cross section is the mean of the values published in the literature.[1-3] In view of the confusion concerning polarization, no estimate is made of the reliability of these absolute magnitudes.

V. RESULTS

Figures 2–4 show the results of the present experiments; the cross sections are expressed in units of cm^2 per molecule. It is emphasized that the relative values of cross sections for the different targets are obtained directly from the present measurements; the absolute values of the whole set of data are established by normalization to a value of 2.9×10^{-17} cm^2 for the cross section in $H^+ + Ar$ at 20-keV impact energy. Reproducibility of the present data was within ± 5%. Systematic errors in the energy dependence of the data should not distort the ratio of the values of a cross section measured at the extreme energies of the available range by more than 10%. The ratios evaluated for smaller differences in energy will be of higher accuracy. The data represent the cross section for formation of the 2*s* state by charge transfer and include cascade from higher levels.

The present results may be compared with previous experiments using targets of He and Ar. There is considerable disagreement between the absolute values in the previous data, reflecting the difficulty of carrying out accurate absolute determinations of emitted light intensity. Since the present data are relative rather than absolute, the most valuable comparisons may be carried out in terms of the relative variations of cross section with impact energy. Such a comparison is shown in Figs. 5 and 6, which include all available data for targets of He and Ar normalized together at

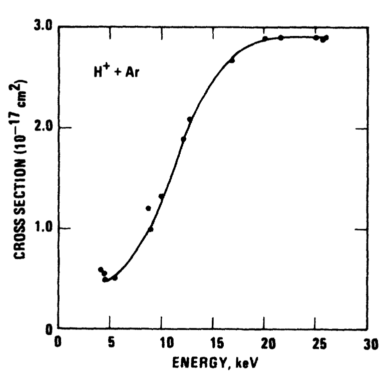

FIG. 3. Cross sections for the formation of H(2*s*) by impact of H^+ on Ar.

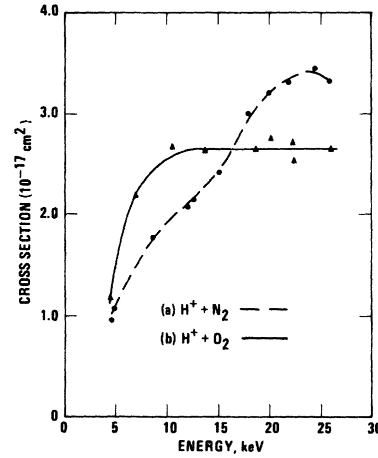

FIG. 4. Cross sections for the formation of H(2*s*) by impact of H^+ on (a) N_2 and (b) O_2.

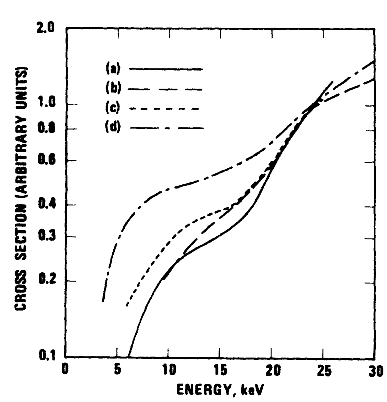

FIG. 5. Comparisons of data for formation of H(2*s*) by impact of H^+ on a He target. All data are shown normalized at an energy of 24 keV. (a) Present work, (b) Andreev *et al.* (Ref. 2), (c) Jaecks *et al.* (Ref. 1), (d) Dose (Ref. 7).

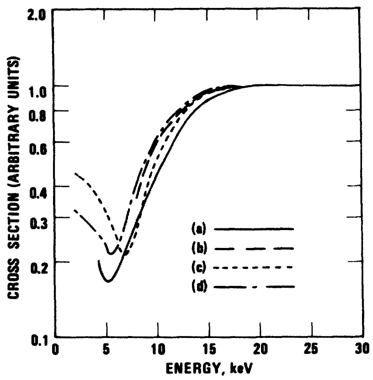

FIG. 6. Comparisons of data for formation of H(2*s*) by impact of H^+ on an Ar target. All data are shown normalized at an energy of 24 keV. (a) Present work, (b) Andreev *et al.* (Ref. 2), (c) Jaecks *et al.* (Ref. 1), (d) Bayfield (Ref. 3).

an energy of 24 keV. It is clear that appreciable disagreement exists among the various authors. In the case of the helium target, if one neglects the work of Dose,[7] the remaining values diverge by up to 12% from the mean. For argon all data lie within 15% of the mean down to an energy of 6 keV; below this point the divergence is more serious. The various apparatus used in these measurements are of different design; thus instrumental errors in the data may differ from one experiment to another. The contribution of cascade, coming primarily from the $3p \rightarrow 2s$ transitions, will vary from one experiment to another due to the different geometrical configurations. However, the direct measurements by Andreev *et al.*[8] of the 3*p* cross section at energies above 10 keV indicate that cascade cannot exceed 4% for any of these determinations; thus cascade is a small contribution to the measured cross sections and cannot contribute significantly to the discrepancies. In all experiments there is a danger that the detection sensitivity will vary with energy; the work of Jaecks *et al.*[1] contains such an error amounting to 10% or more due to Doppler shift of emission[9]; the techniques of Andreev *et al.*[2] inherently exclude this type of error; the present work has assessed such errors to be less than 10%; the remaining experiments ignore the problem.

It is not possible to make a definite identification of sources of error to explain the discrepancies between measurements. Consequently, one cannot use objective criteria to select one set of data as being more accurate than the others. Excluding the work of Dose, which exhibits a considerable disagreement with the other determinations, one may conclude that the available data establish the energy dependence of the cross sections to an accuracy of ± 15%. Within this uncertainty the results of the present experiment are consistent with previous work.

Gaily[10] carried out a comparison with theory of the cross-section data for a helium target. It was shown that a coupled-state calculation[11] provides the best description of the process at energies below 30 keV, but there remains a considerable discrepancy between theory and experiment.

There are no previous data for N_2 and O_2 targets with which the present measurements may be compared. It is interesting that these two cross sections are of about the same magnitude but exhibit different dependence on energy; this indicates perhaps that the charge-transfer mechanism is sensitive to the detailed electron structure of the target. Mapleton[12] has calculated capture into the 2*s* state for targets of O and N using the orthogonalized Brinkman-Kramers (OBK) approximation. It might be expected that the cross section for atomic O or N will be roughly equal to half that for the O_2 and N_2. In fact the theory lies as much as one order of magnitude higher than experiment and exhibits a greatly different dependence on energy. This is not too surprising since the OBK approximation is designed for use at higher impact energies than the present experiment.

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