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Formation of metastable hydrogen atoms by charge exchange of protons in cesium vapor

Vu Ngoc Tuan and G. Gautherin
Institut d'Electronique Fondamentale, Université Paris-XI, 91405 Orsay, France

A. S. Schlachter
Institut d'Electronique Fondamentale, Université Paris-XI, 91405 Orsay, France, and
Service de Physique Atomique, Centre d'Etudes Nucléaires de Saclay – 91190 Gif sur Yvette, France

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Formation of an intense beam of hydrogen atoms in the metastable 2*s* state is not only of basic interest, but is also useful for polarized-ion sources of the Lamb-shift type. The nearly resonant reaction $H^+ + Cs \rightarrow H(2s) + Cs^+$ is often used for this purpose. The fraction f of metastable atoms relative to the number of neutral atoms in all $n = 2$ states formed in the collision of a proton in a thin Cs-vapor target is reported here for the energy range 0.4–3.0 keV. An apparent maximum of $f = 0.43 \pm 0.03$ at 0.5 keV is found. For energies above 0.75 keV, $f = 0.25 \pm 0.01$, which would be expected if the $n = 2$ states of H are statistically populated. Previously reported values for σ_{+m}, the cross section for electron pickup in the metastable 2*s* state of hydrogen for $H^+ + Cs$ collisions, differ by two orders of magnitude for a given energy. It can be assumed that essentially all electron pickup is into the $n = 2$ states at low energy. Thus measured values of f can be multiplied by reported values of σ_{+0}, the cross section for electron pickup into any neutral state of the H atom, to find values for σ_{+m}. As this method is independent of calibration or normalization of the photon measurements, these values for σ_{+m} are more reliable than those previously reported. Our results for σ_{+m} are between 4.3×10^{-15} and 1.5×10^{-15} cm^2 in the energy range 0.5–3.0 keV.

I. INTRODUCTION

Formation of an intense beam of hydrogen atoms in the metastable 2*s* state is not only of basic interest but is also useful for subsequent collision studies. An important application is in nuclear physics for sources of polarized H^+ and H^- ions of the Lamb-shift type,[1] in which a polarized-ion beam is formed by charge exchange of polarized H(2*s*) atoms in an appropriate gas target.

The usual method to form a beam of fast H(2*s*) atoms is by charge exchange of protons in a gas target. This method has the disadvantage that the cross section for formation of H(2*s*) atoms is small and that the maximum value of the cross section occurs at energies higher than is convenient for many applications. For example, the cross section for formation of H(2*s*) in H_2 has a maximum value of 2.7×10^{-17} cm^2 at an energy of 30 keV.[2]

An improved method for creating an intense beam of H(2*s*) atoms is by the nearly resonant charge-exchange collision of protons in Cs vapor, first proposed by Donnally *et al.*[3] [Eq. (1)]:

$$H^+ + Cs \rightarrow \{H(2s), H(2p)\} + Cs^+ - 0.49 \text{ eV}. \qquad (1)$$

The cross section for this process is large, and the maximum should occur at low energy because of the small energy defect.

The following notation is used in this paper: σ_{+m} is the cross section for electron pickup by a proton to form H^0 in the metastable 2*s* state; σ_{+0} is the cross section for electron pickup by a proton to form H^0 in any state (total charge-exchange cross section). The metastable fraction f is the ratio of the number of atoms formed in the 2*s* state to the number of atoms formed in all $n = 2$ states (2*s* and 2*p*) as the result of a single collision of a proton in Cs vapor. In the limit that Eq. (1) is the only possible reaction—that is to say, that all neutral atoms formed in the collision of a proton in Cs vapor are in $n = 2$ states—f can be called the metastable fraction of the neutral beam. This is a reasonable approximation at low energies (<2 keV), as discussed in III E. In this limit we can also replace σ_{+0} by the cross section for formation of neutral atoms in the $n = 2$ states. Thus $f = \sigma_{+m}/\sigma_{+0}$ in this limit.

There have been several experiments involving reaction (1). However, the metastable fraction f as a function of H^+ energy has not previously been reported. It is this fraction f that we have measured in the energy range 0.4–3.0 keV. Because the method employed does not require calibration of the metastable-atom detector, the measured values of f can be combined with previously measured values of σ_{+0} to obtain more-reliable values for σ_{+m} than have been previously reported. Preliminary results have been recently published.[4]

A summary of reported measurements of cross

TABLE I. Summary of reported measurements of collisions of hydrogen atoms and ions in alkali-metal targets.

Reference		Measured	Target	Energy range (keV)
Il'in *et al.*	5, 6	σ_{+0}, hes[a]	Li, Na, K, Cs	10–180
Schlachter *et al.*	7	σ_{+0}, σ_{g+}, σ_{+-}, σ_{g-}, F_{+}^{∞}, F_{0}^{∞}, F_{-}^{∞}[b]	Cs	0.5–20
Spiess *et al.*	8	σ_{+0}	Cs	0.5–2.5
		σ_{+m}, f[c]	Cs	2.4
Spiess *et al.*	9	σ_{+0}, σ_{+-}, σ_{-0}, σ_{0-}, F_{+}^{∞}, F_{0}^{∞}, F_{-}^{∞}	Cs	2.5
Grüebler *et al.*	10,11	σ_{+0}, σ_{+-}, F_{+}^{∞}, F_{0}^{∞}, F_{-}^{∞}	Li, Na, K, Cs	1–20
Donnally *et al.*	3	σ_{+m}	Cs	0.16–3.0
Cesati *et al.*	12	σ_{+m}	Cs	0.5–5
Sellin and Granoff	13	σ_{+m}	K, Rb, Cs	2–30
Roussel *et al.*	14-16	σ_{+m}, σ_{+g}, f	Cs	0.3–3
D'yachkov *et al.*	17, 18	F_{0}^{∞}, F_{-}^{∞}	Li, Na, K	1.5–40
Bohlen *et al.*	19	F_{-}^{∞}	Cs, K	0.5–2
Khirnyi and Kochemasova	20	F_{-}^{∞}	Cs	~0.2–6
Nieman	21	σ_{+0}, F_{0}^{∞}	Na, K	4–30
Leslie *et al.*	22	σ_{-0}, σ_{-+}	Cs	2–30
Futch *et al.*	23	hes	Li, K	5–35
Schlachter *et al.*	24	σ_{+m}, σ_{+g}, σ_{mg}, σ_{m-}, σ_{g-}	Cs	0.5

[a] Yield of highly excited states.

[b] F_{i}^{∞} is the equilibrium charge fraction for the charge state i.

[c] Subscript notation is as follows: +, −, m, g, and 0 refer, respectively, to H^+, H^-, H(2s), H(1s), and H^0 in any state. f is metastable fraction (see definition in text).

sections and charge-fraction yields is shown in Table I for hydrogen ions and atoms in alkali targets. Reported values of σ_{+0} in Cs are generally in agreement at low energy (up to 5 keV), but differ seriously at higher energies. Values of σ_{+m} in Cs, however, depend either upon absolute calibration of the metastable-atom detector or upon a normalization process. The disparity between values reported at the same energy can be very large; for example, at an H^+ energy of 2.4 keV, reported values of σ_{+m} differ by more than two orders of magnitude. It is therefore clear that it is not possible to compute reliable values of f from reported values of σ_{+m} and σ_{+0}.

The only previously reported value of the metastable fraction is a value of 0.27 ± 0.08 at 2.4 keV by Spiess *et al.*[8] Donnally *et al.* have stated that between 0.25 and 0.5 keV (0.5–1.0 keV D^+) about 25% of the neutral atoms emerging from $D^+ + Cs$ collisions are in the 2s state.[25] Donnally and O'Dell[26] have also determined a lower bound for f by an indirect method. They found that this bound has a maximum value of 0.33 at 0.4 keV.

We mention certain well-known properties of the H(2s) and H(2p) states necessary to understand the present experiment. The field-free lifetime of an H atom in the metastable 2s state is very long (0.14 sec), while that of the radiative 2p state is 1.6×10^{-9} sec. In the presence of an applied electric field, the 2s state is Stark mixed with the nearby radiative 2p states, with the resulting emission of a Lyman-α photon having a wavelength of 1216 Å. This process is called quenching. In a sufficiently strong applied electric field ($E \sim$ several thousand V/cm), the lifetime of an atom in the 2s state approaches twice the lifetime of the 2p state. A field of 500 V/cm is sufficient to reduce the 2s lifetime to 3.5 times the 2p lifetime; i.e., the 2s lifetime is 5.6×10^{-9} sec in this field.

The Lyman-α radiation resulting from field-induced quenching of H(2s) is known to be polarized.[27-31] This is further discussed in Sec. III D.

II. EXPERIMENTAL APPROACH

A. General description

The apparatus, shown in Fig. 1, consists of a proton beam which traverses a thin Cs-vapor target. This target is a Cs-vapor jet issuing from an oven, and intersects the H^+ beam at 90°. The collision zone (Fig. 2) is surrounded by a cooled box to trap Cs atoms. An electric field can be applied in the collision zone to quench the H atoms in the 2*s* state without seriously perturbing the proton trajectories. Located at 90° to the plane of the H^+ and Cs beams and centered above the collision zone is a uv-sensitive photomultiplier which is the detector of Lyman-α photons. From the photon signals without and with applied quenching field, i.e., from the H(2*p*) and the H(2*p*) plus quenched H(2*s*) atoms, the fraction of the neutral beam in the 2*s* state can be calculated without absolute calibration of the Lyman-α detector.

B. Proton beam

Protons from a duoplasmatron are extracted at 10 keV, focused by three electrostatic lenses, analyzed by a 90° electromagnet, and then decelerated to their final energy. Ion energy was determined by measuring the voltage of the ion-source anode (the Cs-target system was at ground potential). This potential gives the ion energy to within 10 eV, which corresponds to the possible difference in potential between the anode and the ions due to the distribution of potentials in the ion source.

The proton beam is collimated by two circular apertures. The first (3-mm diam) is located 70 mm before the entrance to the cooled box in which the collisions take place (see Fig. 2). The second aperture (1-mm diam) is located at the end of a tube which extends outside the box. The purpose of the tube is to ensure that any photons which might be created by the H^+ beam striking the aperture are well outside the field of view of the photomultiplier (PM).

After traversing the collision zone, the proton beam passes through a large hole in the box, where it is detected by a suppressed Faraday cup sufficiently large to intercept the entire beam. This cup is recessed for the same reason as the second beam-collimating aperture— to avoid a photon signal from the beam striking the Faraday cup. Typical proton current is 1×10^{-6} A.

Although the proton beam is measured after it traverses the Cs target, it is essentially undiminished by charge exchange, as the Cs target is very thin (of the order of 3×10^{11} atoms/cm^2). As σ_{+0} is of the order of 7×10^{-15} cm^2, the product of σ_{+0} and target thickness is of the order of 0.002.

In order to extend the measurements to low energies, a D^+ beam was substituted for the H^+ beam. Deuteron results were shown to be the same as proton results when compared at the same velocity, i.e., with the D^+ results at half the nominal energy.

C. Cesium jet

A jet of Cs is used as a charge-exchange target, rather than an oven, in order to have an unobstructed view of the collision zone for photon measurements. The jet is created by a stainless-steel oven located inside the vacuum chamber. The oven consists of two sections: body and head, separated by a valve operated from outside the vacuum system, so as to permit background measurements without Cs while the oven is hot. The Cs jet leaves the oven by a 6.0×0.5-mm horizontal slot in the head. The jet is further collimated by a heated diaphragm with a 2-mm-diam circular aperture in the center and located 5 mm in front of the oven. Typical oven temperature is 250 °C. The head is

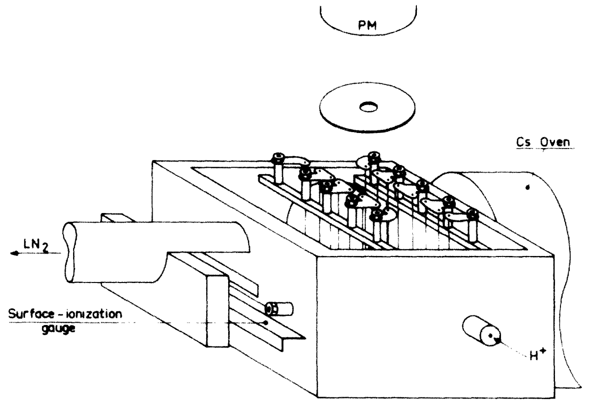

FIG. 1. Beam interaction region in perspective. The proton beam enters the cooled Cu box by the small aperture shown in front, and leaves by the large aperture behind. The cesium-vapor jet is created by the oven at the right and is detected by the surface-ionization gauge at the left. Photons are detected by the photomultiplier (PM) shown at the top. The quenching field is provided by the vertical wires.

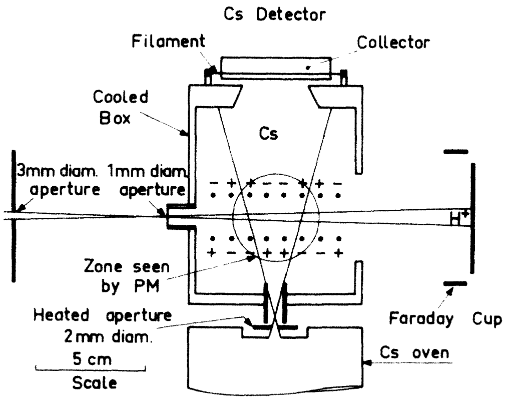

FIG. 2. Beam interaction region viewed from above. Circle indicates field of view of PM in the collision plane. Solid circles indicate wires (normal to the plane) used to obtain the quenching field (relative polarity indicated).

separately heated, and is maintained approximately 20 °C hotter than the oven, in order to prevent blocking of the slot by Cs.

Cesium density is of the order of 1×10^{11} atoms/cm^3 at the working distance of 6 cm from the slot. This density was roughly determined in a separate experiment using a surface-ionization detector. An accurate value of the density is not required in this experiment, the only requirement being that the density be sufficiently low to ensure single-collision conditions for the proton beam.

The collisions take place in a rectangular box composed of four Cu plates, cooled to approximately -80 °C in order to trap the Cs jet. The box is in contact with a liquid-nitrogen reservoir by means of a large Cu rod passing outside the vacuum system.

The Cs jet enters the box by a 10-mm-diam tube located in one wall of the box (see Fig. 2). This tube and the heated aperture at the exit of the oven are such that all Cs atoms see a cold surface (and are therefore trapped), rather than the walls of the vacuum chamber or the insulators which support the quench-field wires, in order to prevent contamination of the vacuum system and degradation of the insulators, and to avoid obstruction of the Cs jet by condensed Cs.

Cesium density is monitored by a surface-ionization gauge located behind a 30×5-mm horizontal slot in the wall of the Cu box opposite the entry slot.[32] The gauge consists of a heated W wire parallel to the slot. Ions are collected by a biased plate located below the filament. We thus have a relative measure of the density of a rectangular sample of the Cs jet. This gauge was designed so as to create no appreciable electric field inside the box which could cause quenching of H(2*s*) atoms. A cold Cu plate behind the Cs gauge traps those Cs atoms which pass through the gauge slot.

D. Lyman-α detector

It is necessary to measure the Lyman-α signal in the absence of electric field and with an applied electric field sufficient to quench essentially all the H atoms in the 2*s* state.

The Lyman-α detector is an EMR 541 G photomultiplier with a LiF window, located at 90° to the plane of the H^+ and Cs beams, and centered over the collision zone. It is sensitive in the range 1150–1800 Å. The PM is diaphragmed so as to have a circular field of view of approximately 4 cm in the collision plane; i.e., it views the entire collision region.

The spectrum of the radiation emitted in the collision zone was measured using a vacuum uv spectrometer which was mounted above the collision zone in the place normally occupied by the PM. The only detectable radiation (i.e., the only measurable radiation in the range 1150–1800 Å) was Lyman-α (1216 Å) when the proton beam was incident on the Cs-vapor target, both with and without applied electric quenching field. It was therefore justifiable to utilize the PM as a Lyman-α detector without filter or spectrometer.

The quenching electric field was designed to provide a field sufficiently strong to quench H atoms in the 2*s* state without seriously perturbing the proton trajectories. This is important because a change in trajectory would change (i) the path length of the H^+ beam in the Cs, (ii) the solid angle for radiation seen by the PM, and (iii) the number of excited H atoms formed, as the Cs density is not uniform. The geometry chosen, a field which al-

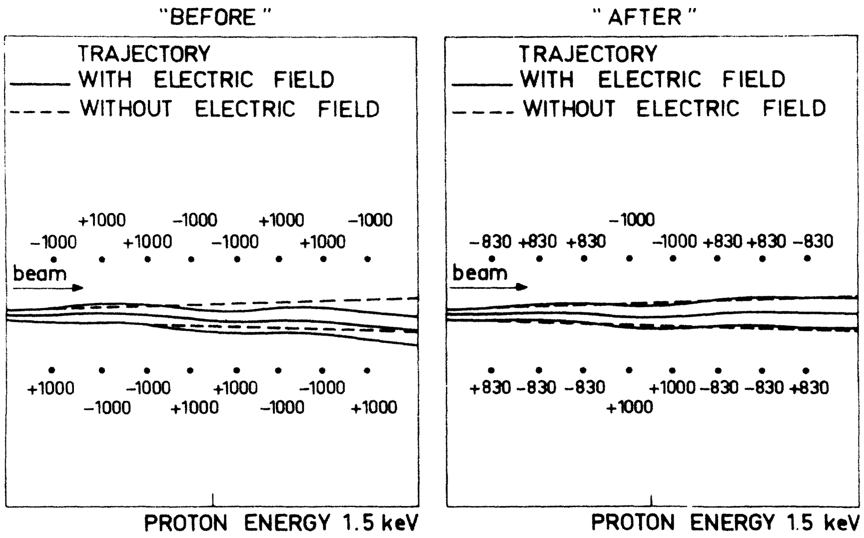

FIG. 3. Calculated proton trajectories for two quenching-field configurations. The improved "after" configuration was used for all reported measurements. Figures next to each point (wire perpendicular to the plane) indicate the applied voltage. Exterior rectangle is at ground potential.

ternates in direction, is well known.[33] It is created by two parallel rows of eight wires each, 8 mm between each wire and 20 mm between the two rows. The voltage applied alternates in sign every two wires (see Figs. 2 and 3). It was found, however, that the photon signal depended upon the sign of the applied electric field. We therefore modified the field by increasing the voltage on the four central wires. The correction necessary for optimal trajectories was determined by numerically integrating Laplace's equation in two dimensions (the collision plane) using an accelerated-convergence iterative method. Proton trajectories in the resulting field were then calculated for three cases: the center of the H^+ beam and the geometrical limits. It was found that a voltage V applied to the four central wires and $0.83V$ applied to the remaining 12 minimized the perturbation of the proton trajectories. "Before" (1000 V on all wires) and "after" (1000 V on the central wires, 830 V on the others) calculated trajectories are shown in Fig. 3 for a proton energy of 1.5 keV. Equipotential lines for the after configuration are shown in Fig. 4.

It was confirmed experimentally that the after configuration indeed gave a Lyman-α signal independent of the sign of the applied voltage. As a precaution, however, all data were taken with the applied voltage in both directions (see Fig. 5).

E. Measurements

Atoms in the H(2s) and H(2p) states are created by the collision of a proton in the Cs target. The H(2p) atoms decay almost immediately to the ground state ($\tau = 1.6\times10^{-9}$ sec) with the emission of a Lyman-α photon. In the absence of applied fields, the H(2s) formed do not decay and leave the collision chamber ($\tau = 0.14$ sec). The measured Lyman-α signal S_1 is therefore proportional to the number of H(2p) formed. In the presence of a strong electric field, however, the H(2s) atoms formed also decay almost immediately ($\tau \simeq 6\times10^{-9}$ sec) with the emission of a Lyman-α photon. Thus, in the presence of an electric field sufficient to quench essentially all the H(2s), the Lyman-α signal S_2 is proportional to the number of H atoms in the $2p+2s$ states. (See Sec. III E for a discussion of possible cascade effects.)

Typical data for Lyman-α signal as a function of applied quenching voltage are shown in Fig. 5. The signal S_2 is seen to be independent of applied voltage above a certain minimum value. Thus the fraction f of atoms in the $2s$ state relative to the number in the $2s+2p$ states is

$$f = (S_2 - S_1)/S_2 .$$

It is necessary to correct for background signal due to PM dark current and to collisions of protons in the residual gas. Typical ambient pressure is 1×10^{-6} Torr. The cross section for formation of H ($n=2$) in a collision of a proton with typical residual gas (H_2) is roughly a factor of 400 smaller than in Cs.[2] As the Cs-target density corresponds to a pressure of the order of 10^{-5} Torr, collisions of protons with residual gas leading to formation of H atoms in the $n=2$ states are relatively unimportant. Indeed, typical background signal (mainly PM dark current) is generally less than 8% of the signal with Cs, and is subtracted before calculating f. In order to correct for small fluctuations in H^+ beam intensity and Cs density,

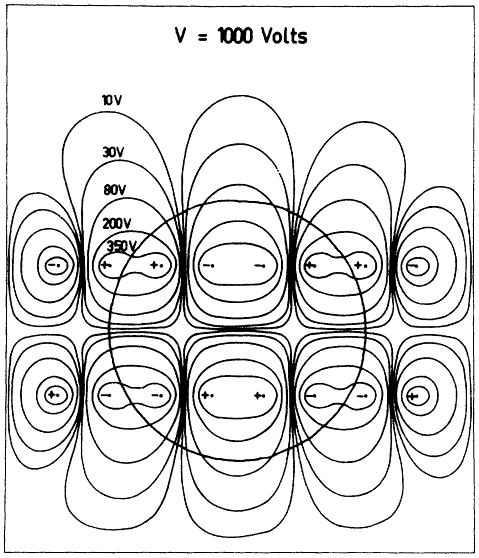

FIG. 4. Calculated equipotential lines for quenching field in "after" configuration of Fig. 3. Circle indicates field of view of the PM in collision plane.

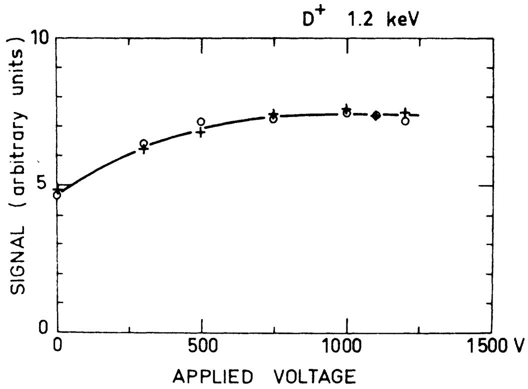

FIG. 5. Typical data: Lyman-α signal as a function of applied quenching voltage. The symbols + and ○ refer to the quenching voltage applied in the usual sense and reversed.

the PM signal at each point is normalized to the H$^+$ current and to the Cs-gauge current.

III. RESULTS AND DISCUSSION

A. Metastable fraction

Experimental results for the metastable fraction f are shown in Fig. 6 and Table II. It is to be noted that f has an apparent maximum of 0.43 ± 0.03 at 0.5 keV and that, for energies above 0.75 keV, $f = 0.25 \pm 0.01$, which would be the result to be expected if the 2s and 2p states are statistically populated. These results are for a thin target (single-collision domain), as it is known that the cross section for destruction of H(2s) in Cs is very large (of the order of 5×10^{-15} cm^2).[24]

The only direct result which can be compared with the above is a measurement by Spiess *et al.*[8] of $f = 0.27 \pm 0.08$ at 2.4 keV, which was determined in an absolute measurement by measuring Lyman-α photons with a calibrated Channeltron and by simultaneously measuring the flux of H^0. The agreement is very good.

Donnally and O'Dell have determined a lower bound for f [26] by measurement of the H$^-$ current after two single collisions in Cs: the first to form H(2s), the second to form H$^-$. By applying a quenching field between the two collisions, and by assuming that $\sigma_{g-} \gg \sigma_{m-}$ (σ_{g-} and σ_{m-} are the cross sections for electron capture by an H atom in the ground state and in the metastable 2s state, respectively), they found that the lower bound of f has a maximum value of 0.33 at a proton energy of 0.4 keV. This is in satisfactory agreement with the present results. Furthermore, there is reason to believe, contrary to the claim of Donnally and O'Dell, that σ_{g-} and σ_{m-} are of approximately the same order of magnitude in the energy range of interest.[24] Thus the value of f would be greater than the lower bound of Donnally and O'Dell, which could improve the agreement.

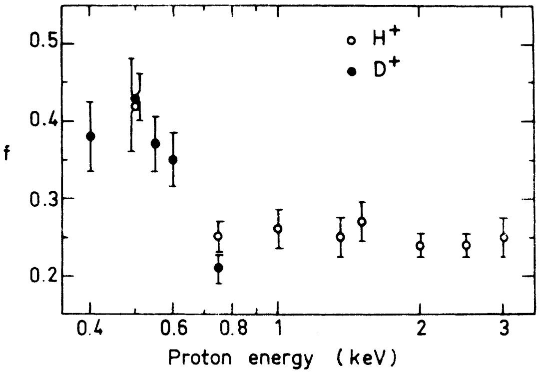

FIG. 6. Metastable fraction f [ratio of H(2s) atoms to total number of atoms in the $n = 2$ states], for H$^+$ (D$^+$) incident on a thin cesium-vapor target. D$^+$ points are shown at the equivalent H$^+$ velocity.

B. Cross section σ_{+m}

Although the cross sections σ_{+m} and σ_{+0} have been measured, only the values of σ_{+0} can be considered to be reliable. The measurements of σ_{+m} depend either upon calibration of the metastable-atom detector or upon normalization, either of which can be rather difficult. This is evident in the great disparity of published values (Fig. 7). Thomas[34] has criticized the experiments of Donnally *et al.*, Sellin and Granoff, and Cesati *et al.*, concluding that, because of doubtful calibration and/or normalization, the results cannot be considered to be reliable. Thus only the recent results of the companion experiment to this one[14-16] can be seriously considered.

In the limit that the fraction of neutral atoms formed in states other than $n = 2$ is negligible, the cross section σ_{+m} can be determined by multiplying reported values of σ_{+0} by measured values of f, i.e., $\sigma_{+m} = f\sigma_{+0}$. That this approximation is reasonable at low energy (<2 keV) is discussed in Sec. III E. Results are shown in Fig. 7 and in Table II.

Error bars for σ_{+m} were calculated using the stated uncertainty in f and using ±25% uncertainty for the values of σ_{+0} found in Ref. 8. These results are in acceptable agreement with the results of Roussel *et al.* (taking the absolute error to be ±30%)[14-16] and Spiess *et al.*[8] The results of Donnally *et al.*[3] are in agreement at low energy, as far as magnitude is concerned, but the behavior with energy is not similar.

TABLE II. Metastable fraction f [ratio of H(2s) atoms to total number of atoms in the $n = 2$ states] for protons in a thin Cs-vapor target; the cross section σ_{+m} for electron pickup into the 2s state calculated using f and σ_{+0} (Ref. 8).

E (keV)	f	σ_{+0} (10^{-15} cm^2)	σ_{+m} (10^{-15} cm^2)
0.4	0.38 ± 0.045	~10[a]	~3.8
0.5	0.43 ± 0.025	10 ± 3	4.3 ± 1.3
0.55	0.37 ± 0.04	9.8	3.6 ± 1.0
0.6	0.35 ± 0.035	9.3	3.3 ± 0.9
0.75	0.24 ± 0.015	8.6	2.1 ± 0.6
1.0	0.26 ± 0.025	8.2	2.1 ± 0.6
1.35	0.25 ± 0.025	7.8	1.9 ± 0.5
1.5	0.27 ± 0.03	7.1	1.9 ± 0.5
2.0	0.24 ± 0.02	6.4	1.5 ± 0.4
2.5	0.24 ± 0.02	6.4 ± 1.6	1.5 ± 0.4
3.0	0.25 ± 0.025	6.3	1.6 ± 0.4

[a] By extrapolation.

C. Errors

Errors bars shown in Fig. 6 include all identified sources of uncertainty except those due to polarization (see IIID) and cascade (see IIIE). As the accuracy of the results for f does not rely upon absolute calibration of the Lyman-α detector, the primary source of uncertainty is the determination of the photon signals at $E = 0$ and for strong field, which is of the order of ±5%, including normalization to incident-proton current and to cesium-target thickness. Thus the over-all error is of the order of ±10%.

In order to avoid systematic error between measurements made with and without applied electric field, it is important that the spatial distribution of emitting particles be essentially unperturbed by the application of the field. We have already discussed in Sec. II the precautions taken to create a field which does not seriously perturb the proton trajectories. The mean distance traveled by an excited atom between its creation and its deexcitation is not identical for H($2p$) and H($2s$) but, even at the highest energy (3 keV), the difference is only of the order of a few millimeters. The difference in solid angle seen by the detector is therefore negligible. The only region where this would not be true is where the electric field changes direction (Fig. 4). Here there is a "hole" in the field, so that an H($2s$) atom could travel several millimeters further than an H($2p$) atom before deexcitation. If the hole were to occur just at the edge of the region seen by the PM, this would allow some H($2s$) atoms to leave the field of view of the PM without emitting a photon. The experiment was designed to avoid this situation (the circle in Fig. 4 shows the field of view of the PM).

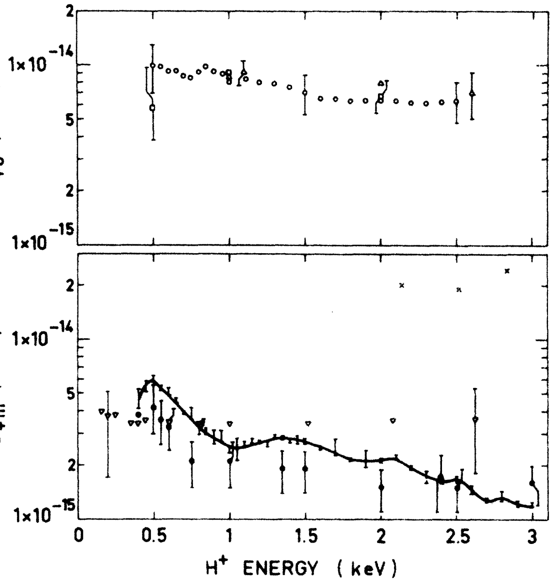

FIG. 7. Cross sections σ_{+0} and σ_{+m} for protons in cesium vapor. σ_{+0} is the cross section for electron pickup into any neutral state; σ_{+m} is the cross section for electron pickup into the metastable $2s$ state. All reported values of these cross sections are shown except those for σ_{+m} of Cesati *et al.* (1966), which are between 5 and 10×10^{-17} cm^2 in the given energy range. Symbols are as follows: ○, Spiess *et al.* (1970, 1972); △, Grüebler *et al.* (1970); □, Schlachter *et al.* (1970); ×, Sellin and Granoff (1967); ▽, Donnally *et al.* (1964); ●, present experiment. Small dots connected by curve, Roussel *et al.* (1973). The error bars for the data of Roussel *et al.* include only the relative error, so as to show the existence of the small maxima and minima; the curve is drawn for clarity.

A possible systematic error would occur if there were a source of electric field, with zero applied field, which was sufficiently strong to quench H($2s$) within view of the PM (> 50 V/cm). Possible causes are (a) static electric charge on insulating surfaces, (b) static charge on conducting surfaces, (c) the charge of the protons in the beam, (d) the charge of Cs$^+$ ions created in collisions, (e) motional electric field due to an ambient magnetic field, and (f) electric field due to Cs gauge, ion gauge, etc. The presence of a weak electric field is not easy to measure. We took all reasonable precautions to shield insulating surfaces and to thoroughly clean conducting surfaces. A measurement of Lyman-α signal with the system of quenching wires and their associated insulators removed showed essentially no change in signal from the usual measurement with quenching system in place. We calculated effects due to (c) and (d) and showed that they were negligible, using our experimental values. The only magnetic field present was Earth's field, which causes a negligible motional electric field. We took all possible precautions to shield the Cs gauge, the PM, the ion gauges, etc., and indeed verified that (except for the PM) their presence did not influence the Lyman-α signal. The PM, of course, could not be turned off, but it is far from the beam and shielded by a metallic diaphragm. We are therefore confident that there was no stray source of electric field sufficiently large to quench H($2s$) atoms in view of the PM.

As pointed out by Thomas,[34] we must tacitly assume that "the mechanism of the collision process is unaffected by the presence of an ambient electric field."

D. Polarization

A source of error not included in the stated error bars is that due to polarization of the Lyman-α radiation. It is well known that the Lyman-α

radiation from the 2p state formed by the collision of H^+ in various gases is polarized,[35,36] although this polarization has not been measured for H^+ in Cs vapor. It is further known that Lyman-α radiation from the 2s state quenched in an electric field is polarized.[27-31] The polarization of Lyman-α radiation from an atom in the 2p state could conceivably change in the presence of an electric field; this has not been measured.

In the present experiment we are unable to correct for the polarization of Lyman-α for several reasons: (i) The value is not known for $H^+ + Cs \to H(2p)$ in Cs; (ii) the electric field used for quenching changes in magnitude and direction in a complicated manner; (iii) there are two polarization axes—the electric field direction and the beam axis. We do note, however, that for a sufficiently strong electric field the Lyman-α signal is independent of the field value (within the error bars), which is to say that the effect of the field is small or independent of the field value for fields of the order of several hundred V/cm.

If we assume that the values of P for the various signals are not larger than ±0.3 [the value of P for H(2s) quenched in a moderate electric field], then, using formula (2), our measured signals are uncertain by ±10%:

$$S_{\text{true}} = S_{90^\circ}(1 - \tfrac{1}{3}P) . \qquad (2)$$

As P could either increase or decrease S_{true}, according to the sign of P, we add the errors independently, obtaining ±14% additional possible uncertainty in the value of f.

E. States other than $n=2$

Results presented in this article must be considered to include cascade effects. Measurements of cross sections for formation of states with n other than 2 have not been made, so that no correction is possible. Such effects are likely to be small, however, in the energy range considered.

Reaction (1) is nearly resonant; the energy defect is only −0.49 eV. The energy defect for the ground state is 9.7 eV. Thus direct formation of atoms in the ground state can be excluded at low energy. The energy defects for the states $n=3$ and 4 are −2.4 and −3.0 eV, respectively. It would be expected *a priori* that the cross sections for formation of atoms in the $n=3$ and 4 states are smaller than for the $n=2$ state, and, furthermore, that these cross sections have their maxima at higher energy than for $n=2$ (and thus are small at energies well below that at which the maximum occurs).

There is evidence that some formation of atoms in states with $n=3$ or higher does occur.[14-16] Note in Fig. 7 the curve for σ_{+m} measured by Roussel *et al.* Several maxima and minima are evident. The large maximum at 1.4 keV is believed to be an oscillation due to a maximum in the potential difference in the H-Cs system. The smaller maxima near 2.1, 2.5, and 2.8 keV are not regular in v^{-1}, and thus cannot be due to the above effect. These small maxima are believed to be cascading effects from states with $n=3$ and higher.

Consider the states with $n=3$. Only the 3p state decays to the 2s state; the 3s and 3d states decay to the 2p state (and then to the 1s state). Only 12% of the atoms in the 3p state decay, however, to the 2s state; the rest decay directly to the 1s state. However, these levels are Stark mixed by a weak electric field. For example, 2 V/cm parallel to the axis of quantization is sufficient to completely mix the $3p_{3/2}$ and $3d_{1/2}$ states[34]; likewise 58 V/cm to mix the $3s_{1/2}$ and $3p_{1/2}$ states. Thus a weak electric field is sufficient to enhance cascading into the 2s state.

It is not possible to find cross sections for formation of H atoms in states with $n=3$ and higher from the curve of Roussel *et al.* The maxima are not sufficiently clear, and the relative population of the various sublevels is not known (and cannot be assumed to be proportional to the statistical weights). Furthermore, the well-known n^{-3} law for population of highly excited states is probably not applicable for the low values of n considered.

The effect of cascades on the measurement of f is very difficult to estimate. The relative population of the 2s and 2p states by cascading could change according to the presence or absence of a field of the order of a few volts/cm. Furthermore, 88% of the decay which cascades via the 3p state gives a photon which was not detected at all.

We conclude, therefore, that our measurements of f cannot be corrected for cascade effects. However, it is extremely unlikely, at energies below 2 keV, that states other than $n=2$ are significantly populated.

We did consider measuring either Lyman-β or Balmer-α radiation in the collision zone, in order to determine population of the $n=3$ states. However, in the case of Lyman-β, our spectrometer was not suitable for the measurement, and in the case of Balmer-α, the signal-to-noise ratio was unsatisfactory.

F. Comparison with theory

We know of no theoretical calculation of the metastable fraction for the system $H^+ + Cs$. However, Vinogradov and co-workers[37,38] have calculated the cross sections σ_{+m} and σ_{+0} for this system using the Brinkman-Kramers approximation. They point out that it is necessary to take into account the possibility of electron capture from the internal shells. They find for σ_{+m} a calculated curve which appears to be roughly in agreement with the measurements of Sellin and Granoff,[13] and therefore larger than the present results. For σ_{+0} they find a curve which at high energies is much too high in comparison with the results of Refs. 5 and 6. They say that this is to be expected, as the Brinkman-Kramers method gives results which are too high at all energies. It appears that, at lower energies, the calculated value of σ_{+0} is of the order of 10^{-14} cm^2, which is in agreement with measured values; the theory, however, is of doubtful utility at low energies.

Perel and Daley[39] have obtained a semiempirical relationship between the velocity at the cross-section maximum and the absolute value of the energy defect for nonresonant charge-transfer collisions. This predicts the cross-section maximum near 1 keV for σ_{+0}. As the collision H^+ + Cs forms primarily $n=2$ states of H^0, the cross sections σ_{+m} and σ_{+2p} are predicted to have a maximum near 1 keV. The observed maxima lie at somewhat lower energy. Perel and Daley note also that "the large values of the cross section (σ_{+0}) at low energy indicated the dominance of transfer to excited states ($\Delta E = 0.50$ eV) over that to the ground state where $\Delta E = 10.70$ eV." Olson[40] has recently justified theoretically the semiempirical result of Perel and Daley.

Olson has also recently suggested[41] that, at low energies, the metastable fraction f is 50%, and at higher energies 25%. This would not be seriously inconsistent with the data, as the experimental apparent maximum is based on only one point at a proton energy of 0.4 keV. It would have been desirable to make measurements at lower H^+ energies, but this was not possible with the present apparatus.

G. Conclusion

We have measured the fraction f of metastable atoms, H(2s), relative to the total number of atoms in the $n=2$ states, for a single collision of a proton in a thin cesium-vapor target. At energies below 2 keV essentially all neutral atoms formed are in the $n=2$ states; thus f can be considered to be the fraction of metastable atoms in the neutral beam. The apparent maximum value of f is 0.43 ± 0.03 at a proton energy of 0.5 keV. For energies between 0.75 and 3.0 keV, $f = 0.25 \pm 0.01$, which is what would be expected if the sublevels of the $n=2$ state are statistically populated.

As it can be assumed that essentially all electron pickup is into the $n=2$ states at low energy, the fraction f can be multiplied by known values of the total charge-exchange cross section σ_{+0} to determine σ_{+m}, the cross section for electron pickup into the metastable 2s state. This cross section has a maximum value of $(4.3 \pm 1.3) \times 10^{-15}$ cm^2 at 0.5 keV, and decreases to $(1.6 \pm 0.4) \times 10^{-15}$ cm^2 at 3.0 keV.

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