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Charge-Exchange Collisions between Metastable Hydrogen Atoms and Iodine Molecules[†]

L. D. Knutson
University of Wisconsin, Madison, Wisconsin 53706

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Beams of hydrogen atoms in the (2*S*) metastable state and ground state were passed through an iodine vapor target at energies between 0.2 and 2.5 keV. Measurements of the positive-ion current after the iodine target indicate that the cross section for ionization of the metastable atoms by iodine is much larger than the cross section for ionization of ground-state atoms, especially at the lower energies. The results indicate that this reaction can be effectively used for production of polarized protons in a Lamb-shift polarized-ion source.

I. INTRODUCTION

Charge-exchange collisions of hydrogen atoms are of considerable interest because they provide insight into the atomic processes and because of various applications. Charge-exchange reactions are of importance for polarized-ion sources which make use of the Lamb shift.[1] In this type of device, fast metastable H(2*S*) atoms are produced by charge exchange of protons in cesium vapor.[2,3] Some of the hyperfine substates are then quenched by the application of suitable electric and magnetic fields, and the remaining metastable atoms, which have net nuclear polarization, are ionized by a reaction of the form $H(2S)+X \rightarrow H^- + X^+$.

To obtain substantial polarization, the cross section for ionization of metastable atoms must be much larger than the cross section for ionization of ground-state atoms. It is desirable that the charge-exchange cross section be larger than the cross section for all other processes which remove metastable atoms from the beam so that high beam intensity can be obtained.

Donnally and Sawyer[4] showed that the negative ions produced in the reaction $H + Ar \rightarrow H^- + Ar^+$ arise almost entirely from the H(2*S*) state when the hydrogen atoms are produced by charge exchange of protons in cesium with energies near 0.5 keV. This reaction has subsequently been used for the production of polarized negative hydrogen and deuterium ions for injection into Tandem accelerators.

For accelerators which accept positive ions, it is desirable to convert the H(2*S*) atoms directly to positive ions. Attempts to find a suitable gas or vapor target for this purpose have been made.[5,6]

A theoretical study by Lodge[7] had indicated that for alkali metals the cross section for the reaction $H + X \rightarrow H^+ + X + e^-$ should be much larger for metastable hydrogen atoms than for ground-state atoms. A preliminary study was undertaken at this laboratory on the reaction using cesium vapor as a target. At energies from 5 to 10 keV, the cross sections for ground-state and metastable atoms are not significantly different, and at lower energies the cross sections are too small to be useful. It appears that this reaction is not suitable for the production of polarized ions.

The conditions under which the reaction $H(2S) + X \rightarrow H^+ + X^-$ would be expected to have a large cross section can be determined by simple considerations of the pseudocrossing theory.[1,8] The ionization energy of the electron in the H(2*S*) atom is 3.4 eV. If the ionization energy of X^- (electron affinity of *X*) is smaller than 3.4 eV, the potential energy of $H^+ + X^-$ for infinite separation will be larger than the potential energy of $H(2S)+X$. As the particle separation decreases, the potential energy of $H^+ + X^-$ decreases because of the Coulomb interaction, so that at some separation *R* the potential-energy curves will cross. In pseudocrossing theory it is assumed that (under appropriate velocity conditions) the electron is exchanged near this point. For the cross section to be large, *R* should be large so that the two atoms have a high probability of passing within a distance *R* of each other. At the same time, *R* should be small enough that the wave functions of the two atoms overlap when the potential-energy curves cross.

Tables of the electron affinities of various elements and molecules are given in a review article by Pritchard.[9] Molecular iodine (I_2), which has an electron affinity of 1.7 ± 0.5 eV,[10] was chosen for the present study. Iodine is convenient to use since it is readily available and has a vapor pressure high enough to form vapor targets without heating.

II. APPARATUS

The apparatus used is shown schematically in Fig. 1. Protons were extracted from a duoplasmatron at energies which were varied between 0.2 and 2.5 keV. The ions entered a heated cesium tube of 15-cm length and 1.2-cm diam. Cesium vapor entered the tube from a reservoir heated to approximately 220 °C through a bakeable stainlesssteel valve which was used to control the cesium density. Cesium leaving either end of the tube was trapped by a Freon-cooled copper baffle.

The beam emerging from the cesium tube consisted of hydrogen atoms in the $1S$ and $2S$ states along with H^+ and H^- ions. The charged components were removed by passing the beam between a pair of electrostatic deflection plates 15 cm long and 7 cm apart. The presence of an electric field reduces the lifetime of the $H(2S)$ state[11]; thus some of the metastable atoms were quenched in the deflection plates. A field strength of 3 to 7 V/cm (depending on beam energy) is sufficient to remove most of the ions from the beam while quenching less than 2% of the metastable atoms.

After passing through the deflection plates, the fast neutral atoms entered an iodine charge-exchange tube 30 cm long and 0.9 cm in diameter. This tube is connected by a copper tube to an iodine reservoir which is outside the vacuum system. The iodine density in the tube was regulated with a needle valve. No heating was necessary to obtain sufficient iodine density. A second needle valve allowed argon to enter the tube for production of negative ions. A liquid-nitrogen cold trap prevented the iodine from contaminating the vacuum pumps.

The H^+ ions emerging from the iodine charge-exchange tube were momentum analyzed by a 24° magnetic deflection and collected in a suppressed Faraday cup. The remaining H atoms were collected in a neutral detector, which consisted of a polished-copper beam stop surrounded by a ring which was biased to + 300 V for collection of secondary electrons emitted from the copper surface.[12]

Several defining apertures (see Fig. 1) were used to limit the diameter of the beam to 0.5 cm. These prevented the beam from striking the walls of the charge-exchange tubes and allowed the charged ions to be removed from the beam by smaller electric fields.

The residual gas pressure near the cesium cell was due mainly to hydrogen from the duoplasmatron and was typically 2×10^{-5} Torr. In the iodine region, the pressure was typically 3×10^{-6} Torr.

III. DATA

The quantity which we would like to determine is the ratio of the cross section for ionization of metastable hydrogen atoms by iodine (σ_{m+}) to the cross section for ionization of ground-state atoms by iodine (σ_{g+}). If f is used to denote the fraction of the atoms which enter the iodine charge-exchange tube in the metastable state, one can determine σ_{m+}/σ_{g+} by measuring the positive-ion current leaving the tube for two known values of f.

If N hydrogen atoms per unit time are incident on an iodine target, and if l is the effective path length of the hydrogen atoms in the iodine vapor which contains ρ molecules per unit volume, the H^+-ion current leaving the tube will be

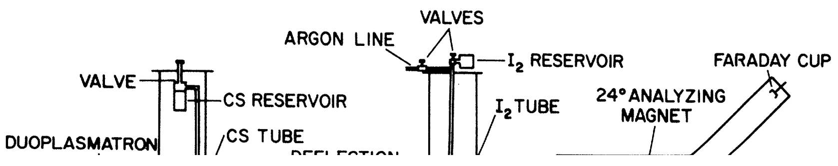
$$I = eNpl[f\sigma_{m+} + (1-f)\sigma_{g+}] \ ,$$

where e is the elementary charge. The equation holds provided the iodine density is low enough that the number of multiple collisions is insignificant.

The experimentally measured currents contained a contribution from sources other than charge exchange in the iodine. This background current, which was measured and subtracted in all cases, is believed to be due to ionization of neutral hydrogen atoms in the residual gas and to the positive beam, which was not completely removed by the deflection plates. The background current was measured by closing the valve to the iodine reservoir and was found to range from 4% of the total current at low energies to 50% at high energies.

In this experiment f attains its maximum value F when the cesium target thickness is low enough that collision quenching of the metastable atoms in the cesium vapor is insignificant and when no quenching occurs in the deflection plates. The H^+-ion current which would be obtained under these conditions is

$$I_1 = eNpl[F\sigma_{m+} + (1-F)\sigma_{g+}] \ .$$



In practice, I_1 was determined in the following manner: The current in the Faraday cup was measured with just enough voltage on the deflection plates to remove most of the charged ions from the beam; this measured current was then corrected for the small number of metastable atoms quenched in the deflection plates and for the background current.

The H^+ ion current obtained with $f = 0$ is

$$I_2 = eN\rho l\sigma_{g+} \ .$$

This quantity was determined by noting the current with enough voltage on the deflection plates to quench virtually all the metastable atoms. The measured current was corrected for the few remaining metastable atoms and for the background current.

The ratio of I_1 to I_2, which will be called Q, is

$$Q = I_1/I_2 = [F\sigma_{m+} + (1-F)\sigma_{g+}]/\sigma_{g+} \ . \qquad (1)$$

It is desirable that Q be a large number. The measured values of Q are shown in Fig. 2. The values of Q are found to be quite large, especially at the lowest energies. They start at 125 for an energy of 0.2 keV and then decrease rapidly and monotonically with increasing energy.

Donnally and Sawyer[6] measured F and found it to be roughly 0.25, independent of energy, up to 1.5 keV. Since Q and F are known, one can solve Eq. (1) for σ_{m+}/σ_{g+}. The ratios of the cross sections are surprisingly large, starting at 500 for the lowest energy and then decreasing to 9 at 1.5 keV. The accuracy of σ_{m+}/σ_{g+} obtained in this manner is somewhat doubtful because of the difficulty in measuring F.

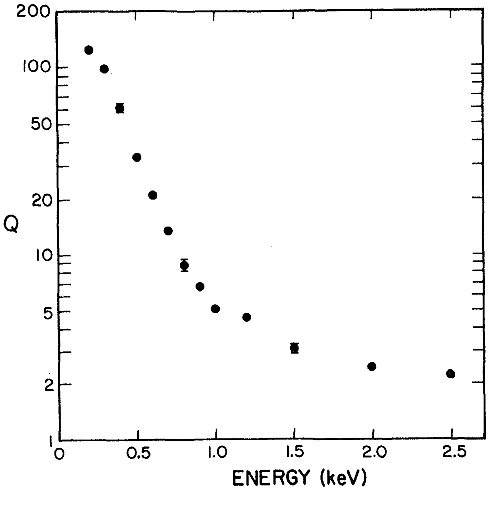


FIG. 2. Semilogarithmic plot of Q as a function of hydrogen atom energy. Typical error bars are shown.

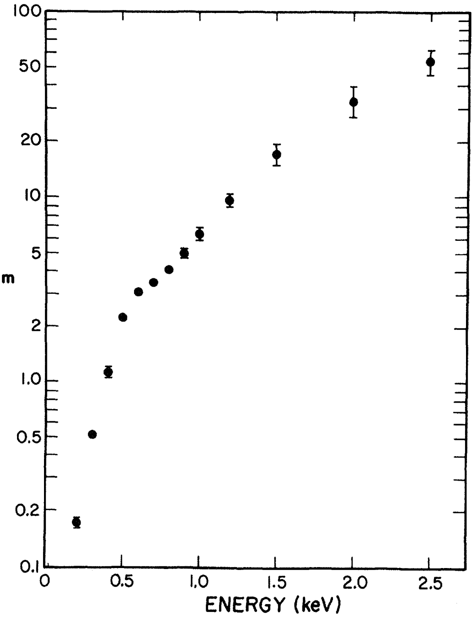


FIG. 3. Semilogarithmic plot of the maximum positive-ion current obtainable from the sequence of reactions $H^+ + Cs \rightarrow H + Cs^+$ and $H + I_2 \rightarrow H^+ + \cdots$. The data are normalized to the maximum negative-ion current obtained from the sequence of reactions $H^+ + Cs \rightarrow H + Cs^+$ and $H + Ar \rightarrow H^- + Ar^+$ at an incident proton energy of 0.5 keV.

IV. APPLICATION

In order to obtain an indication of the beam intensity which can be expected from a Lamb-shift polarized-ion source using the cesium and iodine charge-exchange reactions, the restriction of low density in the charge-exchange tubes was dropped and the following measurement was made. With just enough voltage on the deflection plates to remove most of the charged ions from the beam, the iodine and cesium densities were adjusted to obtain the maximum current in the Faraday cup. The background current was measured and subtracted as described in Sec. III, and the result is denoted by I_m.

Measured values of I_m are shown in Fig. 3. The error bars indicate the reproducibility of the results. The data are normalized to the negative-ion current obtained in the same manner when the iodine was replaced by argon and the proton energy fixed at 0.5 keV. The reason for this normalization is that the argon charge-exchange reaction is normally used in Lamb-shift negative-polarized-ion sources.

The portions of the current in the Faraday cup which were due to metastable atoms, ground-state atoms, and background were determined with the iodine and cesium densities adjusted for the maximum current. With these numbers it is possible to calculate the proton polarization which would be obtained in a Lamb-shift source under the same conditions. It was assumed that $\frac{1}{2}$ of the metastable atoms would be quenched and that the remaining metastable atoms would obtain complete nuclear polarization. This can be done, for example, by a scheme which was proposed by Sona[13] and verified experimentally by Clegg *et al.*[14] It was further assumed that in the process the metastable atoms which were quenched would be completely polarized in the opposite direction. The background and the atoms which were initially in the ground state were assumed to be completely unpolarized. The calculation at a proton energy of 0.7 keV indicates that a polarization of 0.7 can be expected.

The large values of Q and L indicate that the iodine charge-exchange reaction can be effectively used for production of positive polarized ions.

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