Charge Production in Collisions Between Hydrogen Atoms, and He, Ne, Ar, and H_2 in the Energy Range From 50-1000 eV*

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Experimental results on negative-charge production in collisions of hydrogen atoms with He, Ne, Ar, and H_2 are reported. For the three noble gases a rather smooth rise of the cross section with energy was found, absolute values ranging from about 10^{-18}–10^{-16} cm^2. For collisions of H on H_2, an interesting structure appears which seems to extend structure indicated in earlier high-energy measurements. From extrapolations of our results and comparison with high-energy stripping measurements, a trend in the ratio between target ionization and projectile stripping from He to Ne to Ar is indicated.

1. INTRODUCTION

So far, ionization reactions occurring in collisions between neutral atoms or molecules have attracted interest mainly for applicational reasons. The theory of these reactions still is in a rather rudimentary stage. From simple theoretical models, the corresponding cross sections are expected to rise continuously with energy from the ionization threshold up to a maximum at an energy given by the Massey criterion[1] and then to fall steadily. At small energies, this basic energy dependence may be modified by molecular effects containing information on subreactions and the structure of the the intermediate compound molecule. Experimentally, for beam energies above a few keV, some material has been accumulated for a number of projectiles of generally gaseous origin and for alkali atoms.[2,3] More recently, some other heavy metal projectiles were investigated.[4] In these experiments, generally, the stripped projectiles were measured, and in most cases the expected general behavior of the cross sections was found. At lower energies, down to the ionization threshold where the scattering of the projectiles during these reactions becomes appreciable, only a few measurements have been reported of negative-charge production in encounters between various projectiles and gases.[5–8] As expected, also in most of these combinations a fast smooth rise of the cross section with energy was revealed. In a number of cases, however, and most pronounced for Ar + Ar, some additional structure appeared superimposed on the general rise. So far, probably mainly due to the complexity of these reactions, no theoretical analysis of these structures beyond pure suggestions has been published.

These reactions and structure would be best accessible for theoretical treatment if at least one of the collision partners were a neutral hydrogen atom. However, up to now no experimental data on collisional ionization involving hydrogen atoms exist in the low-energy region. This gap seems to be due mainly to the non-existence of a gas allowing sufficiently resonant charge transfer with protons into the hydrogen ground state. Only for this case, the scattering of the resulting hydrogen atoms was expected to be sufficiently small to allow a deduction of the absolute intensity of the neutral beam from the ion current and a known charge transfer cross section.

In an earlier paper,[9] we reported on measurements of the angular distribution of hydrogen atoms produced in the nonresonant reaction

$$H^+ + O_2 \rightarrow H + O_2^+ .$$

Using these results, we have produced a neutral-hydrogen beam of known intensity and used it for measurements of total negative-charge production in collisions of these neutrals with He, Ne, Ar, and H_2 at particle energies between 50 and 1000 eV. In this, the experimental arrangement was similar to the one described in Ref. 5, using parallel-plate geometry to extract the produced negative charge from the reaction region. For the three noble gases a rather smooth rise of the cross section with energy was found, whereas the case $H + H_2$ exhibits an interesting structure which seems to extend structure indicated in older high-energy measurements. From extrapolations of our results to higher energies and comparison with stripping measurements of the hydrogen projectile, a steady increase of the ratio between projectile stripping and target ionization from He to Ne to Ar appears indicated. Also, some results on secondary-electron emission from copper under hydrogen impact are given.

2. EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown in Fig. 1. As in the charge transfer measurements,[9] with minor modifications, the ion beam as described in detail by Fite *et al.*[10] was used. In short, a proton beam was extracted from a duoplasmatron source, accelerated, focused, mass analyzed at 1–2 keV, and then decelerated to the desired energy. As in Ref. 9, the angular spread of the beam entering the charge transfer cell was limited by two apertures, 1 mm wide and about 8 cm apart. Generally, these were at ground po-

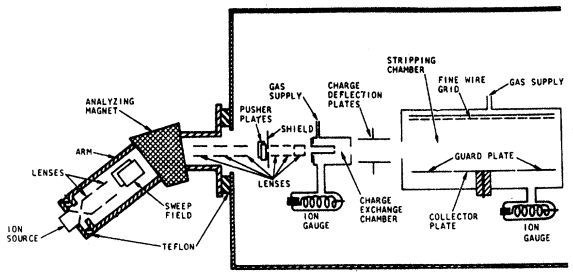

FIG. 1. Experimental arrangement.

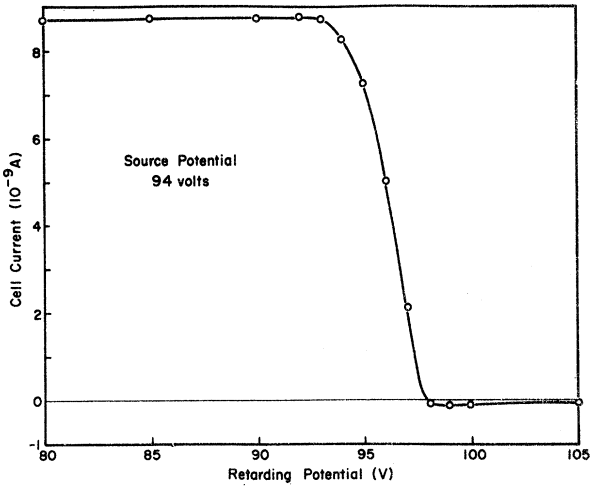

FIG. 2. Energy distribution of proton beam in charge transfer cell measured by retardation method.

tential as the cell itself, but insulated from it. The charge transfer cell also had already been used in the small-angle measurements of Ref. 9. Using an effective cell length of 16 mm and an exit hole of 4.5 mm, all hydrogen atoms resulting from a charge transfer in the cell and scattered less than 7 deg could emerge. Using again 2–4 $\times 10^{-4}$-Torr oxygen, a few percent of the original proton beam was neutralized.

Behind the cell exit, the ion beam was deflected onto a ground shield and measured. To determine the contributions from secondary electrons emitted from this shield, the cell exit was closed by a flap and the total current entering the cell measured. Thus a correction factor α for the current measured at the mentioned ground plate was determined. It varied from about $\alpha = 1.05$ at 60 eV to $\alpha \approx 1.9$ at $E = 1000$ eV. Under a wide variety of conditions, for any given energy, this factor was constant to better than 5%. As could also be inferred from other measurements, electrons produced in other parts of the beam duct contributed less than that to the total current entering the cell. The secondary-emission factors for impact of protons inferred from this correction factor rather closely coincide with the results of direct secondary-emission measurements for impact of neutral hydrogen atoms given in Fig. 12. Over most of the investigated energy range, no difference between both secondary-electron-emission coefficients would be expected.

To determine the energy spread of the ions entering the cell, a retarding potential was applied to the cell and the current measured as a function of this potential. A typical result for a source potential of 94 V is shown in Fig. 2. For various beam settings, the indicated surplus of the average beam energy varied between 2.5 and 4 eV.

The collision chamber used for the actual measurements (in the following is called stripping chamber) was very similar to the one described by Utterback and Miller.[5] The arrangement of the electrodes extracting the charges from the collision region between the plates is shown in Fig. 3. The guard plate, the negative plate, and the grid were 20 cm long and 12 cm high. The collector plate itself was 10 cm long and about 6 cm wide. The distance between the collector side and grid was 4.5 cm. The grid was made of 0.0007-in. gold-plated tungsten wire spaced 0.13 in. This grid was mounted 1.0 cm from the back plate. All surfaces were gold plated. During the measurements, the back plate was held at +30 V with respect to the grid which proved sufficient to prevent secondary electrons produced at the plate from reaching the collector. A few measurements also were made with a wire spacing of 0.2 in., but no essential difference was found.

This electrode assembly was contained in a box of similar dimensions. The neutral beam entered the chamber through an aperture 8 mm wide admitting all neutral particles which were scattered less than 3 deg in the neutralizing charge transfer. In front of this aperture, two collimating tubes were mounted shielding the entrance hole from particles coming from places other than the charge transfer chamber. The fraction of hydrogen atoms neutralized in this cell which entered the stripping

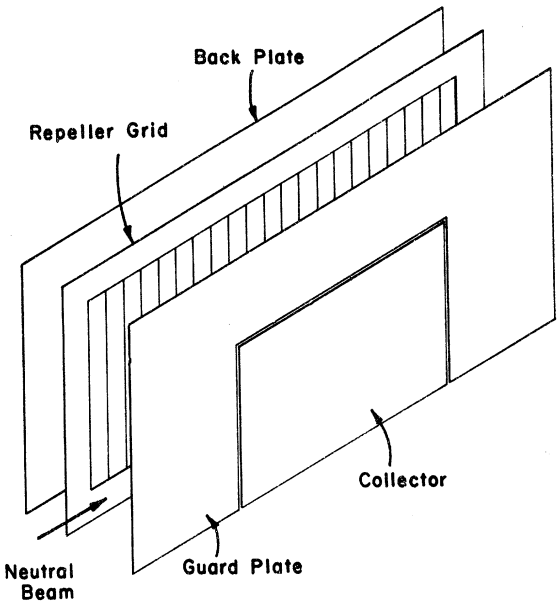

FIG. 3. Ionization chamber.

chamber as derived from our earlier scattering measurements is shown in Fig. 4 together with the corresponding fractions for other maximal scattering angles.

The target gas was introduced into the stripping chamber through the lid by means of combined leak and on-off valves and could escape only through the front aperture. The resulting pressure gradients in the cell thus were negligible. As in the neutralization chamber, the pressures were measured with an ionization gauge. For absolute measurements, both gauges were calibrated agains a capacitive MKS Baratron. In the stripping chamber, generally, pressures of a few times 10^{-4} Torr were used. The background pressure in the general vacuum tank containing both reaction chambers generally was a few times 10^{-6} Torr whereas in the ion-acceleration and mass-analysis region pressures of a few times 10^{-5} Torr prevailed.

The ionization currents were measured with a Cary vibrating Reed Electrometer, using 10^{10}–10^{11} Ω as load resistor.

3. MEASUREMENTAL PROCEDURES

At the beginning, the collection efficiency of the stripping chamber was determined for each target gas. In this, for a fixed potential difference of 30 V between the grid and the back plate, the ionization current onto the collector plate was measured as a function of the grid potential. The results were very similar to that of Utterback.[5] A typical curve is shown in Fig. 5. Higher values of the mentioned potential difference between grid and back plate did not influence these curves. In the final measurements a grid potential of 180 V was selected. At this level no efficiency correction had to be applied to the final results. The microphonic noise generally still was negligible.

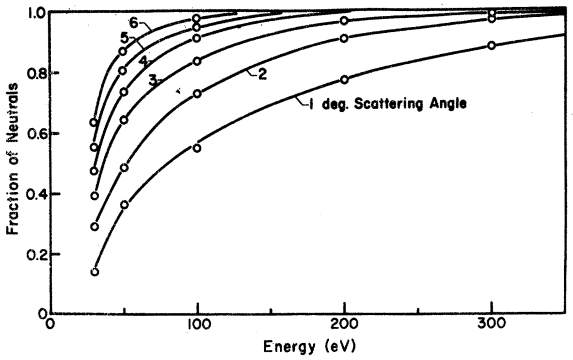

FIG. 4. Fraction of neutral hydrogen atoms with scattering angles less than indicated.

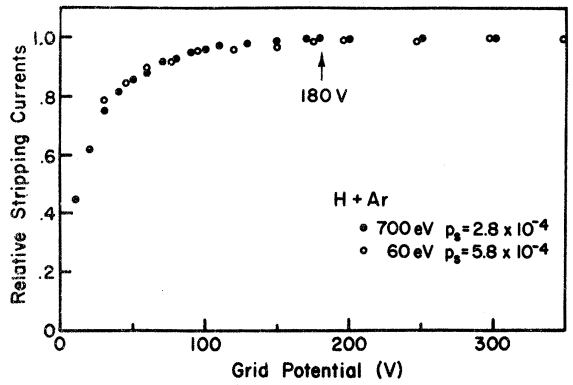

FIG. 5. Collection efficiency of stripping cell as function of extraction voltage.

For a measurement of the real ionization currents, a background determination had to be made. In addition to generally negligible leakage currents, this background, to a first order, was due to ionizations in collisions of the fast beam neutrals with the background gas in the stripping cell and to neutral atoms or electrons produced either outside the charge transfer cell (collisions of the fast ions with the gas molecules or metal parts) or in its background gas. To separate the various contributions, the apparent ionization currents were measured for all combinations of the possible choices: target gas on-off valve, charge transfer gas on-off valve, and ion beam admitted to the charge transfer cell – or repelled at its entrance aperture. By properly combining the various results, the various first-order background contributions were deduced. As to be expected, these contributions depended strongly on the tuning of the ion beam. In the final measurements, the total background corrections generally were smaller than 20%. Real ionization currents derived by different combinations of the mentioned submeasurements, differing only by second-order background contributions, generally agreed to better than 5%.

Using the real ionization currents thus derived, their dependence on the pressure in the stripping cell was measured for each gas and a number of beam energies. For argon, Fig. 6 shows the resulting pressure dependence of the apparent cross section, i.e., the deduced real ionization currents divided by the ion current and the stripping pressure. For the other target gases this dependence was even smaller. The necessary corresponding corrections to the later experimental results never exceeded 10%.

After these checks for a given target gas, the energy dependence of the cross section was investigated. To this end, the ionization currents were measured for various beam energies using fixed pressures in charge transfer and ionization chamber. From these, the energy dependence of the cross section was derived in the usual manner applying the mentioned secondary emission correction to the measured ion current, using the scattering factor of Fig. 4 and the total charge transfer cross sections as measured by Stebbings *et al.*[11] and by Koopman.[12] Since the results of the latter references differed slightly, the mean value of both results was taken.

At the end, the relative cross sections of the gases and their absolute values were determined for a fixed beam energy of 150 eV. For this purpose, the gas inlet of the ionization chamber was

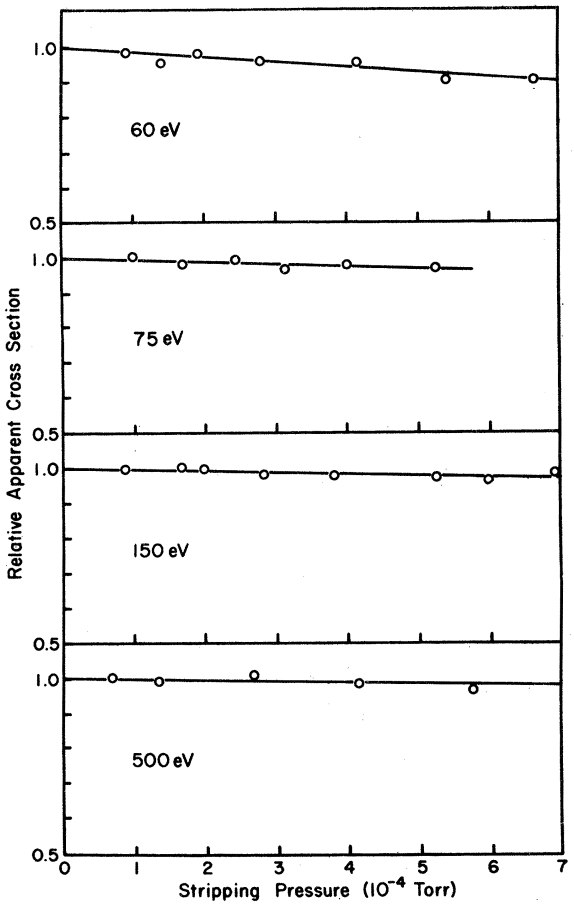

FIG. 6. Pressure dependence of apparent ionization cross section for argon.

connected to a manifold which, through on-off and leak valves, was connected to supplies of all 4 gases; and alternately all gases were admitted to the chamber. Before and after these measurements, the calibration curves of both ionization gauges for the various gases and also the calibration of the current meters were checked and corresponding corrections applied.

4. ERROR ESTIMATES

Based on the described experimental procedures, we would expect the following limits for the various error contributions:

Background determination	
high energies	< 5%
low energies	5–10%
Charge collection in ionization chamber	< 5%
Secondary emission effects in current determination	
$E < 300$ eV	< 5%
$E > 300$ eV	5%
Charge transfer scattering	
$E > 100$ eV	< 5%
$50 < E < 100$ eV	10–5%
Secondary electron effects in ionization chamber	< 5%
Pressure dependence	< 5%
Pressure measurements (each)	5%
Current meters	5%
Total charge transfer cross sections	5–10%
Miscellaneous	5%

In total, we would expect the relative cross-section values among the various gases to be accurate within about 10%. For the absolute values at 150 eV, we estimate an uncertainty of about 15%. Relative to this point, the energy dependence of the cross sections should be off not more than about 10% at the limits of the investigated energy ranges.

To these errors have to be added the possible deviations of our energy scale. These may arise from instrument calibration (2–3%), fluctuations of the beam energy in the charge transfer cell due to surface charges and duoplasmatron conditions (≈ 2 eV) and our ignorance about the energy transfer in the neutralization reaction[9] (1.5 eV).

As a check on some of these error estimates, in the course of these experiments, the data of Utterback and Miller[5] were redetermined using their charge transfer cross sections. As shown in Fig. 7, there is reasonable agreement between both results, our values generally being smaller by about 10%, which can easily be accounted for by the accuracy of both experiments. It seems interesting to note that this agreement exists in spite of our use of a duoplasmatron source with anode voltages around 100 V where the admixture of vibrationally or even electronically excited N_2^+ ions cannot be excluded.

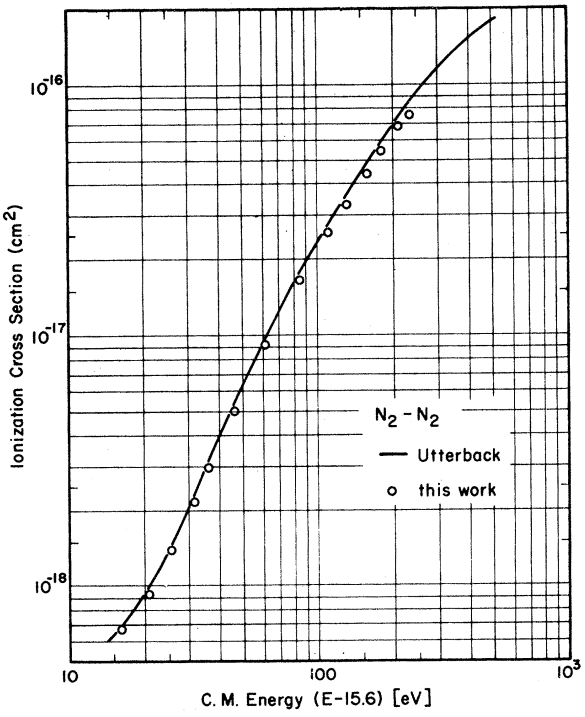

FIG. 7. Negative-charge production cross section for collisions between two nitrogen molecules.

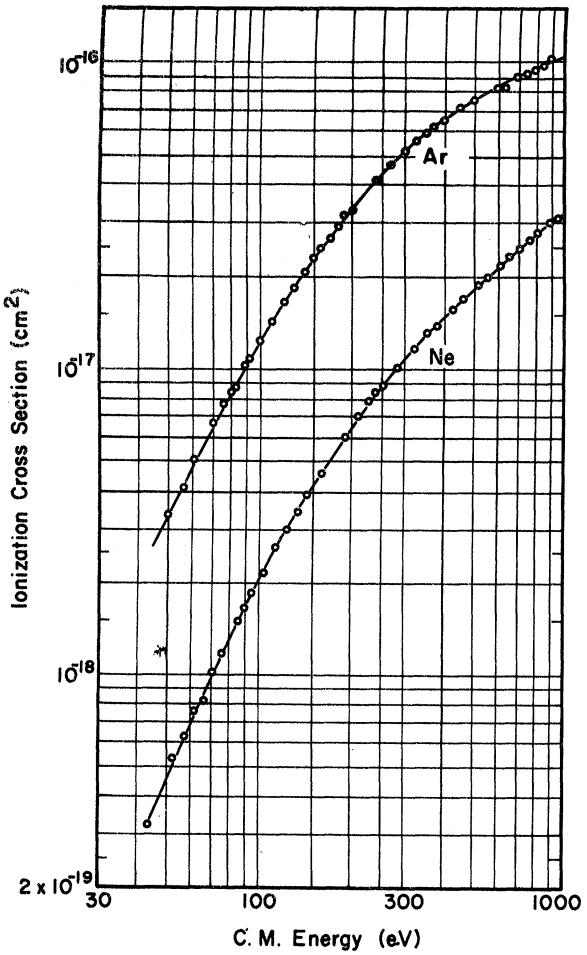

FIG. 8. Negative-charge production cross section for collisions of hydrogen atoms with neon and argon.

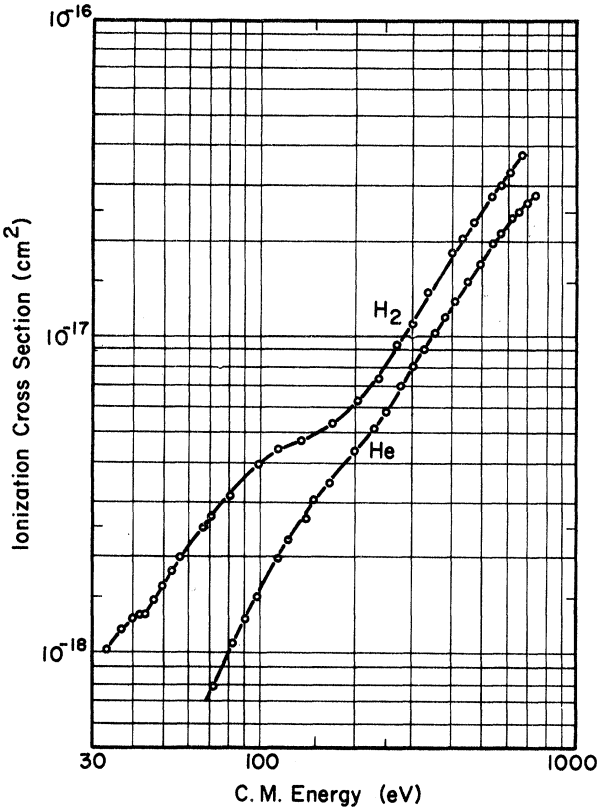

FIG. 9. Negative-charge production cross section for collisions of hydrogen atoms with helium and molecular hydrogen.

5. RESULTS AND DISCUSSION

Results of the above measurements are shown in Figs. 8 and 9. The curves for Ne and Ar show the steep smooth increase with energy generally expected for nearly adiabatic transitions. In view of the lack of any good theory of these reactions, a comparison with Firsov's[13] formulas (15) and (16) was tried, though the large mass ratio of the two collision partners is outside the range claimed by Firsov. In both cases, his predictions come out considerably larger and slower rising than our results. We tried to fit his curves by proper selection of his parameters σ_0 and u_0, i.e., of his excitation energy E_0. In the case of Ne, values $\sigma_0 \approx 2\times10^{-16}$ cm^2 and $E_0 \approx 22$ eV were obtained instead of the theoretical values 6.7×10^{-16} cm^2 and 13.5 eV. For argon, the fitting yielded $\sigma_0 \approx 4.6\times10^{-16}$ cm^2 and $E_0 = 40$ eV, comparing with Firsov's values 5.1×10^{-16} cm^2 and 13.5 eV. However, in both cases, the fitted Firsov curves still bend appreciably sharper and the reached agreement with our curves was not very convincing.

In the cases of He and H_2, some structure appears superimposed on the general rise of the cross section. Speculatively, the slight bump in the curve for He could be explained as a superposition of separate smooth curves for the stripping of the hydrogen projectile and for the ionization of the helium target. However, this explanation would necessitate cross sections for the ionization of the helium atom which are clearly incompatible with higher-energy results of Barnett and Stier[14] for the stripping of helium atoms in molecular hydrogen. Likewise, no supportable explanation can be offered for the pronounced structure of the curve for $H+H_2$. However, this structure seems to be an extension of some structure already indicated in earlier high-energy measurements of the stripping of hydrogen atoms in hydrogen gas by Stier and Barnett[15] (compare Fig. 11). If we put, though somewhat arbitrarily, the "maxima" of this structure at 60, 170, and 4000 eV, one would obtain approximately equidistant $(1/v)$ values for these maxima which, if true, would suggest an oscillatory origin of this structure.

So far, no measurements of total charge production caused by neutral hydrogen atoms in gases have been published for a comparable energy range. Thus comparison can be made only with higher-energy measurements of cross sections for the stripping of hydrogen atoms in various gases, as plotted in Figs. 10 and 11. Measurements

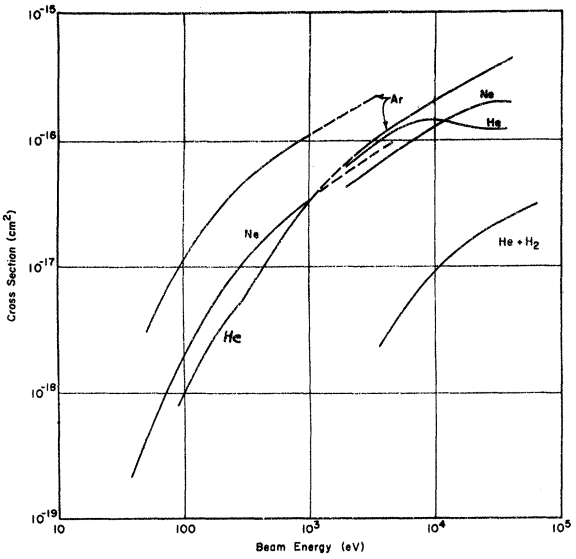

FIG. 10. Comparison with high-energy stripping data for helium, neon, and argon [averaged from the results of Stier and Barnett (Ref. 15) and Williams (Ref. 16)].

of this type have been done by Stier and Barnett[15] for energies above 4 keV on a number of gases including the ones investigated by us, and more recently by Williams[16] for energies down to 2 keV and the same gases, and by Mc Clure[17] on H and H_2 down to 1.25 keV. As already analyzed by Williams and McClure, these measurements agree within about 10% for energies above 10 keV. For lower energies, the H_2 results of Stier, Barnett, and McClure still agree similarly well. With respect to these, the H_2 results of Williams decrease faster with decreasing energy, probably[17] due to a smaller upper limit for the scattering angle of the detected protons. For the noble gases, the results of Stier, Barnett, and Williams again agree within about 10–20% down to 4 keV. In Fig. 10 an average of the noble-gas results of these two papers is compared with our results, whereas in Fig. 11 only the results of McClure are used for comparison. (Notice that in this case our data as well as theirs are plotted for beam energies rather than center-of-mass energies.)

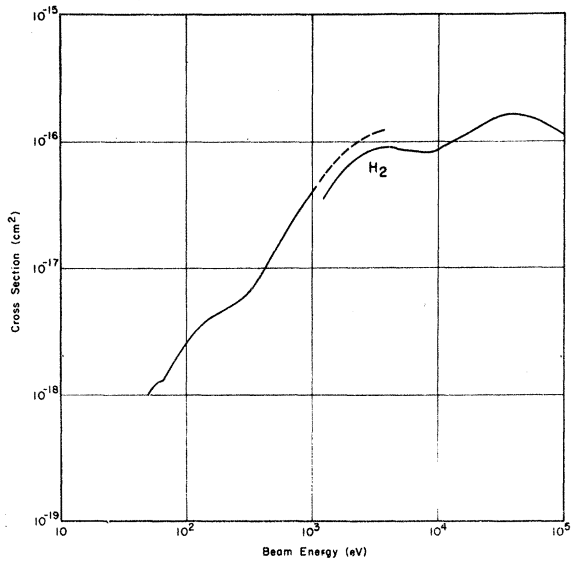

FIG. 11. Comparison with high-energy stripping data for molecular hydrogen from McClure (Ref. 17) and Stier and Barnett (Ref. 15).

As shown, a good interpolation between both sets of data can be obtained for helium, including both the absolute magnitude and the slopes of the cross section. The small difference shown in Fig. 10 around 2–4 keV very closely matches the already mentioned results of Barnett and Stier[14] for the stripping of He atoms in molecular hydrogen. The corresponding curve, also shown in Fig. 10, gives the cross section per target atom which would seem a reasonable first approximation of the cross section for stripping of helium atoms by atomic hydrogen. Taking the same relative velocities, this cross section is about one tenth of ours in the questionable energy region, and this ratio only slightly changes between hydrogen beam energies of 1 and 4 keV.

For neon, a smooth extrapolation of our curve into the region around 2–4 keV very well meets the slope of the high-energy measurements. However, a larger difference appears to exist in the absolute values. In the case of argon, the end slope of our curve agrees quite well with the slope of the almost linear part of the high-energy curve above 4 keV, and a corresponding extrapolation results in a cross section ratio of almost 2. For energies below 4 keV, the stripping curve falls somewhat faster than suggested by our extrapolation. However, this part has only been measured by Williams and this droop may very well be caused by the mentioned limitations[17] of the accepted scattering angles which should be more pronounced for heavy target gases.

Of course, all these extrapolations contain a certain degree of arbitrariness. However, they appear quite natural with respect to the measured curve below 1 keV and also are not unreasonable in view of the already existing similarity of the slopes in both curves. If true, these extrapolations would indicate that, at these beam energies, the probability ratio between ionization of the target atom and stripping of the projectile increases from helium to neon to argon. More experimental data appear necessary to test the general significance of such a trend and its origin. In the mentioned sequence, the ionization energies of the target atoms decrease and also the number of available electrons increases, and both of these influences could be reasonably expected to establish this trend. On the other hand, the absolute cross sections do not exhibit any comparable regularity in the keV range. Only at low energies a similar trend may prevail.

For the case of hydrogen, again the slopes of the adjoining curve ends meet quite well, whereas the absolute values differ by about a factor of 1.6 which

again falls into the range exhibited by the other gases. Using Williams's results[16] for comparison, the gap between the corresponding slopes would become somewhat larger, and the factor between the absolute values would increase to about 2. This value would still fit into the general range, whereas the absolute values of the negative-charge production at small energies does not fit into the mentioned He-Ne-Ar pattern.

In all our measurements, it is assumed that the hydrogen atoms are in the ground state. Because of the considerably smaller energy defect, the charge transfer may be expected to occur mainly to the ground state, particularly at small beam energies. The small percentage of hydrogen atoms possibly produced in an excited state will be considerably reduced due to the short radiation times and the existing electric deflection field quenching the metastable states.[18]

In the course of the experiments also the secondary-electron-emission coefficient has been determined for a 45-deg impact of the neutral hydrogen atoms on a copper surface only cleaned with hydrochloric acid and alcohol. The result is shown in Fig. 12.

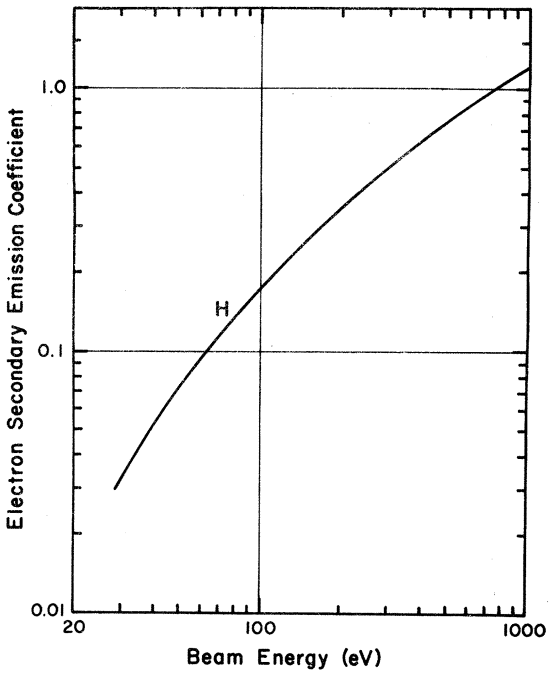

FIG. 12. Secondary-electron-emission coefficient for impact of hydrogen atoms on a copper surface under an angle of 45 deg.

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