Formation of Negative Hydrogen Ions in the Passage of Protons through Thin Metal Foils

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By means of a dual capability mass spectrometric apparatus, the ratio of negative hydrogen ions to protons has been determined for a beam of protons with energies in the interval from 11.5 to 28 kev which have passed through thin foils of Be, Al and Cu. It has been found that approximately 10 percent of the protons which fall on a thin Be foil are changed into negative hydrogen ions.

INTRODUCTION

WITH the passage of fast positive ions through matter, collisions between the ions of the beam and the atoms of matter occur which are accompanied by a capture or loss of electrons by the ions. As a result the composition of a beam which has traversed several layers of matter can be substantially different from that of the incident beam. Thus, for example, if the incident beam consists of singly charged ions, then the traversing beam can contain, in addition to the singly charged ions, multi-charged ions, neutral atoms and singly charged negative ions. Negative ions will occur in the traversing beam only in the case that the corresponding atoms pass a positive electron affinity.

In the case of protons passing through matter, the processes of electron capture or loss imply that the traversing beam will contain neutral atoms and negative hydrogen ions in addition to protons. Negative hydrogen ions will be produced in a beam as a result of protons capturing two electrons in a single collision event with the particles of the medium* , or as a result of two successive collisions with the capture of one electron in each collision.

The percentage of neutral atoms in a proton beam which was passed through thin foils of Be, Al, Ag, and Au was studied by Hall[2] for protons with energies ranging from 20 to 400 kev. In this work the presence of negative hydrogen ions in the emergent beam was ignored for small proton energies. For this reason the ratio of hydrogen atoms to protons as determined by the author for these energies must be corrected.

The first mention of the formation of negative hydrogen ions in the passage of a proton beam through a thin aluminum foil is contained in a work by Ribe[3] dedicated to the study of the neutralization of protons in hydrogen. With the idea of evaluating the influence of the presence of negative hydrogen ions on the results of his experiments, this author performed measurements for protons with energies from 35 to 90 kev. At 35 kev, the percentage of ions H_1^- reached 3 percent of the number of protons in the emergent beam, while at 90 kev, within the limits of experimental errors (of the order of 0.1 percent), the presence of negative hydrogen ions was not detected in the emergent beam.

Some data on the percentage of negative deuterium ions in a beam of deuterons which had passed through an aluminum foil of thickness 0.01 milligrams/cm^2 are presented by Alvarez[4] .

One can conclude from the data that, for deuterons with energies of 10 kev, the number of negative deuterium ions formed in a beam passing through metal foils reaches as much as 26 percent of the number of deuterons incident on the foil. Finally, in a recently published short report by Phillips[5] some results are presented on investigations of the equilibrium composition of proton beams with energies from 4 to 140 kev which have passed through thin foils of Al, Al_2O_3 , SiO and Au.

It appears from the preceding short summary that the study of the concentration of negative ions in a proton beam which has passed through metal foils and its dependence upon proton energies and the nature of the foil is not sufficiently complete. In this connection we undertake to report experiments performed with the aim of studying the formation of negative hydrogen ions for proton beams

* An essentially similar process was discovered by us in the passage of a proton beam through hydrogen[1].

[1] Ia. M. Fogel, L. I. Krupnik , and B. G. Safranav, J. Exper. Theoret. Phys. USSR **28**, 589 (1955); Soviet Phys. 1, 415 (1955)

[2] T. Hall, Phys. Rev. **79**, 504 (1950)

[3] F. L. Ribe, Phys. Rev. **83**, 1217 (1951)

[4] L. W. Alvarez, Rev. Sci.Instr. **22**, 705 (1951)

[5] J. A. Phillips, Phys. Rev. **91**, 455 (1953)

which have passed through thin foils of Be, Al, and Cu.

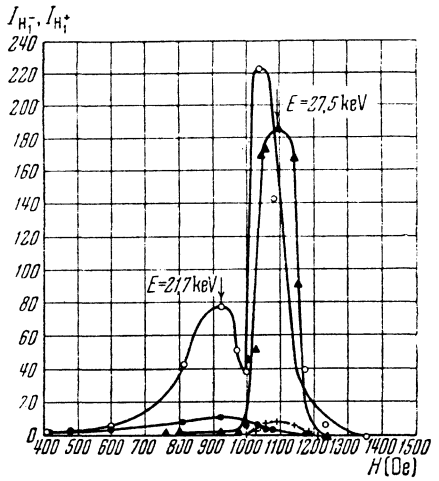

FIG. 1. ▲ – positive ion current obtained without a foil (the scale is reduced 200 times); + – negative ion current obtained without a foil; O – positive ion current, with foil in place; ● – negative ion current, with foil in place.

DESCRIPTION OF APPARATUS AND METHOD OF MEASUREMENT

The apparatus used in this experiment has been reported in an earlier paper[1] (consider Figure 1 in that report). The proton beam was defined by a diaphragm (diameter of 2 mm) which covered the entrance window of a Faraday cylinder *21*. The beam current passing through the diaphragm was measured by the Faraday cylinder. The foil holder *30* was introduced through a slide sealed within a sleeve *25*. The beam, after passing through a foil which was carried by one of the windows in the holder, is analyzed with the help of the magnetic analyzer *23*. To study the composition of the incident beam, it was allowed to pass through one of the windows in the holder which did not contain a foil.

Since we had in mind to work with protons with energies from 10 to 30 kev, it was necessary to prepare sufficiently thin foils so that the protons could penetrate through the foils, i.e., thicknesses of the order of 0.01 milligram/cm^2 . We prepared two types of foils – with and without backing. The method of preparing foils with backing was as follows: A drop of enamel lacquer thinned by amyl acetate was placed on the surface of distilled water. The lacquer spread over the surface of the water and, when it dried, formed a thin film. In the container with the water was a brass plate with a window lying on a small table in such a fashion that the plate surface was beneath the water. After the formation of the enamel film the water was let out of the container, and as the water level fell, the enamel film, settling gradually with the water, settled onto the brass mask and closed the window.

The thickness of the enamel film could be changed by varying the degree of dilution of the lacquer in the amyl acetate. The thickness of the enamel film was obtained by weighing a known area of the film on a microbalance. The backing thickness with which we worked was of the order 0.01 milligram/cm^2 . To obtain backings of smaller thickness appeared too difficult because of their instability.

After the introduction of the enamel film across the window of the brass plate, the film was exposed to air drying for several hours. The brass plate with its enamel film was then inserted in a vacuum apparatus for the deposition of a metal layer onto the enamel film. The determination of the thickness of the metal layer was accomplished by first weighing the backing and then weighing the backing together with the metal layer. For plating the backing with a metal layer, the time of deposition and the rate of heating the evaporator were fixed. In the subsequent deposition of a metal layer, we maintained a constant heating and were able to obtain one or another layer thickness by varying the evaporation time. In this fashion we obtained foils of Be, Al, and Cu with thicknesses from 0.01 to 0.03 milligram/cm^2 .

For the preparation of metal foils without backing we employed another method as follows: A carefully cleaned glass plate was dipped into a solution of cellulose nitrate varnish and acetone. After removing the plate from the varnish it was set vertical, the excess varnish running off. After a short time a film of varnish was formed on the surface of the plate. This film was then removed from the glass and fastened to a thin brass ring which was placed in a chamber where a thin metal layer was deposited on the film by an evaporation process. After this, the ring with its foil and varnish backing was placed on the surface of acetone which had been poured into a suitable container. The varnish backing dissolved in the acetone and the thin metal foil was left floating on the surface. Within the acetone container was a small table containing an inclined brass plate with a window. As the acetone was slowly let out, it was possible to set the metal foil over the window of the brass plate. It was not possible to determine the thickness of such foils (i.e. without backing), but, judging from the foil transparency, they were believed to have thickness of the same order as foils with backing.

After the foil had been set over the window in the brass plate, the plate was attached to a holder which in turn was introduced into a sleeve *25*, in the magnetic analyzer chamber (Fig. 1, reference 1).

Before we approached the study in question (i.e., the dependence on proton energies and foil material of the composition of a beam of protons which has been affected by passage through a metal foil), we had performed some preliminary measurements. In these we had studied the dependence of the proton and negative hydrogen ion currents in the Faraday analyzer on the magnetic field intensity in the analyzer cell. In Fig. 1 the characteristic curves for a beam which had passed through an uncovered window** and for a beam which had passed through a beryllium unbacked foil are represented.

The noticeable width of the curve $I = f(H)$ is due to the fact that the diameter of the aperture of the Faraday cylinder (= 15 mm) was larger than the diameter of the incident beam. Nevertheless, the sharp fall off of the curves testifies to the monoenergetic nature of the incident beam.

The curve $I = f(H)$ for protons which have passed through a Be foil possesses two maxima. The presence of two maxima is due to the fact that the observed curve is a composite of two curves: one for protons which have passed through holes in the foil (i.e., have suffered no interaction) and the other for protons which have suffered energy losses and scattering in their passage.

A first glance comparison of the nondisplaced and displaced maxima suggests that a considerable fraction of the incident beam has passed through holes in the foil. In reality this is incorrect, since one must keep in mind that only a small fraction of the protons passing through the foil enter the Faraday chamber of the analyzer because of the scattering of protons. A calculation which included this effect shows that about 3 percent of the protons pass through openings in the foil. Further observation showed that an undisplaced maximum was always found for foils without backing testifying to the presence of holes in these foils.

The proton and negative hydrogen ion currents for a backed beryllium foil is illustrated in Fig. 2 (the incident proton energies was 28.3 kev). The absence of an undisplaced maximum testifies to the absence of holes in foils with enamel backing. This is characteristic for all foils of this type. By measuring the difference in the magnetic field intensity for the displaced and the undisplaced maximum, we determined the energy loss for passage through the foil.

After these measurements we embarked on the problem under question. One other point that had to be considered was the conditions for an equilibrium composition of the proton beam. The above experiments implied that a foil thickness of about 10^{16} atoms/cm^2 is needed (compare Phillips[5]). The aluminum foils which we used had a thickness of the order of 2×10^{17} atoms/cm^2. The thickness of the beryllium was even larger. Thus the thicknesses of our foils were sufficiently larger than the thickness needed for equilibrium for the incident proton energies in question. In this connection it must be remembered that the equilibrium composition of the emergent beam corresponds to the energy of the emergent protons. If the deviation of the maximum of the emergent energies is a significant fraction of the incident energy, then the results obtained deviate considerably from that characteristic for the incident energies. Thus it should be kept in mind that the composition of the emergent beam, for a given incident energy, may depend on the thickness of the foil.

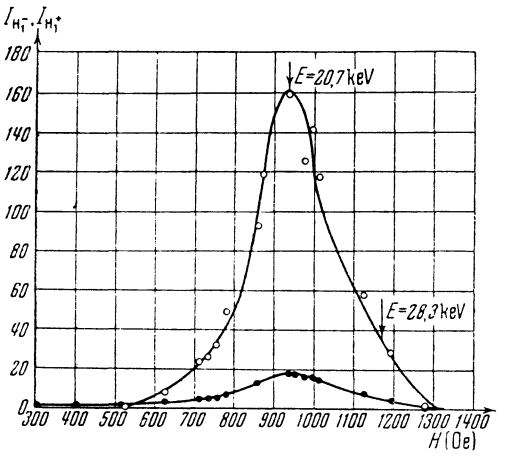

FIG. 2. O – positive current; ● – negative current

The basic measured quantity in our experiment was the ratio of the negative hydrogen ion current to the proton current in the Faraday cylinder of the magnetic analyzer. The values of these currents were obtained from the maxima of the curves $I = f(H)$. Because of the identical width of the proton curve and the negative ion curve, the ratio of the maximum currents gives the correct characteristic percentage of respective particles in the emergent beam.

** In the incident beam there were a few negative hydrogen ions which were formed by collisions between the protons and the residual gas molecules in the path to the analyzer.

The magnetic analyzer was set up in relation to the beam in such a fashion that the maximum of $I = f(H)$ for the negative ions and for the protons occurred at the same magnetic field intensity. Thanks to this arrangement, the measurements of one current could be carried out immediately after the other, thus minimizing errors introduced by variations in the incident beam during the time of a measurement.

The proton and negative ion current in the Faraday cylinder of the analyzer was measured by a string electrometer employing a constant deflection wiring scheme. Two electrometer resistances were used for measuring the two currents, viz., 3.9×10^8 ohms and 3.3×10^{11} ohms. The error in the measurement of the incident beam of protons was of the order of 3 percent and in the measurement of the ratio of currents ~ 7-10 percent.

RESULTS

The majority of our measurements were performed with unbacked foils in order that the results not be influenced by the enamel backing. For those cases when a backed foil was employed, no burning out under the influence of the proton beam was observed. On the other hand, a carbonaceous layer was observed if the backed foil was exposed to the beam for a long time. This was produced by the decomposition of the butyrous film under the action of the proton beam. Consequently, the exposure time was kept small for the measurements to be described, and no carbonaceous layer was formed.

In the process of making measurements we obtained soma data on the relative stability of foils under the continuous action of protons with energies from 10 to 30 kev. Greater foil stability was obtained by the use of small window diameters. For this reason nearly all measurements were made with foils on windows having 4-5 mm diameter. Enamel backed foils showed the greatest stability. They sustained a continuous current density of $3\,\mu A/cm^2$ for 6 hours. Unbacked foils sustained a current density of $1\,\mu A/cm^2$ for one or two hours. Foil collapse for the current densities employed (not greater than $10\,\mu A/cm^2$) occurred because of the gradual loss of material due to the particle blow-off produced by the proton beam.

Unbacked aluminum foils were the first to be studied. The results for four foils are shown in Fig. 3 where the ratio of $I_{H_1^-} / I_{H_1^+}$ is plotted against incident proton energies. The figure shows an increase in the ratio for decreasing energies, reaching ~ 20 percent for 15 kev. In this instance, foils of the same thickness (foils 18*a* and 18*b*, $\Delta E = 2.2$ kev) gave identical ratios. For foils of greater thicknesses this ratio was somewhat larger, in which case the deviation enters into the determination of measuring errors. This result is completely understood since the characteristic composition of the beam is dependent upon the energies of the emergent protons.

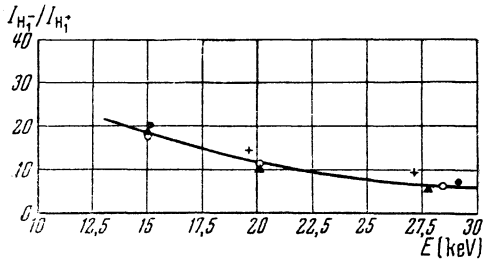

FIG. 3. Here, + = Al-foil No. 23, $\Delta E = 4.5$ kev; ● = Al-foil No. 15, $\Delta E = 5$ kev; o = Al-foil No. 18a, $\Delta E = 2.2$ kev; ▲ = Al-foil No. 18 *b*, $\Delta e = 2.2$ kev.

Beryllium and copper foils were employed next. Figure 4 shows the results for these foils along with the data for aluminum. It is evident that the ratio for the Be foil is quite different from that for Al or Cu.

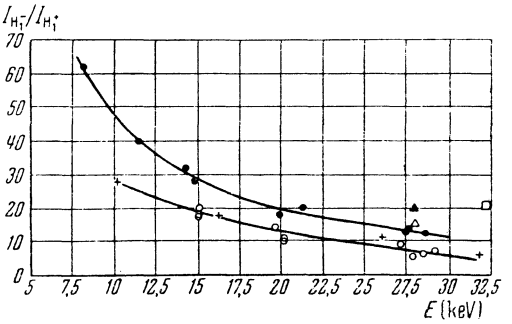

FIG. 4. Value of $I_{H_1^-} / I_{H_1^+}$ for a beam of H_1^+ passing through: o = Al-foil; ● = Be-foil; + = Cu-foil. Values for I_{D^-} / I_{D^+} for a beam of D^+, passing through: Δ = Al-foil; ▲ = Be-foil; Value of $I_{H_2^-} / I_{H_2^+}$ for H_2^+ beam passing through: □ = Al-foil .

Measurements on beryllium with and without backing are presented in Fig. 5. In the case of the backed beryllium foil, the proton beam passed through the metal layer first, i.e., the composition of the beam was characterisitc of the backing material[*] . It is seen that a difference exists in the composition of the emergent beam for a metal layer and an enamel backing. These differences enter into the determination of measurement errors.

[*] Measurements on an enamel film were not possible because of their rapid deterioration in the proton beam.

TABLE I

Foil Material	Proton energy in kev	$\frac{p}{p+1}$	$\frac{I_{H_1^-}}{I_{H_1^+}} \cdot 10^2$	$\frac{I_{H_1^-}}{I_{H_1^+\text{incident}}} \cdot 10^2$
Be	27.4	0.6	13.4	7.1
	21.3	0.55	20.1	9.2
	14.3	0.47	32.3	11.5
	11.5	0.43	40.0	12.3
Al	28.2	0.6	6.0	3.4
	20.2	0.55	10.9	5.4
	15.0	0.47	18.1	7.2
	28.0 [4]	0.47 [4]	15.5 [4]	6.3 [4]

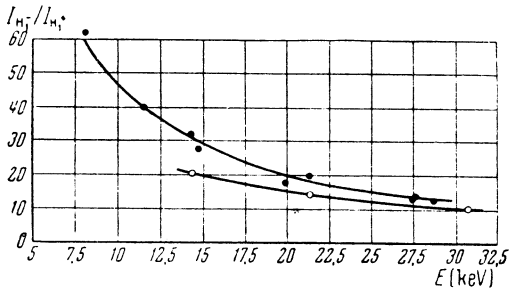

FIG. 5. ● = Be-foil without backing, ΔE = 5-9 kev; O = Be-foil with backing, ΔE = 10-12 kev.

One would expect that in a beam of deuterons passing through a metal foil the negative deuterium ion content should be larger than for a proton beam of the same energy because of the lower deuteron velocity ($v_{D_1^+} = \frac{1}{2} v_{H_1^+}$). Measurements with deuterons having an energy of 28 kev were performed for an aluminum and beryllium foil. The results, confirming our expectations, indicate a ratio I_{D^-}/I_{D^+} of 15.5 percent and 19.4 percent for Al and Be respectively, or approximately equal to the ratio $I_{H_1^-}/I_{H_1^+}$ for 14 kev protons (Fig. 4).♀

Finally, we made measurements with a beam of molecular hydrogen ions H_2^+ with an energy of 32 kev. During the beam's passage through the aluminum foil complete dissociation occured so that the beam was found to contain only protons and negative hydrogen ions with an energy of 16 kev. The ration of $I_{H_1^-}/I_{H_1^+}$ was found to be close to that for incident protons of 16 kev energy (Fig. 4).

♀ *Translator's note*: Appears to be more like 17.5 to 19 kev.

The measurements which we made do not permit the calculation of the negative ion content of the emergent beam since the intensity of the neutral component was not measured. However, if we use our data together with those of Hall[2] we can calculate the negative ion content. Actually, Hall determined the quantity $p = (\bar{N}_1 + \bar{N}_{-1})/N_0$, that is the ratio of the charged to the neutral component. It can be easily shown that the conversion efficiency, $I_{H_1^-}/(I_{H_1^+})_{\text{incident}}$ has the form

$$\frac{I_{H_1^-}}{I_{H_1^+\text{incident}}} = \frac{p}{p+1} \frac{(I_{H_1^-}/I_{H_1^+})}{1+(I_{H_1^-}/I_{H_1^+})}. \qquad (1)$$

To the accuracy to which the number of incident protons equals the sum of the number of emergent protons, atoms, and negative ions, the ratio $I_{H_1^-}/(I_{H_1^+})_{\text{incident}}$ is representative of the conversion factor for a proton beam passing through metal foils.

Employing Hall's data[2] for beryllium and aluminum foils, we calculate with formula (1) the negative hydrogen ion content in the beam passing through these foils. Table 1 summarizes these results. The presented conversion coefficients can be compared with the results of others. Our conversion coefficient for 28 kev deuterons (conversion into negative deuteron ions) in aluminum foils agrees quite well with Alvarez's data[4] for 30 kev deuterons. A comparison of our results for aluminum with those of Phillips[5] shows our negative hydrogen ion content to be significantly larger. Since the method of measurement used by Phillips is not known, it is difficult to say anything about the reason for this discrepancy.

In conclusion it appears from Table 1 that approximately 10 percent of the protons incident on a metal foil are transformed into negative hydrogen ions. Thus the usefulness of this method for obtaining a sufficiently intense beam of negative hydrogen ions for experimental purposes is confirmed.

In conclusion we wish to express our gratitude to Prof. A. K. Bal'teru for his continuing interest and attention.

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