Det Kongelige Danske Videnskabernes Selskab
Matematisk-fysiske Meddelelser, bind **26**, nr. 12

Dan. Mat. Fys. Medd. **26**, no. 12 (1951)

TOTAL CHARGES OF FISSION FRAGMENTS AS FUNCTIONS OF THE PRESSURE OF THE STOPPING GAS

BY

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København
i kommission hos Ejnar Munksgaard
1951

Printed in Denmark.
Bianco Lunos Bogtrykkeri.

Introduction.

In recent papers [1] [2] some measurements on the total charges of fission fragments were reported. The deflections suffered by the fragments along their paths in the magnetic field of the cyclotron were measured. In some experiments the fragments travelled in vacuum, in which cases the curvatures of the paths were determined by the total charges with which the fragments left the uranium layer. In other experiments the fragments passed through a gas at a rather low pressure, and the curvatures were now determined by the average charges of the fragments in the gas. By the passage through the gas the fragments exchange electrons with the gas molecules, and the charges are determined by an equilibrium between capture and loss of electrons. These equilibrium charges were found to be slightly different in different gases and, especially, to differ markedly from the charges in solids. They were, furthermore, found to vary with the pressure of the stopping gas, a variation which may be explained as a result of a competition between loss of electrons from excited states of the fragment ions and radiative transitions to lower states, from which loss of electrons is less probable. The higher the pressure, the more frequent are the collisions and the more predominant will be the loss processes; consequently the total charges equal to the number of lacking electrons of the fragment ions will increase with the pressure. For low pressures such an increase was actually found; for somewhat higher pressures the increase was more slow. Unfortunately the experimental arrangement did only allow the measurements to be carried out for pressures lower than about 35 mm of argon or about 100 mm of the light gases, hydrogen and helium. The purpose of the present work was to extend the investigations to higher pressures in order to see whether the increase of charge is continued for the high pressures or whether a constant charge value is reached.

§ 1. Experimental arrangement.

The method used is the same as in the earlier measurements, the curvatures of the paths being measured by means of three parallel slits, the central one of which is movable in a direction perpendicular to the path of the fragments; however, the geometry

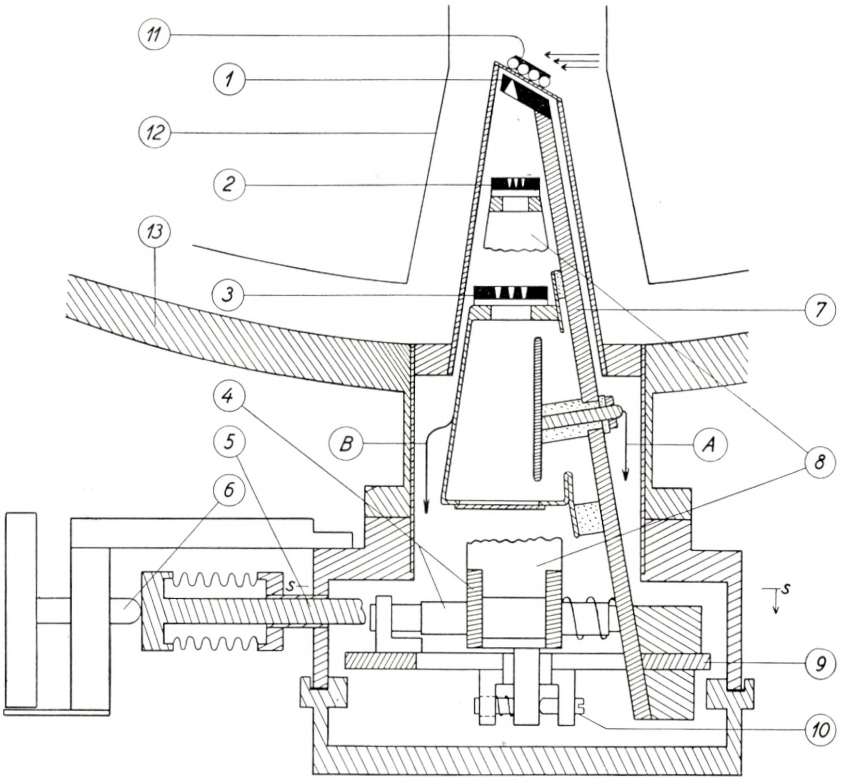

Fig. 1. Experimental apparatus. Horizontal section.

of the apparatus is somewhat altered. It is shown in Figs. 1 and 2. The distance from the uranium layer (1) to the entrance slits (3) of the ionization chamber is about 70 mm instead of the 200 mm depth of the deflection chamber in the earlier apparatus; therefore, for a certain residual energy of the fragments reaching the ionization chamber the pressure in the deflection chamber can be three times higher in the new than in the old arrangement. Moreover, the mica foil covering the slit of the ionization chamber

in the old arrangement has now been omitted, partly to simplify the construction, but partly also to obtain higher residual energy in the ionization chamber so that a still higher pressure can be used in the deflection chamber.

The smaller depth of the deflection chamber results in much smaller displacements of the midmost slit (2); in fact, the displacements are reduced by a factor of 7, so that when we had to measure a displacement of 9—11 mm in the earlier work we now

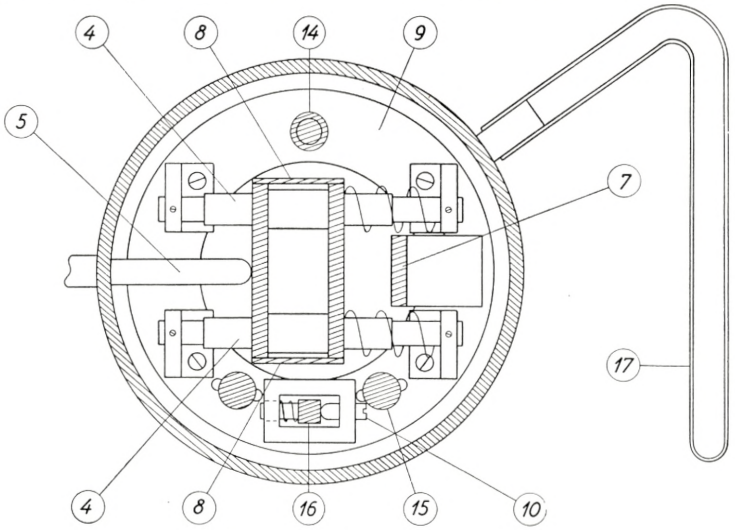

Fig. 2. Experimental apparatus. Section s—s.

have to measure displacements of about 1.5 mm. It is therefore necessary to use very narrow slits to obtain sufficient accuracy. The uranium layer, evaporated on a mica sheet, is placed behind a slit (1) of width 0.18 mm. The slits (2) and (3) are of widths 0.16 mm and 0.46 mm, respectively.

Compared with the earlier arrangement the intensity is severely reduced; to compensate for this result of the narrowness of the slits each of the slits (2) and (3) is replaced by three slits beside each other, so that the apparatus acts as three spectrographs in parallel. The slits (2) are cut in the same piece of brass and are thus mutually fixed; the same holds for the slits (3). When the slits (2) are placed exactly midway between the slits (1) and (3), the mutual distances between the slits (2) must be exactly half the corresponding distances between the slits (3). The distances

must be so large, that when some fragments pass through the central slits of the systems (2) and (3) no other fragments are at the same time able to pass through the central slit of (2) and through one of the outermost slits of (3). From the earlier measurements the width of the charge distribution is known approximately and the present choice of 2 mm as the distance between the slits (2) could be foreseen to exclude undesired interference between the ion beams. On the other hand, the mutual distance should not be chosen too large, because then geometrical differences between the three spectrographs will play a role; for the same reason it is not advisable to increase the number of parallel spectrographs. In the arrangement used the error introduced due to geometrical differences is smaller than one percent and can thus be neglected.

The brass piece in which the slit (1) is cut and which supports the mica sheet with the uranium layer is screwed and soldered to the brass bar (7). The two electrodes of the ionization chamber are fixed to the same bar, but insulated by means of perspex. The one electrode, which is connected to the negative terminus of a battery (B), has the shape of a box surrounding the second electrode, which is simply a brass plate. Free electrons are collected on the latter electrode, which is connected to a linear amplifier (A). The former electrode carries the brass plate with the slits (3). The recording of the fragments is done in some cases by a thyratron and a mechanical counter, in other cases by a cathode-ray-oscillograph, which is photographed on a continuously moving film. The brass piece with the slits (2) is supported by two plates (8), one below and one above the ionization chamber. These plates are fixed to the slide (4), which by means of two springs is pressed tightly against the rod (5); the position of the slits (2) is determined by the micrometer screw (6). The bar (7) is screwed to a brass plate (9), which also supports the slide. This brass plate is fastened to the outer walls of the apparatus, which are the same as was used in the earlier arrangement. It can be taken out together with the essential parts of the apparatus, slide, ionization chamber, and all slits. It is fastened to three brass supports (14) and (15); one of these (14) has a tap, on which the brass plate can rotate, until it is fixed by pressing it against the supports (15) by means of screws. A fourth brass

rod (16) goes through a hole in the brass plate (9); the rotation of the latter is governed by the screw (10) and a spring. The lengths of the brass supports (14) and (15) are adjusted in such a way as to bring the uranium layer close to the neutron source, a beryllium plate (11) soldered to four cooling tubes. As will be clear from the figures the space is rather limited, and only small movements of the slits (2) and only small rotations of the whole apparatus can be allowed; several stops (not shown) serve to

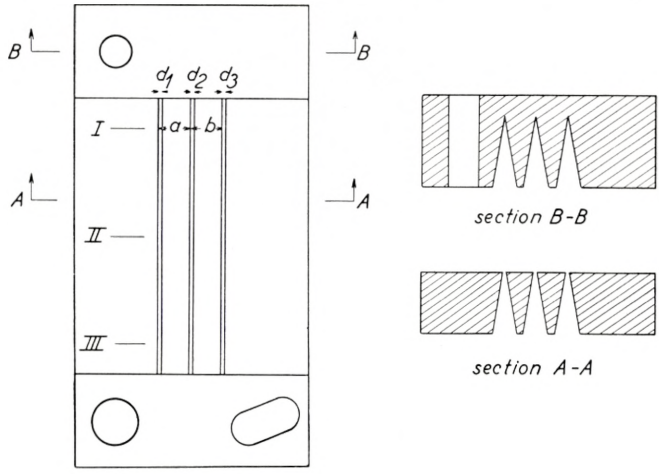

Fig. 3.

avoid damage to the apparatus. In Fig. 1 are shown the edges of the Dees (12) and the wall of the cyclotron chamber (13).

It is clear that the slits must be made and arranged with utmost care, great accuracy being necessary. In order to show the precision obtained some details shall, as an example, be given about the slits (2). The widths of the slits are denoted d_1, d_2, and d_3, and the distances between them a and b (Fig. 3); the latter are measured from the left (or right) edge of one slit to the corresponding edge of the adjacent slit. Measurements at the three places I, II, and III gave the figures in Table 1. As seen, they do not vary by more than 0.01 mm. The same is valid for the corresponding figures for the slits (3). The mutual distances between the latter slits were intended to be 4.00 mm, but they turned out to be 3.96 mm and 3.97 mm. To compensate for this deviation the distance from the uranium layer to the slits (2)

was put equal to 35.0 mm, but the distance between the systems (2) and (3) to 34.4 mm.

TABLE 1.

Widths and mutual distances of slits (2) in mm.

	d_1	a	d_2	b	d_3
I.....	0.16	1.99	0.16	2.00	0.16
II.....	.15	2.00	.16	2.00	.16
III.....	.15	2.00	.15	2.00	.15

The brass plate with the slits (2) is fastened to the support in such a way that it can be rotated so as to make the slits (2) parallel to the slit (1). The arrangement is similar to the one described above, the rotation being carried out by means of a screw and a spring, but due to the limited space and to avoid sharp edges and points the whole mechanism for adjustment is made very small and buried in a recess in the supporting plate; the spring is made of phosfor bronce so as not to disturb the magnetic field. The adjustment is obtained using a suitable light source, a lamp with one vertical filament placed near the focus of a lens. Light from this source is sent through the slit (1) so as to make a "shadow image" of the slit (1) on the plate with the slits (2). This image is made parallel to a line which previously is drawn exactly parallel to the slits (2). By repeating the adjustment one may from the orientation of the screw estimate the uncertainty. The deviation from exact parallelism is believed to be smaller than 0.03 mm along the whole length (15 mm) of the slits.

In similar way the slits (3) are made parallel to the slits (2).

It may be mentioned that even if small deviations from parallelism occur they need not give rise directly to errors in the measuring results, but they will strongly affect the uncertainty. An obliquity has the effect to make the slits broader with respect to resolving power but not with respect to intensity, so that it will make the observed deflection distribution broader and more flat.

When the apparatus is mounted it is adjusted in such a way that the slits are parallel to the lines of magnetic force. The direction of the latter is made visible by means of a piece of

thin steel wire movable about a horizontal axis through its center of gravity. This needle is held by a long thin arm and placed close to the slits (3), the back-wall of the ionization chamber being removed. Using the screw (10) the apparatus is rotated until the shadow of the needle when using the above mentioned light source is parallel to the slits; a telescope is used for observation. The adjustment is repeated after the needle has been turned 180° about its length axis. The uncertainty is about 0.03 mm on the whole length of the slits (3) (18 mm) or 0°.1. The lines of force are found to be vertical within the limits of error.

Great care was taken to obtain a high purity of the gases. The helium was filtered through charcoal in liquid air; the hydrogen and the argon were of purity higher than 99.9 per cent. It was found that with 300 mm H_2 in the chamber an admixture of a few millimeter of air produced a shift in the deflection distribution, the peak being moved from the hydrogen curve to about the argon curve (see later). Since, according to the earlier measurements [1]) the cross-sections for capture and loss of electrons by the fragments are very much smaller in hydrogen than in the heavy gases, it should just be expected that a small admixture of air in the hydrogen would have a considerable influence on the charge.

§ 2. The measurements.

A difference from the earlier experiments lies in the fact that now the pressure is the same between the uranium layer and the ionization chamber as it is inside the latter. At low pressures the residual range of fragments reaching the chamber is long, but it cannot be fully employed, the depth of the chamber being too small; at high pressures the equivalent depth of the chamber is great, but the residual range is only short. From an earlier work of the author [3]) the mean energy spent by the fragments inside the ionization chamber may be estimated, and Table 2 gives the figures. By the estimation it has been taken into account that the thickness of the uranium layer is considerable, 1.2 mg/cm^2.

Since the ionization chamber is placed rather close to the Be-target in the cyclotron the background due to γ-rays and

TABLE 2.

Gas	Pressure	Energy loss inside the ionization chamber	
		Light fragment	Heavy fragment
H_2	100 mm	16 MeV	12 MeV
—	200 —	24 —	12 —
—	300 —	22 —	10 —
—	400 —	16 —	8 —
A	38 —	17 —	14 —
—	76 —	25 —	11 —
—	114 —	22 —	8 —
—	152 —	12 —	4 —

neutrons is high. This background together with the thickness of the uranium layer involve a large spread in the energy measurements. Therefore one cannot expect to be able to distinguish between the two fragments in the same manner as earlier, when the energy difference is so low as is the case at a pressure of 100 mm H_2. At this pressure the recording is performed simply by means of a thyratron biased properly. The number of fragments is counted for each position of the slits (2) within the interval of interest, this interval being crossed several times in both directions. The deflection distribution is shown in Fig. 4a. For comparison, in Fig. 4b is shown a curve obtained in a similar way at a pressure of 300 mm H_2. At this pressure the background is so high, that the heavy group of fragments disappears. Only the light group is counted, and correspondingly the peak in Fig. 4b is narrower than the one in Fig. 4a, which includes both groups of fragments.

At an intermediate pressure, 200 mm H_2, the difference between the energy losses inside the ionization chamber of the two groups is great and the background is not higher than to allow at least some of the heavy fragments to be counted. At this pressure the pulses are recorded by means of the photographic arrangement; when the films have been measured, the material is divided into two parts according to the sizes of the pulses. The larger pulses are ascribed to the light group, the smaller pulses to the heavy group (cf. ref. [1])). Since not all heavy fragments are included, some having disappeared in the background, one should not divide in such a way that the two parts are equal in number. By trying to divide in different ways one

finds that the position of the peak for the light group can be determined rather accurately, while the position of the peak for the heavy group is only roughly indicated.

It was planned to measure the deflections at pressures of 100 mm H_2 and higher; of course it would also be of interest to follow the deflection pressure curve down to lower pressures in order to compare with the earlier measurements. However, this turned out to be somewhat difficult; the ionization chamber was found to work well at pressures of 100 mm H_2 and higher,

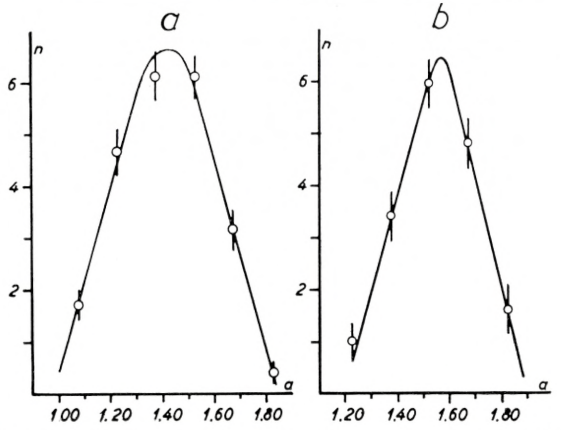

Fig. 4. Deflection distributions obtained at 100 mm H_2 (a) and at 300 mm H_2 (b).

but not at very low pressures. At 30 mm H_2 an ion collection can be obtained, but when the voltage across the chamber is too low ($\sim$ 50 volts) only a few and small pulses occur, and when it is too high ($\sim$ 200 volts) corona discharge takes place. At pressures of 10 and 15 mm H_2 attempts to get the chamber to work have been unsuccessful. The reason for the difficulties is presumably, that the ionization chamber is situated inside a magnetic field of strength 18,000 oersteds. In vacuum a very high voltage would be needed to draw the ions to the collecting plate, and for usual voltages the path of an ion would consist of a series of cycloidal loops, so that the resulting displacement of the ion would be in a direction perpendicular to the electric force and the ion would never reach the collecting plate. By a field strength $\sim$ 100 volts per cm the length of a single loop will be about 2×10^{-5} cm for the negative ions, which are free electrons, as above mentioned. When the ionization chamber is filled with a gas to a sufficiently

high pressure, however, the elastic collisions between the molecules of the gas and the ions will be so numerous, that the movement of the ions will be completely altered and they will be collected almost as easily as without the magnetic field. When the gas pressure is reduced the number of collisions decreases, and difficulties may be expected to arise when the pressure is lowered so much that the mean free path for collisions becomes much larger than the length of the cycloidal loops. Already at atmospheric pressure the mean free path for the negative ions in hydrogen is comparable with the length of a loop. This would seem to account for the observed difficulties at pressures below 30 mm H_2, since at such pressures the electronic mean free path is considerably longer than a loop.

In helium the chamber works well at pressures af 150 mm and higher; it can work at 100 mm, but corona takes place at a considerably lower voltage than in hydrogen.

In argon the chamber will work at pressures of 35 mm and higher. Yet, while the countings can be reproduced when the two light gases are used, the pulse sizes found with a certain pressure of argon vary from one experiment to the next and consequently the number of pulses higher than a certain limit will also vary ($\sim$ 30 %). Even in the same experiment the conditions may change in the course of a few hours. However, by repeated measurements the most probable deflection value has been found to be unchanged. The reason for the changing sensitivity of the chamber is not known, but in this connection it may be mentioned that in the beginning of the experiments very much larger changes occurred. These were due to condensation of vapor on the uranium layer, which took place although the glass tube (17) (see Fig. 2) was cooled down to $-65°$ C; probably the vapor originated from the oil used for greasing the slide. This trouble is completely overcome, at least when the light gases are used, by pre-heating the cooling water for the Be-target some 15 degrees; as seen in Fig. 1, this water also cools (or heats) the surroundings of the uranium layer.

In some experiments a mica foil of thickness 0.47 mg/cm$_2$ has been placed close in front of the uranium layer. In order to do this it was necessary to take out the whole apparatus; then it was of great importance that the micrometer-screw would show

the same zero point when the apparatus was replaced. Now, to the plate (9) a stop-screw is fastened, which prevents the slide from moving too far the left. When the micrometer-screw is loosened the springs already mentioned will press the slide against the stop-screw. The figure which is read on the screw-head, when the micrometer-screw just touches the slide in this position, was used as a relative zero point; it was found to be unchanged within one hundredth of a millimeter from one run to the next run. Of course, this does not necessarily mean, that the apparatus is unchanged; however, after opening and replacing of the apparatus, the deflections were found, within the limits of error, to be the same as before.

Determination of the absolute zero-point for the micrometer-screw—the figure corresponding to zero deflection—has been made by means of a photo-multiplier. The inner part of the apparatus was taken out and fastened to a flange by means of three supports in the same way as in the actual arrangement. Light was sent through the slits and measured by the photo-multiplier; by means of a micrometer-screw the position of the slide giving maximum of light was determined relative to the position in which the slide is pressed against the stop-screw.

§ 3. Results.

In Fig. 5 the most frequent deflections corresponding to the various pressures are plotted against the pressure.

For hydrogen the peaks obtained at 300 and 400 mm correspond to the light group, as mentioned above. At 200 mm the point for the light fragment is also directly measured while at 100 and at 30 mm H_2 the observed points correspond to some sort of average values between the two groups. The peak corresponding to 100 mm H_2 was obtained by means of the thyrotron, but in one experiment photographic recording was used; this experiment gave as results, firstly that the position of the peak is independent of the minimum pulse size counted, i. e. it is impossible, as was also expected, to distinguish between the two groups by means of the pulse size; secondly, that the fragment pulses are clearly separated from the background pulses, the smallest fragment pulses being greater than the largest pulses of

the background. This latter result tells us, that in the peak shown in Fig. 4a equal numbers of fragments from the two groups occur. If the distribution curves for the two groups both were equal in shape to the curve in Fig. 4b it would seem reasonable to analyse the curve in Fig. 4a, and the position of the peaks corresponding to each of the groups thus determined would be

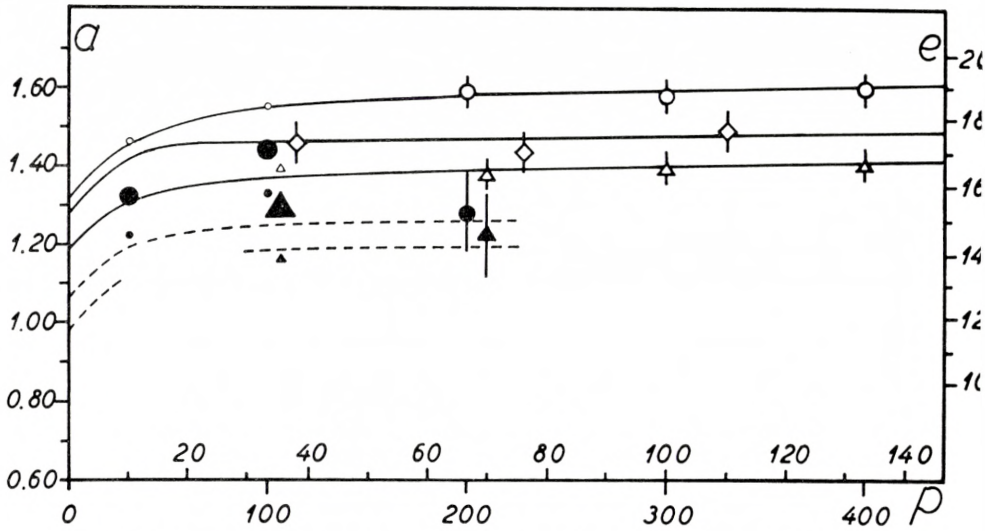

Fig. 5. Total charge of fission fragments as function of the pressure of the stopping gas.
Ordinates: Deflection a in mm. The scale to the right side gives the approximate charge in units of the electron charge.
Abscissae: Pressure of the stopping gas in mm Hg. Upper scale valid for argon, lower scale for hydrogen and helium.
Big and small open circles, squares, and triangles correspond to the light fragment in H_2, A, and He, respectively; regarding the difference between the small and the big points, see text. The small and medium sized full circles and triangles correspond to the heavy fragment in H_2 and He, respectively. The biggest full circles and the big full triangle give the most frequent deflections for all fission fragments and thus correspond to "mean values" between the figures for the two groups.

plausible. The small circles in Fig. 5 give the result, the open and the full circle corresponding to the light and heavy group, respectively. Since the assumption of equality in shape of the distribution curves is certainly not fulfilled the points are somewhat uncertain; especially, the point for the heavy group may possibly lie too high. The small points for 30 mm pressure are obtained in a similar way.

In the earlier experiments the deflection pressure curve was determined in the range 0—100 mm of hydrogen. The corresponding curve valid for the present arrangement may be calculated in the following way. Fragments with an initial radius of curvature $\varrho = 35$ cm will in the old arrangement correspond to an a-value of 12.1 mm [4]; this is calculated from the known variation of the magnetic field along a radius of the cyclotron, a variation which in relative measure is essentially the same for all values of the central field. In the present arrangement the field is constant along the whole path from uranium layer to ionization chamber and one finds that $\varrho = 35$ cm correspond to $a = 1.72$ mm. Hence, one mm deflection in the old arrangement corresponds to 0.142 mm in the present arrangement. Using this result the curve for the light fragment in the interval 0—100 mm H_2 is calculated. It is shown in Fig. 5, where it has been fitted together with the curve above 100 mm pressure. By this procedure the zero-point for the micrometer-screw is fixed; the value so obtained agrees within the limits of error with the zero-point measured directly in the way described in the preceding paragraph.

Using the same value for the zero-point the curves for the light group in argon and helium are calculated for the intervals 0—35 mm A and 0—100 mm He. The curve for argon is shown in the figure; the curve for helium coincides with the one shown in the interval 0—20 mm and at 90 mm. As seen, both curves fit in nicely with the experimental points now obtained.

The curves for the heavy group in hydrogen and helium are calculated in a similar way and they are shown by the dashed lines.

In the calculation of the curves for pressures smaller than 35 mm A or 100 mm H_2 or He one did not directly use the deflection pressure curve obtained by the old arrangement; it was taken into account that at pressures of 35 mm of argon and 100 mm of the light gases the deflections were, in the old arrangement, not simply proportional to the initial charges, but corrections were involved by the slowing down of the fragments. In the present experiments the corresponding corrections are, at the same pressures, much smaller, since the length of the deflection path is reduced to one third.

The measurements in helium are very similar to those in

hydrogen, but the measurements in argon exhibit some new features. The deflection distribution obtained at 37 mm A is similar in shape to the one obtained at 100 mm H_2, but the number of fragments per unit neutron dose is much smaller, somewhat less than the half. At 76 mm A nearly the same result is found. Photographic recording shows, that it is not possible neither at 37 nor at 76 mm to get two peaks by dividing the fragments into two groups according to their sizes. At 105 mm A the peak is much broader than at the lower pressures, the half-value breadth being increased by 50—60 per cent. At 140 mm A the curve is smeared out to a very broad and very flat, almost undetectable peak. This is caused by the compound scattering, which according to theory [5]) increases along the path of the fragments and is highest near the end of the track. The small number of fragments indicates, that already at the lower pressures the compound scattering of the heavy fragments is large enough to make the distribution curve so broad that the fragments cannot be detected. It must be concluded that only the light group is observed and, as seen in Fig. 5, this conclusion gives conformity with the earlier measurements. From the cloud-chamber studies by Bøggild, Brostrøm, and Lauritsen [6]) it is possible to estimate roughly the influence of the compound scattering. In the region from 15.5 mm to 10.5 mm from the end of the track the bending of the tracks is, according to these authors, given by a distribution curve with an upper limit of about 7 degrees. At 76 mm A the deflection in the present arrangement takes place for the heavy fragments along the path from about 17 to about 10 mm from the end of the track and, hence, we must expect at least a similar magnitude of the bending or, since some of the fragments have shorter ranges, probably even a somewhat larger bending. The width of the deflection distribution must therefore be expected to be at least 2 mm larger than the width caused by other factors and probably even much broader. Thus, it is to be expected that the heavy fragments cannot be detected at 76 mm pressure, and it is also reasonable that they cannot either be seen at 37 mm.

At low pressures the deflection a is proportional to the initial charge e [4]). At higher pressures, where the fragments are slowed down along the deflection path, one has

$$a = \text{const} \times \frac{e}{mv}.$$

Since the charge varies along the range almost proportional to the velocity, a is approximately proportional to the initial charge also at higher pressures. We find

$$e = 11.9 \times a$$

where a is measured in mm and e in electronic charges. The scale thus obtained is given to the right side in Fig. 5.

Since the variation of e along the range need not be exactly proportional to v, small corrections might be necessary to obtain the initial charge from the deflection a. An attempt to determine these corrections has been made by measuring the deflections obtained when a mica foil of thickness 0.47 mg/cm$_2$ is placed just in front of the uranium layer. This foil is traversed in a direction making an angle of about 30° with the normal and thus corresponds to about 4 mm of air or to about 12 to 15 mm of hydrogen. The mean value of the fragment velocity will be nearly the same at 400 mm pressure and without absorber as it is at 100 mm pressure and with the absorber. The deflections of the light group with the mica absorber in position at 100 mm H_2 and at 150 mm He are 1.55 ± 0.04 mm and 1.40 ± 0.04 mm, respectively. A comparison with the curves in Fig. 5 at once shows that the possible corrections are negligible.

As will be seen from the figure the charge of the light group of fragments is found to vary strongly at low pressures but to be nearly constant for pressures above some 20 mm of argon. It is interesting that the charge is higher in hydrogen than in argon. It was for a long time believed that the charge was almost the same in all gases except in hydrogen, where it could be expected from theoretical considerations [5]) to be slightly higher. This expectation is now experimentally verified; at the same time evidence is found for a difference between the charge in A and He, the latter being the smaller one in conformity with the results of the earlier measurements at low pressure [1]).

With increasing pressure the charges of the light fragments seem to approach the values 19 ε and 18 ε in H_2 and A, respectively. These values are somewhat lower than the corresponding charges

which from measurements of the specific ionization were estimated [3]) to be 23.8 ε and 22.2 ε, respectively. This difference may be explained by the finite size of the fragment ions and the continuous capture and loss of electrons, which involves a somewhat higher ionization effect than would correspond to a simple point charge. In fact, the observed differences are of the expected order of magnitude, as will be discussed in a forthcoming paper by BOHR and LINDHARD, already referred to in [1]), and where a closer discussion of the capture and loss problems will be given. In the meantime Dr. G. I. BELL has kindly informed me that in connection with a study of such problems he also finds that the corrections due to the above mentioned effects are of a magnitude corresponding to the difference between the charges previously determined from ionizatron measurements and the total charges found in the present work.

The present experiments were carried out at the Institute for Theoretical Physics in Copenhagen, and the author wishes to express his heartiest thanks to the Director of the Institute, Professor NIELS BOHR, for his great and continued interest in the work. Furthermore, my thanks are due to Professor J. C. JACOBSEN for helpful advice in experiments and to Mr. J. LINDHARD for valuable discussions.

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Indleveret til selskabet den 31. maj 1951.
Færdig fra trykkeriet den 8. august 1951.