Equilibrium Charges of Fission Fragments in Gases

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A magnetic analysis method was used to measure the equilibrium charges of median-mass light and heavy fission fragments in H_2, He, air, and A, and mass-97 fragments in He for wide ranges of fragment velocity and gas pressure. The data for the fragments in H_2, air, and A showed pressure effects on the equilibrium charge which yield estimates of the mean radiative lifetime of excited ionized fragments which agree within 25%, in contrast with previous estimates differing by a factor of 10. An experimental test of the Bohr assumption, that the equilibrium charge of a heavy ion is equal to the number of electrons whose orbital velocities are lower than the translational velocity of the ion, showed the assumption to be only a rough approximation. Measurements of equilibrium charge as a function of atomic number of the stopping gas agree with previous measurements.

INTRODUCTION

THE charge of a heavy ion which is moving through matter fluctuates about an equilibrium value determined by two competing processes: electron capture and electron loss. The fragments from nuclear fission are a unique source of heavy ions which may be studied experimentally to determine the influence of the various factors which affect the equilibrium charge. In general the equilibrium charge, $\bar{e}$, of a fission fragment moving in a gas is given by

$$\bar{e}=f(Z,v,P,m,\text{ type of gas}), \qquad (1)$$

where Z is the nuclear charge, v is the velocity, P is the gas pressure, and m is the fragment mass.

A convenient method for measuring the equilibrium charge is to pass the fragments through a magnetic field; from the measured radii of curvature of the fragment paths the charge is given by the well-known relation

$$H\rho=mvc/\bar{e}, \qquad (2)$$

where H is the magnetic field strength, ρ the radius of curvature, and c the speed of light.

The dependence of $\bar{e}$ on Z has been studied experimentally by Cohen and Fulmer.[1] Using a magnetic deflection arrangement, Lassen[2] measured the charges of light and heavy fragments, with initial velocities, and after the fragments were slowed down by thin foils placed in their path.[3] These measurements showed the charge to decrease approximately linearly with fragment velocity.

Lassen also observed a dependence of $\bar{e}$ on gas pressure[4] at low values of pressure (principally for fragments that had lost very little energy). This pressure effect was explained by Bohr and Lindhard[5] to be due to incomplete radiative dissipation of the electron excitation energy of the fragments between successive collisions with gas atoms. However, Bohr and Lindhard,[5] using the data of Lassen[4] for the fragments in H_2, He, and A, obtained very discrepant estimates of the mean radiative lifetimes of excited ionized fission fragments.

In the work reported here measurements of the equilibrium charges of median-mass light and heavy fission fragments were made for a wide range of fragment velocity and gas pressure; equilibrium charge data were also obtained for mass-97 fragments in He gas for a wide range of fragment velocity and gas pressure. The data made it possible to study the dependence of $\bar{e}$ on gas pressure and on fragment velocity rather thoroughly. The pressure effect determined for various gases gave consistent estimates of the mean radiative lifetime of excited ionized fragments. The Bohr assumption,[6] that the charge of a heavy ion is determined by the number of electrons whose orbital velocities are smaller than the translational velocity of the ion, was experimentally tested and found to apply only as a rough approximation.

EXPERIMENTAL

The high-resolution magnetic fission fragment spectrograph, previously described,[7] was used for these studies. For some of the data it was desirable to fill only the deflection chamber with gas, so that higher pressures could be used without stopping the fragments. This was accomplished by mounting 100 $\mu g/cm^2$ Formvar windows, supported by 50% transparent Ni grids, at the entrance and exit of the deflection chamber.

Two methods were used for obtaining data. One method employed catcher foils located at the focal plane of the spectrograph, followed by radiochemistry. The 17-hr activity of Zr^{97} was used to determine accurately the $H\rho$ distributions of mass-97 fragments by this method. The other method employed scintil-

* Operated for the U. S. Atomic Energy Commission by Union Carbide Nuclear Company.
[1] B. L. Cohen and C. B. Fulmer (unpublished).
[2] N. O. Lassen, Phys. Rev. **68**, 142 (1945).
[3] N. O. Lassen, Phys. Rev. **69**, 137 (1946).
[4] N. O. Lassen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **26**, No. 12 (1951).
[5] N. Bohr and J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **28**, No. 7 (1954).
[6] N. Bohr, Phys. Rev. **58**, 654 (1940).
[7] Cohen, Cohen, and Coley, Phys. Rev. **104**, 1046 (1956).

lation detection of the fragments at the focal plane. For the detector a very thin (0.003 in.) CsI(Tl) crystal and a Dumont 6292 photomultiplier tube were used. The details of the detector are given in another paper.[8]

Integral pulse counting was used to determine $H\rho$ distributions of the fission fragments for various pressures of gas in the system and in the deflection chamber only. The peaks of the $H\rho$ distributions correspond to the median light and heavy fragments, respectively. The gases used include H_2, He, air, and A. $H\rho$ distributions were determined for mass-97 fragments over a wide range of pressure with He gas in the system and in the deflection chamber only.

RESULTS

The $H\rho$ values of the median light and heavy fragments in the various gases used are shown as functions of pressure in Figs. 1–4. For low pressures of He, the peaks of the $H\rho$ distribution of the heavy fragments were beyond the range of the magnet. Part of the distributions were within range, and thus the positions of the peaks were estimated as indicated by the dotted portions of the heavy-fragment curve in Fig. 2. The $H\rho$ values of mass-97 fragments in He are shown as a function of pressure in Fig. 5. Data obtained with gas filling the entire system are indicated by triangles, and data obtained with gas in the deflection chamber only are indicated by circles. The ordinate scales are greatly expanded and do not start at zero.

The energies of the fragments, at the middle of the deflection chamber, are shown for several data points

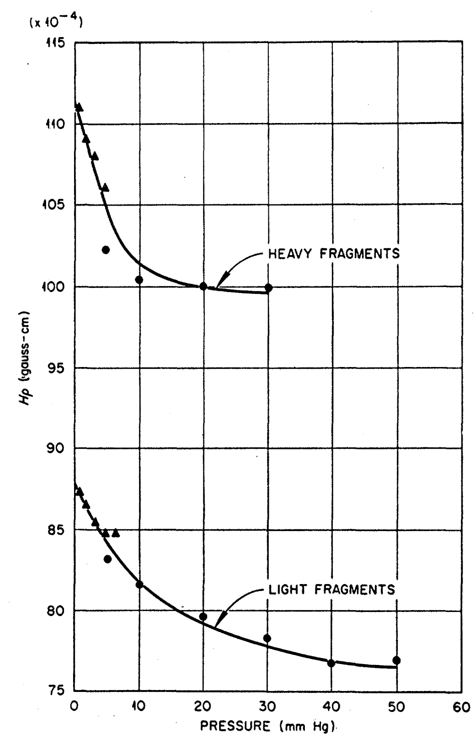

FIG. 1. $H\rho$ values of median-mass light and heavy fission fragments in H_2 as functions of pressure.

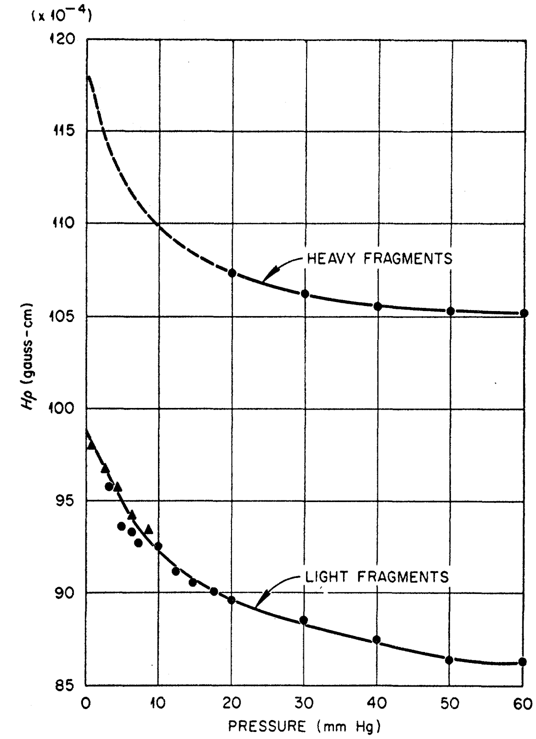

FIG. 2. $H\rho$ values of median-mass light and heavy fission fragments in He as functions of pressure.

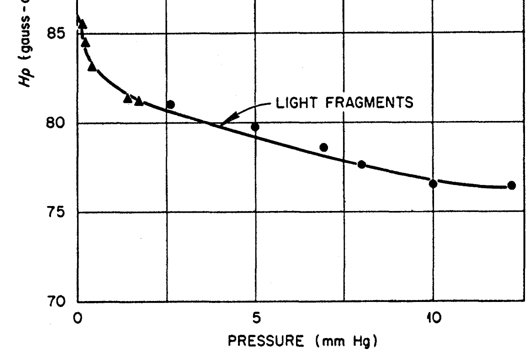

FIG. 3. $H\rho$ values of median-mass light and heavy fission fragments in air as functions of pressure.

[8] C. B. Fulmer, Phys. Rev. **108**, 1113 (1957).

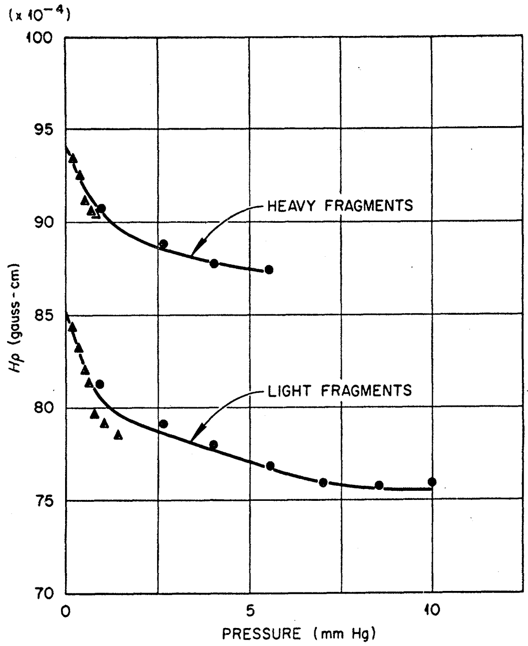

FIG. 4. $H\rho$ values of median-mass light and heavy fission fragments in A as functions of pressure.

in Fig. 5; the data obtained with the two conditions of gas filling (i.e., gas in the entire system and gas in only the deflection chamber) overlap. The energies were determined by the gas pressure, length of path, and range *vs* energy curves.[8] It is seen that fragment energy has very little effect on the $H\rho$ values, and hence the curves in Figs. 1–5 are essentially curves which show the pressure effect on the equilibrium charge. The ordinate values approached by the curves for zero pressure are the $H\rho$ values of the fragments with the pressure effect removed. The flat portions of the curves correspond to saturation of the pressure effect.

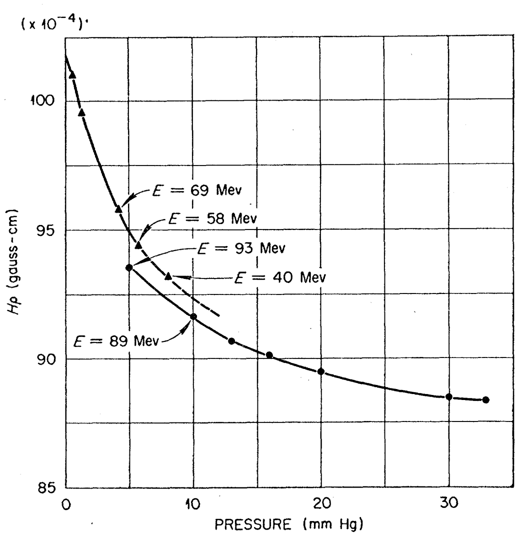

FIG. 5. $H\rho$ values of mass-97 fission fragments in He as a function of pressure. Energies were calculated for the center of the magnetic deflection chamber.

From the zero-pressure values of $H\rho$ in Figs. 1–4 the equilibrium charges of the median light and heavy fragments are obtained in Eq. (2). The results are shown in Fig. 6.

FACTORS AFFECTING EQUILIBRIUM CHARGE

Gas Pressure

Bohr and Lindhard[5] attribute the pressure effect on the equilibrium charge to incomplete radiative dissipation of electron excitation energy of the fragments between successive collisions with gas atoms. The time between collisions is proportional to the number of gas atoms per unit volume and hence the pressure. If the pressure is sufficiently low the radiative process is completed between collisions and the electron loss cross section for the ground state of the ion determines the equilibrium charge. Electrons in excited states are more easily removed from the fragment; thus, excitation energy that is retained between collisions increases the electron loss cross section and hence the equilibrium charge. If the gas pressure is sufficiently high almost none of the excitation energy is radiated between collisions and saturation of the pressure effect occurs.

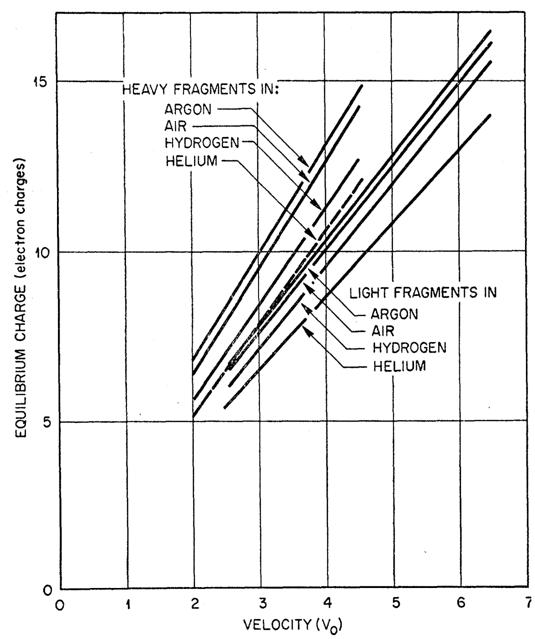

FIG. 6. Equilibrium charges of median-mass light and heavy fission fragments as functions of velocity in various gases.

The range of $H\rho$ for the curves of Figs. 1–4 corresponds to the range of the pressure effect. Values of the maximum pressure effects on the equilibrium charges of the light and heavy fragments, determined from data for the various gases, are given in Table I.

Lassen[4] obtained similar results for the maximum pressure effect on the equilibrium charge of light fragments. The values shown in Table I for the heavy fragments are lower than those obtained by Lassen. The results reported here were obtained with more accurate measurements and larger numbers of data points than were the results of Lassen.

Since the pressure effect on the equilibrium charge of fission fragments is due to incomplete radiative dissipation of the excitation energy between successive collisions, it is easily seen that a given fraction of the maximum pressure effect corresponds to the same fraction, f, of the fragments which remain in excited states between collisions. f is given by the relation

$$f=e^{-\lambda t}, \tag{3}$$

where λ is the probability of decay to the ground state per unit of time.

TABLE I. Maximum pressure effect on $\bar{e}$ of light and heavy fission fragments.

Gas	Maximum increase of $\bar{e}$ for light fragments (%)	Maximum increase of $\bar{e}$ for heavy fragments (%)
H_2	13.6	10.7
He	14	11
air	16	9.1
A	17	11

TABLE II. Mean radiative lifetime of light fission fragments as determined from $H\rho$ data for the fragments in various gases.

Gas	Pv^2 (average)	σ/σ_{air}	$\sigma/\pi a_0^2$	$\tau(\sigma/\pi a_0^2)$ (10^{-11} sec)	τ (10^{-11} sec)
H_2	120	0.104	1.48	6.5	4.3
He	90	0.139	1.98	5.8	2.9
air	12.5	1.000	14.3	68	4.8
A	15.5	0.810	11.6	72	6.2

The time between successive collisions is

$$t=1/kp\sigma v, \tag{4}$$

where kp is the number of atoms or molecules per unit volume and v is the fragment velocity. Equation (4) may be written

$$t\sigma=1/kpv. \tag{5}$$

If the abscissa scales of Figs. 1–4 are multiplied by v they will correspond to the reciprocal of the right side of Eq. (5). A curve of this type for the light fragments in He is plotted in Fig. 7. From Fig. 7 a graph of f *vs* $t\sigma$ is constructed by plotting the fraction of the maximum pressure effect, corresponding to a given abscissa value, *vs* the reciprocal of the abscissa value; this assumes σ to be velocity independent. The resulting graph is shown in Fig. 8. From Fig. 8 a value of $\tau\sigma$ is obtained, where τ is the mean radiative lifetime of the fragments. This value of $\tau\sigma$ and values obtained in the same manner from the light fragment data in H_2, air,

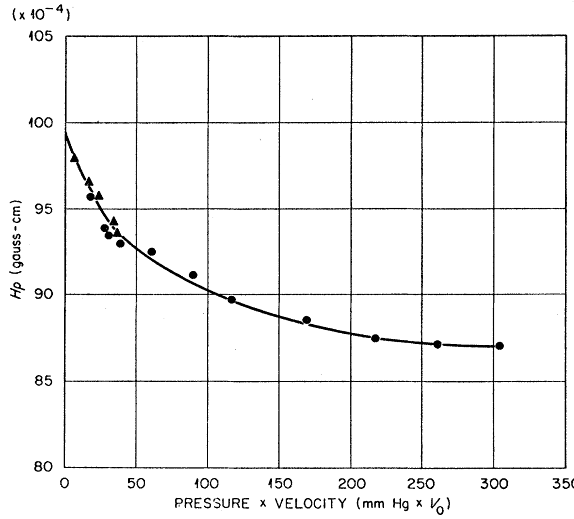

FIG. 7. $H\rho$ values of median-mass light fission fragments in He as a function of (pressure×velocity).

and A are given in Table II. Values of σ for the various gases are needed to determine numerical values of τ.

The charge changing process as the fragments penetrate a gas is a statistical one, and thus there is a distribution of average charge values for fragments of the same mass and velocity. The width, w, of this distribution varies according to the relation

$$w=K/(\sigma P)^{\frac{1}{2}}. \tag{6}$$

This is illustrated by data shown in Fig. 9. It was shown[1] that K is reasonably independent of fragment energy.

Values of w were determined for mass-97 fragments in low pressures of H_2, He, air, and A. These data and Eq. (6) were used to calculate relative values of σ which are shown in Table II. The values for H_2 and air are for molecules rather than atoms.

An absolute value of σ for one of the gases is needed for obtaining estimates of τ. Bohr and Lindhard[5]

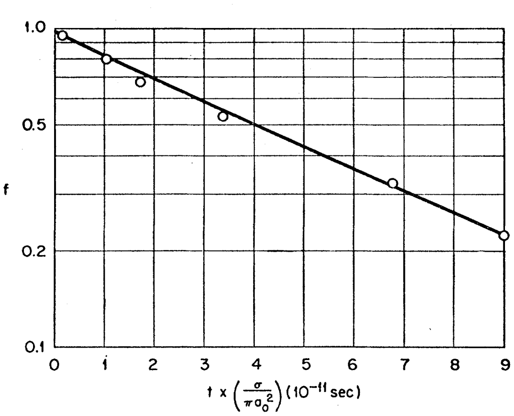

FIG. 8. Fraction of excitation energy retained by median-mass light fragment as a function of time between collisions for velocity—independent cross section—helium data.

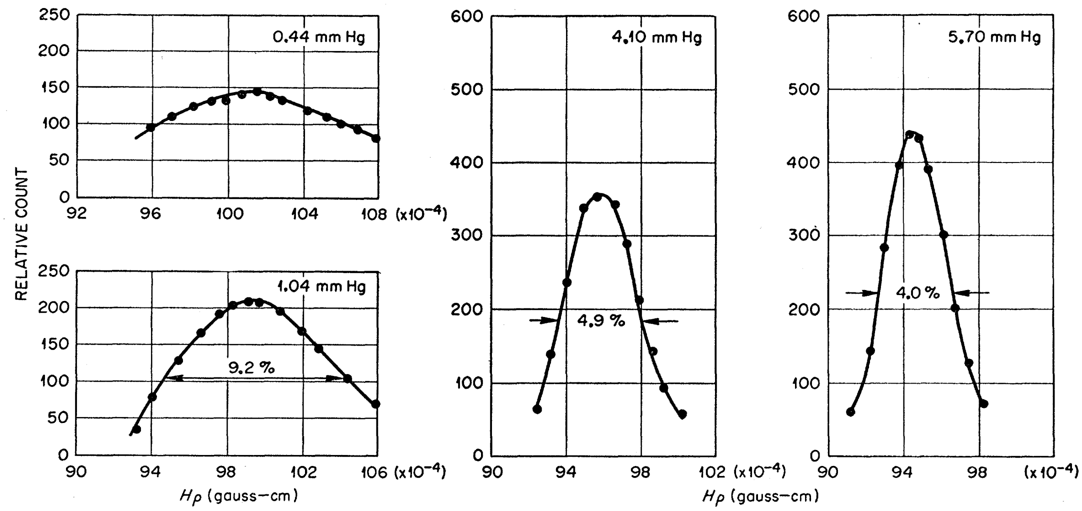

FIG. 9. $H\rho$ distributions of mass-97 fission fragments in He at various pressures.

derived a theoretical formula for calculating σ. The most accurate experimental measurement of the effective electron capture cross section was made by Cohen, Cohen, and Coley[7] for air with mass-97 fragments which had charges considerably above the equilibrium value. The experimental value is 30% larger than that obtained by the theoretical method of Bohr and Lindhard. Accordingly the calculated value of σ for air molecules is increased by 30% and a value, $\sigma/\pi a_0^2=14.3$, is adopted for air. It is noted that this is only a rough estimate.

The corresponding values of σ for the other gases and the values of τ obtained for the light fragments are shown in Table II. The values of τ obtained from data for H_2, air, and A all agree within 25% of the average value; this is within the uncertainty of the several estimates that were made to obtain them. The lower value of τ obtained from He data can probably be attributed to the lower charges of the fragments in He (Fig. 6).

The relative values of τ obtained here remove the

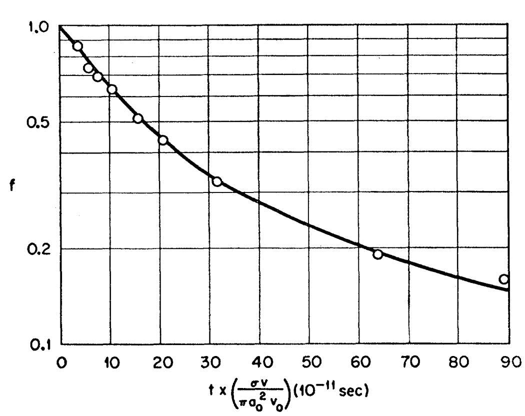

FIG. 10. Fraction of excitation energy retained by median-mass light fragment as a function of time between collisions for $(1/v)$—dependent cross section—helium data.

discrepancies of the values obtained by Bohr and Lindhard[5] from the data of Lassen[4] which had a factor of 10 between the values of τ obtained from H_2 data and A data.

If σ is velocity-dependent, the above estimates of τ must be modified. For instance, from the theoretical calculations of Bohr and Lindhard[5] σ varies as $1/v$. This implies that the curves of $H\rho$ *vs* P (Figs. 1–4) should be used to obtain curves of f *vs* $t\sigma v$. The curve obtained in this manner for the light fragments in He is shown in Fig. 10. Similar curves were obtained for the light fragments in H_2, air, and A. The curve in Fig. 10 implies that if σ is $1/v$-dependent, the excited fragments do not return to the ground state by simple decay.

Fragment Velocity

The data of Figs. 2–5 indicate that $\bar{e}$ varies linearly with velocity. The data in Fig. 5 are for fragments of a single mass and consequently would show any strong dependence of $H\rho$ on fragment velocity more clearly than the data of Figs. 1–4 where the data points represent average masses.

If it is assumed that

$$\bar{e}\propto v^{(1-x)}, \tag{7}$$

and this is substituted into Eq. (2), and only fragments of a single mass are considered, then

$$\Delta H\rho/H\rho=x(\Delta v/v). \tag{8}$$

The data of Fig. 5 were used with Eq. (8) to calculate values of x; for pressures of 5, 8, and 10 mm Hg, the results are $x=0.055$, 0.024, and 0.017, respectively. The wide range of x-values obtained and the manner in which they vary with pressure suggest two possible explanations. Either the relation between $\bar{e}$ and v cannot be represented by Eq. (7), or parts of the x-values obtained are due to a velocity dependence of the pressure effect on $\bar{e}$. For instance, if σ is velocity-independent, the time between successive collisions varies at $1/v$. The larger x-values for lower pressure, where the pressure effect is not approaching saturation, support the latter explanation.

If Eq. (7) is valid and σ is proportioned to v^n, the above values of x are correct if $n=-1$; if $n<-1$ they are too small; if $n>-1$ they are too large.

Bohr[6] estimated the charge of a heavy ion by assuming the ion to be stripped of all its orbital electrons that have smaller velocities than the translational velocity of the ion. The correct application of Bohr's estimate requires that one consider the electron orbital velocities of an ion rather than of an atom as the orbital velocities are different in the two cases.

The Bohr assumption was applied to an ion of nuclear charge 38, and the result is shown by the dotted curve of Fig. 11.[9] Two properties of fission fragments smear this curve out in the regions of the sharp breaks. These are the energy distribution and the nuclear-charge distribution of fragments of a single mass. The dot-dash line shows the smearing effect due to the measured width of the energy distribution[7]; the solid curve shows the additional smearing effect due to the measured width of the nuclear charge distribution.[1] The data points in Fig. 11 are experimental points for mass-97 fragments. The data were corrected for the pressure effect before they were plotted.

The curves in Fig. 11 provide an experimental test of the Bohr assumption and show it to hold only as a first approximation. The structure in a curve of $\bar{e}$ *vs* v, obtained by assuming $\bar{e}$ to be equal to the number of electrons whose orbital velocity is lower than that of the ion, is not verified by experiment.

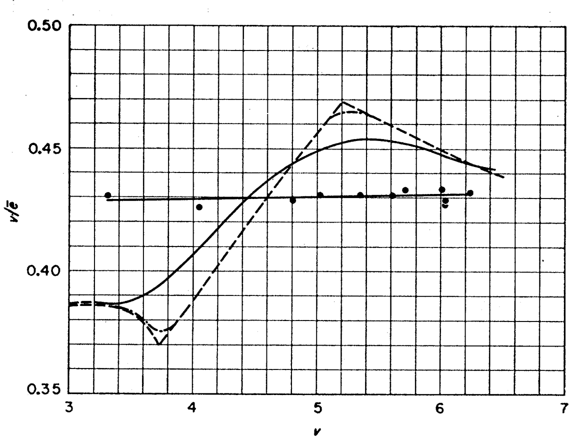

FIG. 11. Ratio of velocity to charge as a function of velocity for fission fragments of nuclear charge 38. The unit of velocity is $e^2/\hbar$; the unit or charge is the electron charge. The dotted line shows the results obtained by applying Bohr's assumption (see text); dash-dot line shows the smearing effect of velocity distribution; solid curved line shows the additional smearing due to nuclear charge distribution.

[9] This curve was calculated from a curve prepared by J. Neufeld of this Laboratory in which charge is plotted as a function of velocity for an ion of nuclear charge 38. Dr. Neufeld kindly permitted the authors to use this curve which is one of several he plans to publish in the near future.

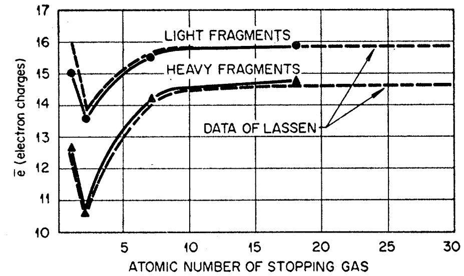

FIG. 12. Equilibrium charges of fission fragments with initial energies as functions of atomic number of stopping gas.

Type of Gas

The equilibrium charges of median light and heavy fragments, with initial velocities, are plotted as a function of atomic number of the stopping gas in Fig. 12. These values are corrected for the pressure effect. The results obtained by Lassen[4] are also shown by the dotted lines. Lassen's data are extended to higher atomic numbers on the basis of a measurement of $\bar{e}$, for which xenon was the stopping gas. There is good agreement between the two sets of measurement except for the light fragments in H_2.

From Fig. 6 it is seen that the curves of Fig. 10 are repeated for lower fragment velocities with very little change except the ordinate values.

Apart from some fluctuations for low atomic number there does not appear to be any appreciable variation of $\bar{e}$ with atomic number of the stopping gas. This is in agreement with the theoretical conclusions of Bohr and Lindhard[5] that the main contribution to the electron loss process arises from the direct action of the bare nucleus in light atoms and the atomic core (the nucleus partially shielded by electrons with velocities larger than that of the fragment) in the case of heavier atoms.

Mass of Fragment

The charges of median light (mass $\sim$95) and median heavy (mass $\sim$139) fragments are shown as functions of fragment velocity in Fig. 6. For the same velocity the ratio of $\bar{e}$ for the heavy fragments to $\bar{e}$ for the light fragments is about 1.25 which is $(139/95)^{0.6}$. However, the nuclear charges of the median light and heavy fragments are about 38 and 54, respectively; the ratio $\bar{e}_H/\bar{e}_L$ equals $(54/38)^{0.64}$. This leads to the conclusion that the $Z^{0.32}$ dependence of $\bar{e}^{(1)}$ is not valid over a large range of nuclear charge.

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