The Capture and Loss of Electrons by Fission Fragments*

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We calculate the effective charge of fission fragments passing through a low pressure gas. We first compute, on a classical basis, the cross section for capture of an electron by a fission fragment of relatively arbitrary charge and velocity. We then develop a rough theory which gives the cross section for loss of an electron through interaction with a gas atom. From these two cross sections we readily obtain the effective charge. The effective charge of a fragment will increase with the pressure of the substance through which it passes. We note the probable importance of anomalous effects in hydrogen and helium. We find general agreement between theoretical predictions and experimental results.

I. INTRODUCTION

IN this paper we will consider the problem of calculating the average number of electrons which a fission fragment possesses as a function of its nuclear charge, velocity, and the density and atomic weight of the material through which it passes. We take the term "effective charge of a fragment" to mean the fragment's nuclear charge minus the average number of electrons which it has under given conditions.

The problem of determining the effective charge of a fission fragment has been discussed by Bohr;[1-3] Lamb;[4] Knipp and Teller;[5] and Brunings, Knipp, and Teller.[6] All of these authors have agreed in treating the electrons of the fragments as a Fermi-Thomas gas. They then made physically reasonable but relatively arbitrary assumptions to obtain the effective charge as a function of fragment nuclear charge and velocity. It is the aim of the present paper to develop a theory which is somewhat less arbitrary or ambiguous than the previous formulations. It is believed that the calculations here made have some claim to quantitative accuracy.

Let us consider the passage through a gas of a fragment of nuclear charge $40e$ or $50e$, effective charge of the order of $10e$, and velocity of the order of $4\alpha c$, where $\alpha=(137)^{-1}$ and c is the velocity of light. Inasmuch as the effective charge of a fragment will at any instant be determined by a statistical balance between the fragment's capture and loss of electrons, we wish to calculate the cross section for the fragment to capture an electron from the gas atom, and the cross section for the fragment to lose an electron in an interaction with a gas atom. Since we are most interested in the ratio of these cross sections, it may be important to treat them by similar methods; we may thus hope that some inaccuracies inherent in the necessarily approximate methods employed will not affect the ratio.

Let us for the present consider that we have the fragment passing through a gas of moderate atomic number (of order 10: The special cases of hydrogen and helium will receive some attention in Sec. IV). Let us further assume that the successive collisions of a fragment with gas atoms are independent. This means that there is sufficient time between collisions in order that fragment electrons which were either raised to highly excited states or newly captured in such states in the last previous collision, will be able to decay to some of the lowest available fragment states before the next collision. We thus ignore ionization in two or more steps and take any captured electron to be no different from other fragment electrons in subsequent collisions. It appears that we hereby limit ourselves to passage through rather rarified gases as will be seen in Sec. V.

Theories of the capture of electrons by α-particles and protons have been developed classically by Thomas,[7] and quantum mechanically by Oppenheimer[8] and Brinkman and Kramers.[9] For our case the fission fragment will have an effective charge of the same order as the nuclear charge of the gas atom, and it is clear that a perturbation or Born approximation method is not valid for a treatment of the capture problem. On the other hand, the electrons will necessarily be captured in fairly high quantum states for which the picture of classical orbits has some validity, and thus a classical approach suggests itself. It is then important to make sure that we can define the positions of our electrons with sufficient accuracy to be able to introduce meaningful impact parameters. It must not be necessary to define electron momenta to an accuracy which would imply position uncertainties as large as the impact parameters. We shall see that, although this limitation is serious, it is generally possible to define the momentum and position of a gas atom electron well enough to justify qualitatively a classical approach.

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[1] N. Bohr, Phys. Rev. **58**, 654 (1940).
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[4] W. E. Lamb, Jr., Phys. Rev. **58**, 696 (1940).
[5] J. Knipp and E. Teller, Phys. Rev. **59**, 659 (1941).
[6] Brunings, Knipp, and Teller, Phys. Rev. **60**, 657 (1941).
[7] L. H. Thomas, Proc. Roy. Soc. (London) **114**, 661 (1927).
[8] J. R. Oppenheimer, Phys. Rev. **31**, 349 (1928).
[9] H. C. Brinkman and H. A. Kramers, Proc. Acad. Sci. Amsterdam **33**, 973 (1930). See also J. D. Jackson and H. Schiff, Phys. Rev. **89**, 359 (1953).

A quantum-mechanical perturbation theory would seem to have more validity for the calculation of the loss cross section, since the influence of the gas atom on the tightly bound fragment electrons is generally small. But in view of the large amount of momentum that must be given to a fragment electron to remove it from the fragment, we may hope that the large allowable uncertainty in electron momentum may permit one to localize the electron position sufficiently to somewhat justify also a classical approach. To this we shall turn our attention in Sec. III, where it will be further argued that the gas atom may be approximately treated as a Coulomb potential, in which happy case classical and perturbation calculations coincide.

II. THEORY OF CAPTURE

It is difficult for a fragment to capture a free electron. This it can do only if the electron collides inelastically with a fragment electron or suffers a radiative collision; otherwise the total energy of the electron in the rest system of the fragment is non-negative, and the electron moves in a hyperbolic orbit. However, it is different for an electron bound in a gas atom: When fragment and gas atom are separated by a distance large compared to atomic dimensions, the fragment exerts virtually no influence on an electron bound in the gas atom. Since the atom is electrically neutral, it will experience only a small net polarization force and may approach quite close to the fragment without having its electrons particularly accelerated toward the fragment. Hence, when the electron is finally liberated from the gas atom, it may have a sufficiently small velocity so that its total energy in the fragment rest system is negative, allowing it to be captured.

During the early stages of a fragment-atom collision, the external force on a gas-atom electron is small compared to the force binding the electron in the atom. For any electron that is to be captured, there will come a time during the collision when the force on the electron caused by the fragment becomes larger than the binding force, and thereafter the force between gas atom and electron is of secondary importance. Thus the following treatment suggests itself as a simplification of the difficult two-center problem posed by capture: In considering the possible capture of any electron we ignore the effect of the fragment on the gas atom, i.e., polarization, until the force on the electron in question caused by the fragment is equal to the force binding the electron in the atom. Thereafter we ignore the effect of the gas atom and consider the newly liberated electron to be captured, if and only if its total energy in the rest system of the fragment is negative.

In this approach we use the Fermi-Thomas model of the atom in order to obtain the space and velocity distributions of electrons in the gas atom. We are favored in this choice because the outermost electrons, which are poorly described by the Fermi-Thomas model, are

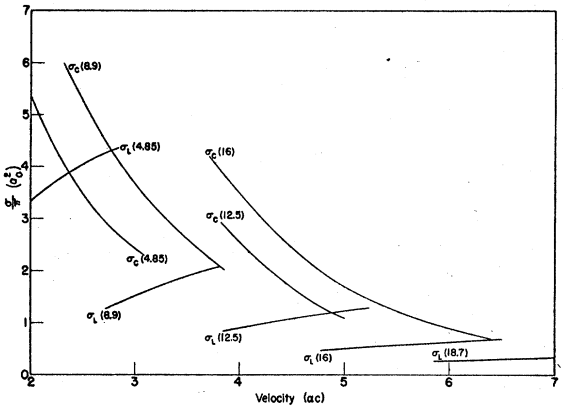

FIG. 1(a). Cross sections for capture σ_c and loss σ_L of an electron by fission fragments of nuclear charge $40e$ passing through low density oxygen; as functions of fragment velocity. The number of electrons z which the fragment lacks for electrical neutrality is indicated as $\sigma(z)$.

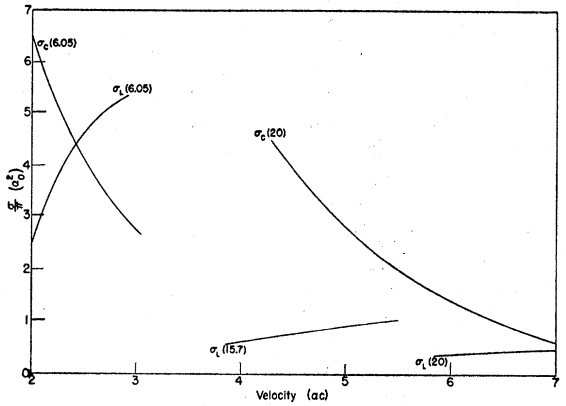

FIG. 1(b). Capture and loss cross sections for fragments of nuclear charge $50e$ in oxygen.

seldom captured and because several electrons contribute almost equally to the capture cross section, thus implying no inconsistency in a statistical model.

Consider the interaction of a fragment of effective charge z and velocity $\mathbf{v}$ with an electron which has a velocity $\mathbf{v}_e$ in its own Fermi-Thomas atom and is subject to a binding force $\mathbf{F}_e$. Assuming that the electron is liberated at fragment-atom separations large compared to the dimensions of the fragment or gas atom, the force on the electron due to the fragment will be equal to the binding force when gas atom and fragment are separated by r_0 such that $F_e=zr_0^{-2}$. At this separation we assume that the electron is liberated, and thereafter we ignore the gas atom. The electron's potential energy is now $-z_0r_0^{-1}$, and its kinetic energy in the fragment rest system is $\frac{1}{2}m(\mathbf{v}+\mathbf{v}_e)^2$. It is convenient to introduce v_0, the minimum speed relative to the fragment which an electron at distance r_0 from the fragment nucleus must have in order to escape. Then the condition for capture is $|\mathbf{v}_e+\mathbf{v}|\leqslant v_0$. From simple geometry it follows that the probability of capture, $P(v, v_0, v_e)$, may

be written

$$\begin{aligned} P(v, v_0, v_e) &= 0, && |v-v_e| \geqslant v_0 \\ &= 1, && v+v_e \leqslant v_0 \\ &= \tfrac{1}{2}[v_0^2-(v-v_e)^2]/2vv_e. && v-v_e \leqslant v_0 \leqslant v+v_e \end{aligned} \quad (1)$$

For an electron which is initially located at a definite position r_e in the gas atom, the value of r_0 is determined (for given fragment charge). Our approximation consists in the assumption that *all* electrons of this type are liberated at r_0 so that the cross section for liberation of such electrons is πr_0^2. The cross section for their capture is $\sigma_c=\pi r_0^2 P(v, v_0, v_e)$. This expression must then be integrated over a distribution of electrons (r_0, v_e, v_0) consistent with the Fermi-Thomas model of the gas atom.

This calculation has been carried through for a number of fragments in oxygen, and some of the resulting total capture cross sections are plotted in Fig. 1. The calculation has also been carried far enough for fragments in argon to show that the total capture cross sections are very nearly twice the values for oxygen. It appears that over most of the periodic table, excluding very light elements, the capture cross section for any given fragment will increase with z of the gas somewhat more slowly than linearly, with the increase becoming slower with larger z.

Some general features of the capture process emerge in our calculation. First of all, as is physically evident, it is difficult for fast fragments to capture slow (outer) electrons or for slow fragments to capture fast (inner) electrons. More quantitatively, it is relatively improbable for a fragment of velocity $\simeq 4\alpha c$ to capture outer electrons which have velocities $\simeq \alpha c$, or inner electrons which have velocities $\simeq Z\alpha c$ if Z, here the atom nuclear charge, is $\geqslant 7$.

Second, we note that the capture cross section for a fragment of given effective charge decreases rapidly with the fragment velocity.[10] This is because, other things being equal, a fast fragment will be less likely than a slow fragment to capture the slower electrons of a gas atom which will be liberated with large cross sections. It will capture faster gas atom electrons better than the slow fragment, but these electrons are liberated with small cross sections. Hence, the total capture cross section will decrease with velocity.

It is of interest to compare this decrease with that recently observed in the case of protons[11,12] and with the theories of Bohr,[3] and Brinkman and Kramers.[9] The observed capture cross sections for protons in hydrogen and air decrease somewhat more steeply than our capture cross sections for fragments. Bohr estimated $\sigma_c \sim v^{-6}$, which is a stronger velocity dependence than found even in the proton experiments. The calculations of Brinkman and Kramers (giving $\sigma_c \sim v^{-12}$) are not strictly applicable to any of the above cases.

For consideration of the loss calculation it is important to note how the electronic structure of a gas atom disintegrates as a fragment passes by. To get an idea of this effect, suppose we divide the Fermi-Thomas oxygen atom into eight spherical shells, each containing one unit of charge, compute the binding force on each shell and thus the liberation radii r_{0i} for the shells. The liberation radii, so computed, for the outer six oxygen electrons in interaction with a fragment of effective charge 8.9 are successively 9.2, 5.9, 2.1, 1.2, 0.90, and $0.65a_0$, (a_0=Bohr radius). Thus the oxygen atom begins to lose electrons already a long way, $\sim 10a_0$, from the fragment, and it loses of the order of half of its electrons in a collision with impact parameter a_0. Very few of these electrons can be captured by the fragment; most of them will simply escape.

We note that any attempt to refine these calculations in the framework of classical mechanics must be viewed with suspicion. If one attempted to localize the electron's position and follow its motion under the influence of the various forces more closely, one would imply uncertainties in the electron energy large compared to to the energy itself, and the whole correction would be spurious.

III. THEORY OF LOSS

We seek the probability for a fragment to lose an electron in a collision with a gas atom. For this problem a quantum-mechanical perturbation approach has some validity, at least if the nuclear charge of the gas atom is smaller than the effective charge of the fragment. One could consider the effect of the passing gas atom as a small perturbation of the electron wave function and calculate the probability for the electron to be knocked out of the fragment.

A difficulty in this calculation is the appropriate choice of the perturbing potential. As we have seen at the end of the last section, the gas atom becomes increasingly ionized as it approaches the fission fragment. In the approximation used in the last section, the successive ionizations occur at definite "liberation distances" from the fragment. Thus an oxgyen atom approaching a fragment of effective charge 8.9 will act like a singly charged ion between distances 9.2 and $5.9a_0$ from the fragment, like a quadruple charged ion between 1.2 and $0.9a_0$, etc. Given the impact parameter b and the velocity v, the perturbing potential due to the oxygen atom can therefore be written down as a function of time and its effect on the fragment electrons calculated by time-dependent perturbation theory.

While this calculation would be perfectly feasible, it was considered too cumbersome for the present work. A simpler method offers itself immediately. Consider first a fragment passing through hydrogen or helium. These light atoms will lose their electrons at relatively large distances from the fragment, and hence, during a

[10] The effect is somewhat mitigated in the physical case of capture, because the effective charge of a fragment will increase with velocity.

[11] H. Kanner and F. L. Ribe, Phys. Rev. **83**, 1217 (1951).

[12] H. Kanner, Phys. Rev. **84**, 1211 (1951).

close collision the fragment will see a proton or α-particle, vaguely accompanied by one or two electrons. We shall say more concerning the motion of these electrons in Sec. V. Anyway, it appears that appropriate perturbing potentials are those for a proton or α-particle.

With more complicated gas atoms the situation is not so clear. However, the consideration is simplified if we again treat the collision between gas atom and fragment electron by classical mechanics. This approach may be suggested by fairly large quantum numbers of the electron, and its justification will be discussed shortly. Because of the large binding energies of the fragment electrons, large amounts of momentum must be transferred from the gas atom to the electron to enable the electron to escape from the fragment. This is only possible classically for close collisions. Now the fragment electron is rather close to the fragment nucleus, and therefore close approach between gas atom and fragment electron automatically means close approach between atom and fragment itself. For such close approach, however, we know that the gas atom will be highly ionized. The perturbing potential at large distances and low degrees of ionization of the gas atom atom is therefore irrelevant, and we may consider the gas atom to have some high effective charge z *all* the time.

Quantitatively, we determine the effective charge z of the gas atom and the loss cross section σ simultaneously by the following consideration. For any *given* z, we can calculate the cross section for removal of a given fragment electron (i.e., one of given binding energy). Because we are now dealing with a perturbing Coulomb potential, this cross section is easily calculated and is essentially the same in classical and quantum mechanics; let us call it πl^2. Then, using a classical picture, essentially all passages with gas atom-electron impact parameters less than l will lead to ionization, those greater than l will not. We shall now choose z in such a way that the electric field at a distance l from the nucleus of the gas atom is just ze/l^2. In other words, z shall be an effective charge at the distance l from the gas atom nucleus, while at the same time πl^2 is the cross section for removal of the fragment electron by a Coulomb field of charge z.

If l and z are simultaneously determined in the manner described, it is found that z corresponds to about the highest degree of ionization which the gas atom can reach, barring direct hits on the fragment. An electron which passes at distance l from the gas atom will not appreciably penetrate the remaining electronic structure of the atom, hence will be perturbed by what is very nearly a pure Coulomb field. Electrons that pass even closer will enter regions of the "gas atom" where the field is stronger than assumed, but they will be lost in either picture.

The question arises as to the validity of these classical arguments. For its escape to be possible, a fragment electron must be given a large amount of energy. This means that a correspondingly large uncertainty in the initial electron momentum may be tolerated and that we can therefore localize the electron to a region small compared with, or of the order of, that given by relevant impact parameters without implying a prohibitively large momentum uncertainty. Interactions which produce loss will correspond to impact parameters of the order of $2Z_e^2/mv^2$, where $Z\leqslant$ the nuclear charge of the gas atom. For oxygen or heavier gases this will be of the order a_0, and the classical approach will have about as much validity as in the case of capture.

We now turn to the quantitative calculation of the loss cross section. Let $\hbar\mathbf{k}_e$ denote the initial momentum of the fragment electron relative to the fragment nucleus and $\hbar\mathbf{k}_m$ the momentum of an electron stationary in the fragment relative to the gas atom. Then $\hbar\mathbf{k}=\hbar(\mathbf{k}_m+\mathbf{k}_e)$ is the initial momentum of the fragment electron relative to the gas atom. If $\hbar\mathbf{k}_f$ is the final momentum of the electron relative to the gas atom, then $k_f=k$, and the momentum change $\hbar q$ will be given by $2\hbar k\sin(\theta/2)$, where θ is the angle of scattering. The final momentum relative to the fragment is $\hbar\mathbf{k}^1=\hbar\mathbf{k}_f-\hbar\mathbf{k}_m$. Now let $\hbar k_0$ be that minimum value of the momentum which the electron in question must have relative to the fragment in order to be free. Then the condition for ionization or loss of this electron which must be imposed on its final momentum is $k^1\geqslant k_0$. All these momenta are shown in a k space diagram in Fig. 6.

For a Coulomb potential of strength z, the differential scattering cross section is

$$d\sigma/d\Omega=(2mz_e^2/\hbar^2)1/q^4. \tag{2}$$

To find the loss cross section for an electron characterized by $(\mathbf{k}_e, k_0)$ we must integrate $d\sigma$ for all $k^1\geqslant k_0$. This integration is outlined in the Appendix. We may then calculate the average loss cross section as a function of the distance r of the fragment electron from the fragment nucleus. From the Fermi-Thomas model of the fragment we compute three parameters as a function of r, namely, (1) $k_0(r)$, (2) $k_a(r)$, where $\hbar k_a(r)$ is the maximum value of the initial momentum which an electron at r can have, i.e., the momentum of an electron at the Fermi level, and (3) $D(r)$, where $D(r)dr$ is the number of fragment electrons between r and rdr.

The loss cross section per electron as a function of r is shown in the Appendix to be

Case I.—$(2k_m-k_0\leqslant k_a)$

$$\sigma_L(k_m, k_0(r), k_a(r))$$

$$=\frac{3\pi z^2}{4a_0^2k_a^3k_m}\left[\left(\frac{2(k_0^2+k_m^2)}{k_0^2-k_a^2}-1\right)\ln\left(\frac{k_m+k_a}{k_0-k_m}\right)+\frac{k_m}{k_m+k_a}-\frac{2k_m}{k_0-k_a}-\frac{2k_m-k_0}{k_0-k_m}\right]. \tag{3a}$$

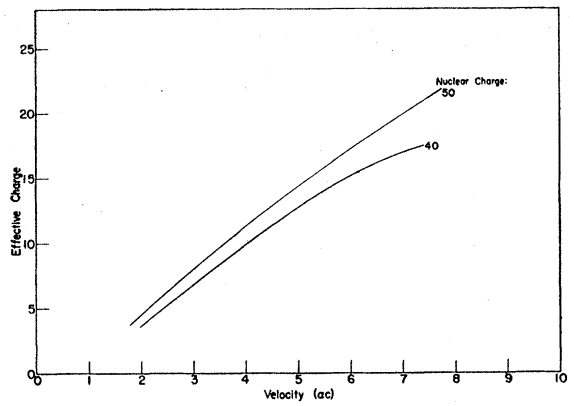

FIG. 2. Effective charges of fragments of nuclear charge $40e$ and $50e$ passing through low density oxygen, as functions of fragment velocity.

Case II.—$(2k_m-k_0\gtrsim k_a)$

$$\sigma_L(k_m, k_0(r), k_a(r))$$
$$=\frac{3\pi z^2}{4a_0^2k_a^3k_m}\left[\left(\frac{2(k_m^2+k_0^2)}{k_0^2-k_a^2}-1\right)\ln\left(\frac{k_m+k_a}{k_m-k_a}\right)-k_ak_m\left[\frac{4}{k_0^2-k_a^2}+\frac{2}{k_m^2-k_a^2}\right]\right]. \quad (3b)$$

In addition, $\sigma_L=0$ if $2k_m-k_0<-k$.

Finally, the total cross section for loss by the fragment is obtained by integrating the cross section per electron multiplied by the electron density over the fragment volume,

$$\sigma(k_m, z)=\int_0^{r_{\max}}\sigma_L(k_m, k_0(r), k_a(r))D(r)dr. \quad (4)$$

The integrand of Eq. (4) has a fairly pronounced maximum. This is because $D(r)$ is very small for r near $r_{\max}$ and σ_L decreases rapidly as r becomes smaller. The charge of the gas atom, z of Eqs. (3), is determined by demanding that the loss cross section per electron at the maximum of the integrand in (4) is equal to the geometrical cross-sectional area of that region of the gas atom within which there are one nucleus of charge Z and $Z-z$ electrons. As remarked earlier, the value of z thus obtained is in each case about equal to the number of gas atom electrons which have been liberated under the influence of the fragment, at the time when the gas atom and fragment are separated by a distance about equal to the fragment radius. We may essentially conclude that a fragment electron is with high probability removed from the fragment, if and only if it is "hit" by the gas atom, i.e., if the gas atom passes close enough to the fragment so that the fragment electron penetrates the remaining atom electronic structure.

Using Eqs. (3), (4) and the above criterion for determining z, we have computed loss cross sections for various fragments in oxygen. Some results are shown in Fig. 1. The values of effective z for oxygen which are obtained vary from about 2 with fragments of effective charge about 5 to about 6 for fragments of effective charge approximately 20. Loss cross sections have also been computed for fragments in argon, and these are found to be larger than those for oxygen by a factor which for effective fragment charge $\gtrsim 8$ is about 2.2 and is smaller for smaller fragment effective charge.

It is worth noting that the loss cross section for a *fixed* effective charge increases with fragment velocity in the region considered. This is apparently contrary to observations with protons[12,13] and the Born approximation, where σ_L decreases with velocity. However, since in reality the fragment effective charge increases with velocity, the loss cross section for a fragment will actually decrease with increasing fragment velocity.

IV. THE EFFECTIVE CHARGE

If we know the capture and loss cross sections as functions of fragment charge and velocity, it is a simple matter to find the most probable number of electrons in a fragment as a function of its velocity. We shall see that the most probable number is essentially the average number, and hence from the intersections of the capture and loss curves in Fig. 1 we may obtain effective charge as a function of velocity and nuclear charge. The effective charges of fragments of nuclear charges 40 and 50 in oxygen are plotted as functions of the velocity in Fig. 2.

Our results may be compared with previous calculations, which are summarized in reference 6. Bohr estimated the effective charge z of a fragment of nuclear charge Z to be $z=Z^{\frac{1}{3}}v$, where v is measured in units of αc. We see that Bohr's values of effective charge are greater than ours by factors of 1.30 or more. The effective charges calculated in (5) and (6), by Knipp, Teller, and Brunings, are greater than ours by the order of at least 30 percent for fragment velocities greater than $3\alpha c$, but are in better agreement with ours for smaller velocities. It appears that their second choice of "critical electron velocity" gives much better (i.e., lower) values of the effective charge for high fragment velocities. Our results are in rather close agreement (of the order of 10 percent) with those of Lamb.[4]

The effective charges of fragments in other gases than oxygen but of comparable atomic number are about the same as those in oxygen. Thus our calculations indicate that the effective charge in argon will be about 2 percent larger than in oxygen when the effective charge >8; smaller for lower values of the effective charge. We may conclude that for a large range of atomic weights of stopping gases, the effective charge is essentially independent of the stopping gas.

From our cross sections, we may deduce the prob-

[13] J. H. Montague, Phys. Rev. **81**, 1026 (1951).

ability for a fragment of velocity v and nuclear charge 40 or 50 to have any arbitrary effective charge. For any definite velocity and nuclear charge of our fragment, we have a most probable value of fragment charge, call it z_0, such that $\sigma_c(z_0)\simeq\sigma_L(z_0)$, where $\sigma_c(z)$ is the cross section for capture of an electron by a fragment of effective charge z, and $\sigma_L(z)$ is the cross section for loss of an electron by a fragment of effective charge z. If $N(z)$ is the number of fragments having effective charge z, the equilibrium distribution of $N(z)$ will be such that the probability of going from z to $z+1$ will equal the probability of going from $z+1$ to z, or

$$N(z)\sigma_L(z)=N(z+1)\sigma_c(z+1). \tag{5}$$

To the extent to which $\sigma_c(z)$ and $\sigma_L(z)$ may be represented by curves of exponential form, $N(z)$ will have a Gaussian form about z_0.[14] Thus, if we set

$$\sigma_L(z)=\sigma_{L0}\exp[\beta(z-z_0)],$$
$$\sigma_c(z)=\sigma_{c0}\exp[\alpha(z-z_0)],$$

we find

$$N(z)=N(z_0)\exp[\tfrac{1}{2}(\beta-\alpha)(z-z_0)^2], \tag{6}$$

provided we set $\sigma_{L0}=\sigma_{c0}\exp[\frac{1}{2}(\alpha+\beta)]$.

From our cross sections and (5) we may also compute $N(z)$, and this has been done for a fragment of nuclear charge 40 and velocity $4.75c$. The results are indicated in Fig. 3 and compared with the Gaussian of Eq. (6) which is seen to be a good fit. We find that most probable, average, and rms values of z agree to within about 2 percent. It appears that for a wide range of fragments the equilibrium distribution $N(z)$ is roughly a Gaussian with width at half-maximum equal to one-third the most probable value z_0.

V. HYDROGEN AND HELIUM

The cross section for loss of an electron by a fragment interacting with a hydrogen atom presents no new problems, for this may be calculated using Born approximation with the perturbing potential of a bare proton. The results of Sec. III may be taken over with the z of the gas atom put equal to 1.

To obtain a simple picture of *capture* in hydrogen, we shall start by adapting our previous treatment of capture from a many electron atom. That is, we retain the notion that before liberation the electron moves in an unperturbed fashion, while after liberation it is aware only of the existence of the fragment. However, we take into account the velocity distribution of the hydrogen electron in detail instead of using the Fermi distribution. The probability that an electron in the hydrogen ground state has velocity between v_e and v_e+dv_e is

$$P(v_e)dv_e=\frac{32}{\pi}\frac{v_e^2}{(1+v_e^2)^4}dv_e. \tag{7}$$

[14] This Gaussian was suggested by Niels Bohr.

We then may multiply $P(v_e)dv_e$ by Eq. (1) and integrate over electron velocities to get a total capture cross section in terms of the liberation distance.

The velocity distribution for large v_e behaves as v_e^{-6}. This makes it very difficult for a fragment of high velocity to capture a hydrogen electron. A hydrogen electron will be liberated at a large distance from the fragment and must have a high velocity relative to its own nucleus, the proton, for capture to be possible; but such high velocities are rare.

To estimate a liberation radius one may extend some considerations introduced by Lamb.[4] By considering the saddle in the potential given by a fragment and a proton, it is easy to show that capture of a hydrogen electron is quite possible for proton-fragment separations of about $(2+4\sqrt{z})a_0$. As a liberation radius, however, we wish a separation for which it is probable that the electron will escape from the proton to the fragment in the order of 3×10^{-16} sec. By estimating barrier transparencies, solid angles subtended by the saddle of escape, and times required for an electron to get through the saddle, one finds that the liberation radius is of the order of $(2\sqrt{z})a_0$.

Fortunately, the total capture cross section is insensitive to the liberation radius. For example, a variation of 25 percent in the liberation radius produces only about a 2 percent change in the effective charge of a fragment.

The cursory character of these estimates is all too apparent, but we might hope that they would give the capture cross section within a factor of 2 or so, and we have used them to compute effective charges in hydrogen. The results are indicated in Fig. 4. The curves

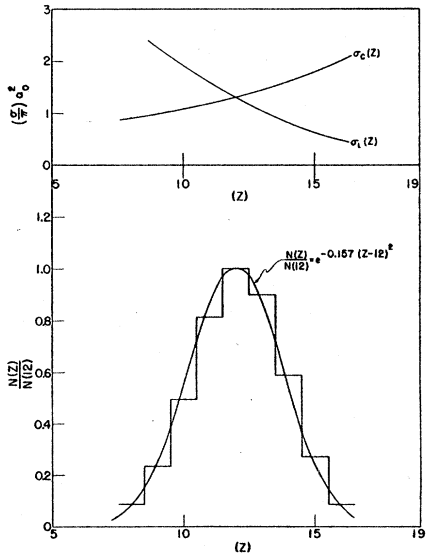

FIG. 3. Distribution of fragment charges about the most probable value, which is 12 for a fragment of nuclear charge 40 and velocity $4.75\alpha c$ in low density oxygen. The capture and loss cross sections are shown above.

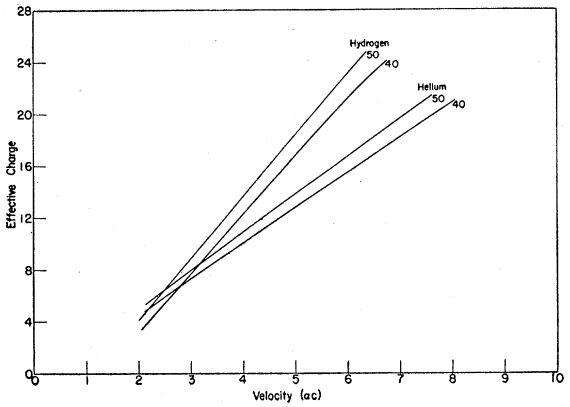

FIG. 4. Effective charges of fragments of nuclear charge $40e$ and $50e$ passing through low density hydrogen and helium; as functions of fragment velocity.

are considerably steeper, straighter, and higher than for fragments in oxygen. This is just a reflection of the difficulty which a fragment, especially a fast one, has in capturing a hydrogen electron. The curve is also, as will be seen in Sec. VI, in marked disagreement with experiment, being too high at least for high fragment velocities.

It may be that because of the smallness of the capture and loss cross sections, new effects will show up in the interactions of fragments with hydrogen atoms. In particular, the capture of liberated electrons through inelastic collisions with the fragment may be of importance. Thus a liberated electron may pass through the outer regions of a fragment and interact with a fragment electron so that in the final state (a) both electrons have been raised to the continuum and are free, (b) one electron is free and the other bound, or (c) both electrons are bound in the fragment. The first process would lead to loss but should be much smaller than proton-induced loss because of the low kinetic energy of the liberated hydrogen electron. The second process does not contribute to net capture or loss. The cross section for a capturing collision of type (c) may be a substantial fraction of the cross section for proton induced loss and thus lead to a large increase in the total capture cross section and a considerable lowering of the effective charge. To obtain a good idea of this effect one would like to consider the interaction between an electron bound in the fragment and a free electron and to calculate the probabilities of transitions to final states in which both electrons are bound. A time-dependent perturbation calculation would seem appropriate.

The above interactions will be of considerably less importance in capture from heavier gases. This is because in a heavier gas the regular capture and loss cross sections will be larger by about the square of the number of electrons liberated from the gas atom, while the cross section for inelastic capture of liberated electrons will go only linearly with this quantity.

We have also considered the possibility of capture of a liberated electron with the emission of radiation. This appears improbable. The natural cross section for measurement of bremsstrahlung is $z^2\alpha^5 a_0^2$. Even for fragment nuclear charge of the order of 50 and photon energies of the order of 100 ev, the relevant cross sections appear too small.

The capture of hydrogen electrons may be somewhat expedited by the diatomic nature of hydrogen. To estimate this effect we may consider the momentum distribution of an electron in a hydrogen molecule or molecular ion to be that of an electron around a "proton" of charge $1.2e$ [*viz.*, H. A. Bethe, *Handbuch der Physik* (J. Springer, Berlin), Vol. 24—1]. This considerably enhances the probability of capture but lowers the effective charge *vs* velocity curve by only about 5 percent.

These considerations have been applied to helium, taking both electrons in $1s$ states and ignoring the electron-electron interaction. The effective charge curve, Fig. 4, is much lower because high internal velocities are much more probable for helium electrons.

VI. COMPARISON WITH EXPERIMENT

To be able to compare our calculations with experimental values of the effective charge, we need to know the properties of fragments from the slow-neutron fission of U^{235}. In particular, we are interested in most probable initial energies, masses, and nuclear charges, and also the expected variations in these quantities.

Leachman[15] has recently completed time-of-flight measurements which give the most probable velocities of U^{235} fragments as $6.49\alpha c$ and $4.42\alpha c$ for light and heavy fragments. This gives most probable masses of about 96 and 140 and most probable energies of 100 and 68 Mev.

These energies are considerably higher than those which have been inferred from measurements of the total ionization produced by fragments.[16–19] For example, Brunton and Hanna obtained 94.5 and 60.2 Mev. However, in converting total ionization to energy loss, it had been assumed that ω, the number of electron volts per ion pair, is the same for fragments as for α particles. Knipp *et al.*[20,21] have recently pointed out that many of the recoiling gas atoms have low ionization efficiencies and that ω should be larger for fragments than α-particles. Agreement between the time-of-flight and ionization measurements is obtained if we assume the energy-ionization ratios ω of the most probable

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light and heavy fragments to exceed ω of uranium α particles by factors of about 1.05 and 1.12, respectively. Leachman's higher energies appear to give better agreement with measured values of the total energy of fission[22] and fragment masses obtained from chemical analysis.[23]

The ionization measurements show that 15 percent variations in energy and mass are very common for both fragments.

The initial effective charges of fragments in low pressure gases have been measured by Lassen.[24] The fragments are deflected in the magnetic field of the Copenhagen cyclotron, and the effective charge is determined by combining the measured value of $H\rho$ with an estimate of the fragment momentum. When the deflecting chamber is filled with oxygen at a low pressure ($\sim$1 mm Hg), the effective charges of the light and heavy fragments are about $16.1\pm\sim0.5$ and $14.8\pm\sim0.5$, respectively. These numbers are somewhat larger than those given by Lassen himself since our value of the momentum exceeds his by about 6 percent. The corresponding numbers from our theory (Fig. 2) are about 15.5 and 13.5. In hydrogen the experimental values are about 16.7 and 13.3, while in helium they are about 14.9 and 12.2. The calculated values, from Fig. 4, are 22.5 and 16 for hydrogen and 16.5 and 12.5 for helium. Taking the molecular effect into account for hydrogen, we get about 21 and 15, so that a marked discrepancy remains. Theory and experiment agree that effective charges are higher in hydrogen than in helium, and in both gases there is a greater difference between the charges of the light and the heavy fragment than in heavier gases, the spread being greatest in hydrogen.

When the region of magnetic deflection is evacuated, it becomes possible to measure effective charges in the solid from which the fragments enter the vacuum. Lassen finds that initial effective charges in solids are about 30 percent higher than in gases. Bohr has noted that this is no doubt due to competition between the radiative decay of excited electronic states in the fragment and removal of the excited electrons by collision. It is observed that for fragments coming from solids, the heavy fragment has more effective charge than the light, contrary to the observation and theory in gases for initial fragment velocities. In general, the effective charge of a fragment increases with the pressure of the substance through which it passes.

Lassen also observes a wide distribution in value of $H\rho$ for both fragments emerging from solids. This should be and is roughly as in Fig. 3.

As a gas is introduced gradually into the deflection chamber, the fragments capture electrons from the gas atoms more or less rapidly depending on the gas pressure. It is of interest to calculate the average values of the effective charges of the most probable fragments as functions of the gas pressure. We take over the experimental values of the effective charges in solids and calculate the average values for the most probable fragments emerging from uranium into argon. Results are indicated in Fig. 5 for a path length of 20 cm and initial charges 21 and 23. There is general agreement with experiment. In particular, the equilibrium charge is approached much more rapidly by the heavy fragment.

Lassen[25] has also measured the specific ionization produced by a fragment in a gas as a function of its residual range. One may obtain information on the variation of effective charge with velocity from these measurements provided one has a theory for the specific ionization produced by such large charges. Such a theory has been developed by Bohr.[3] Using this theory and assuming that the fragment acts as a point charge, one calculates from the measured specific ionization an initial charge of about 22. The measurements were carried out at pressures between $\frac{1}{2}$ and $\frac{1}{10}$ atmosphere, but even admitting some increase of the effective charge with pressure, there is substantial disagreement with the magnetic deflection measurements and our predictions. However, if one multiplies all the charge values as obtained from specific ionization by a constant so as to give the initial effective charges in agreement with deflection experiments, one finds general agreement of the effective charge *vs* velocity with our curves of Fig. 2.

When we take into account the compound nature of the fragment, somewhat better agreement is produced. An actual fragment will lose energy more rapidly than a point charge equal to the effective charge because the actual field, which is stronger than the point Coulomb field within the fragment, will produce more ionization

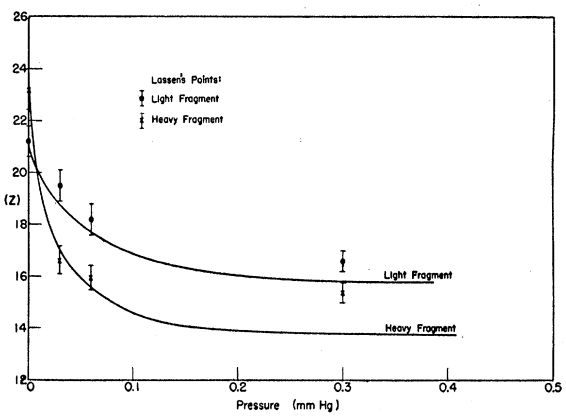

FIG. 5. Effective charges of most probable fission fragments emerging from uranium into low pressure argon as functions of pressure—measurements of Lassen described in reference 24.

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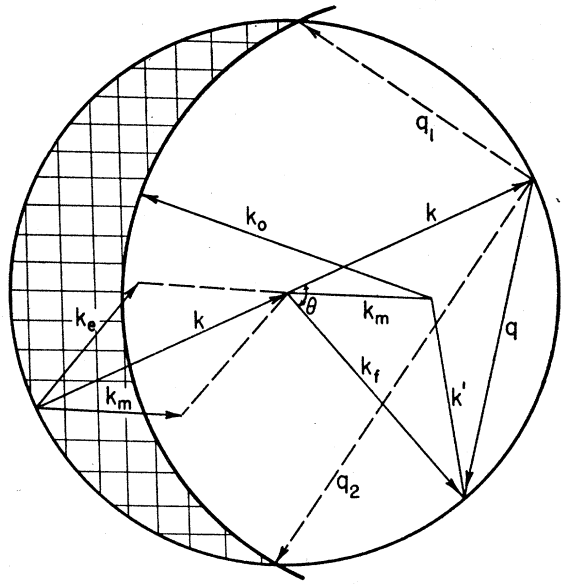

FIG. 6. Momenta used in the loss calculation: $\hbar k_e$=initial electron momentum relative to fragment nucleus. $\hbar k_m$=momentum of electron stationary in fragment relative to gas atom. $\hbar k$=initial electron momentum relative to gas atom. $\hbar k_f$=final electron momentum relative to gas atom. $\hbar q$=momentum change, $2k\sin(\theta/2)$. $\hbar k_0$=Minimum momentum relative to fragment which electron must have to be free. The integration is carried out over the crosshatched surface of the sphere. For $q \geqslant q_2$, loss is certain. For $q \leqslant q_1$, loss is impossible. For $q_2 > q > q_1$, there is a probability of loss between zero and one.

of the gas. This may add about 5 Mev per cm. Perhaps the most important new source of energy loss will arise from loss of electrons by the fragment itself. This is analagous to the charge exchange energy loss, recently discussed by Allison *et al.* for the passage of protons through hydrogen. It may amount to the order of 8–10 Mev per cm for initial fragments. Nevertheless, these two effects do not suffice to establish good agreemnnt between the specific ionization and magnetic deflection experiments.

It is a pleasure to thank H. A. Bethe for many valuable discussions. The author is also indebted to Niels Bohr, N. O. Lassen, and R. B. Leachman for communicating results in advance of publication.

APPENDIX

To get from Eq. (2) to Eq. (3), we must evaluate $\int q^{-4}d\Omega$ over all scattering angles and k consistent with fixed k_e, and k_m, and $(k^1)^2 \geqslant k_0^2$. (The k's are explained in Fig. 6.) We may write

$$Q=\int \frac{d\Omega}{q^4}=\frac{1}{k^2}\int_0^{2\pi} d\phi \int_{q_{\min}(\phi)}^{2k} \frac{dq}{q^3} \\ =\frac{1}{2k^2}\int_0^{2\pi} d\phi\left(\frac{1}{q_{\min}^2(\phi)}-\frac{1}{4k^2}\right), \tag{A1}$$

where ϕ is the azimuthal scattering angle. We wish to express $q_{\min}(\phi)$ in terms of k_e, k_m, k, and ϕ. To do this we make use of the following equations:

$$(k^1)^2=q^2+k_e^2+2\mathbf{k}_e\cdot\mathbf{q}, \\ \mathbf{k}_e\cdot\mathbf{q}=k_eq(\cos\theta_1\cos\theta_2+\sin\theta_1\sin\theta_2\cos\phi), \tag{A2} \\ k_f^2-k^2-q^2=2kq\cos\theta_2, \quad k_f=k,$$

where θ_1 is the angle between $\mathbf{k}_e$ and $\mathbf{k}$, and θ_2 is the angle between $\mathbf{q}$ and $\mathbf{k}$. It follows that

$$(k^1)^2=q^2+k_e^2 \\ +(k_eq/k)[-q\cos\theta_1+(4k^2-q^2)^{\frac{1}{2}}\sin\theta_1\cos\phi]. \tag{A3}$$

When $q=q_{\min}(\phi)$, $(k^1)^2=k_0^2$, so that from Eq. (A3) we find

$$\frac{1}{q_{\min}^2}=\frac{k_e^2}{2k^2(k_0^2-k_e^2)^2}\left[\frac{2k}{k_e}(k_0^2-k_e^2)\left(-\cos\theta+\frac{k}{k_e}\right)\right. \\ \left.+4k^2\sin^2\theta\cos^2\phi\right]+b\cos\phi(e+f\cos^2\phi)^{\frac{1}{2}}, \tag{A4}$$

where b, e, and f are constants independent of ϕ. It is important that simply $\cos\phi$ multiplies the radical for this term gives zero upon integration. If we had taken $|(\cos^2\phi)^{\frac{1}{2}}|$, $q_{\min}^2(\phi)$ would not depend on the sign of ϕ, which is incorrect. When we express $\cos\theta_1$ in terms of k_e, k_m, and k, it follows that

$$Q=\pi\left[-\frac{1}{4(k_0^2-k_e^2)^2}+\frac{1}{k^2}\left\{\frac{1}{2(k_0^2-k_e^2)}+\frac{k_e^2+k_m^2}{2(k_0^2-k_e^2)^2}\right\}\right. \\ \left.+\frac{1}{k^4}\left\{\frac{k_m^2-k_e^2}{2(k_0^2-k_e^2)}-\frac{k_e^4+k_m^4-2k_e^2k_m^2}{4(k_0^2-k_e^2)^2}-\frac{1}{4}\right\}\right]. \tag{A5}$$

To obtain the loss cross section per electron which is at distance r from the fragment nucleus, we must average Q over all velocities which the electron may have; these will be distributed isotropically and weighted proportional to $k_e^2dk_e$ up to a maximum value k_a. The average may be written

$$\frac{\sigma_L(k_m, k_a, k_0)}{4z^2/a_0^2}=\int\int Q\frac{k_ek}{k_m}dkdk_e\Big/\left[2\int_0^{k_a}k_e^2dk_e\right], \tag{A6}$$

where $\sigma(k_m, k_a, k_0)$ is the loss cross section per electron.

The limits on k and k_e must be considered with some care. We must have $k_m+k_e \geqslant k \geqslant |k_m-k_e|$, and for ionization to be possible we must have $k \geqslant k_0-k_m$. It follows that there will be three cases involving different limits of integration, the first for $2k_m-k_0<0$, the second for $0 \leqslant 2k_m-k_0 \leqslant k_a$, and the third for $2k_m-k_0 > k_a$. However, it can be proved that the first two cases may be covered by the same formula. The proof makes use of Q being an even function of k_e. $\sigma_L=0$, if $2k_m-k_0<-k_a$.

By elementary if tedious integration we find that for

Case I.—$(2k_m-k_0\leqslant k_a)$

$$\sigma_L(k_m, k_a, k_0)=A\int_{k_0-2k_m}^{k_a}k_e dk_e\int_{k_0-k_m}^{k_m+k_e}kQdk$$

$$=A\pi\left[\ln\left(\frac{k_m+k_a}{k_0-k_m}\right)\left[\frac{k_0^2+k_m^2}{4(k_0^2-k_a^2)}-\frac{1}{8}\right]\right.$$

$$\left.+\frac{1}{8}\frac{k_m}{k_m+k_a}-\frac{k_m}{4(k_0-k_a)}-\frac{1}{8}\frac{2k_m-k_0}{k_0-k_m}\right]. \quad \text{(A7)}$$

Case II.—$(2k_m-k_0\geqslant k_a)$

$$\sigma_L(k_m, k_a, k_0)=A\int_0^{k_a}k_e dk_e\int_{k_m-k_e}^{k_m+k_e}kQdk$$

$$=\frac{A\pi}{4}\ln\frac{k_m+k_a}{k_m-k_a}\left\{\frac{k_m^2+k_0^2}{k_0^2-k_a^2}-\frac{1}{2}\right\}$$

$$-\frac{\pi k_a k_m}{2}\left\{\frac{1}{k_0^2-k_a^2}+\frac{1}{2(k_m^2-k_a^2)}\right\}, \quad \text{(A8)}$$

where $A=6z^2/a_0^2k_a^3k_m$. This completes the derivation of Eq. (3).

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Radiations from Selenium[75]

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The radiations from 125-day Se^{75} have been examined by means of a thin lens spectrometer and absorption-coincidence techniques. Se^{75} was found to decay to As^{75} by orbital electron capture. The energies of ten transitions (0.066_6, 0.076_6, 0.098_3, 0.124_1, 0.138_4, 0.203_2, 0.268_8, 0.281_4, 0.307_8, and 0.405_0 Mev) were observed in the photoelectron and internal conversion spectra determined with the spectrometer. A decay scheme is proposed on the basis of the absorption-coincidence and spectrometer data. The multipole order of the 98.3-kev transition is discussed.

I. INTRODUCTION

A NUMBER of investigators[1–8] have examined the radiations from 125-day Se^{75}. From lead absorption measurements Burgus *et al.*[2] report two gamma-rays having energies of 0.18 and 0.335 Mev; Friedlander *et al.*[3] found a single gamma-ray having an energy of 0.4 Mev; Cowart *et al.*[5] report two gamma-rays with energies of 0.22 and 0.43 Mev; and Gest and Glendenin[6] found two gamma-rays having energies of 0.18 and 0.35 Mev. The three latter groups found x-rays corresponding to the K lines from arsenic, indicating that Se^{75} decays to As^{75} by orbital electron capture.

Ter-Pogossian *et al.*[7] examined the photoelectron spectra of Se^{75}, as obtained from lead and uranium radiators, in a 180° spectrometer. They report six, and possibly seven, gamma-rays with energies 0.076, 0.099(?), 0.123, 0.137, 0.267, 0.283, and 0.405 Mev. They did not obtain an internal conversion spectrum. Cork *et al.*[8] have examined the internal conversion spectrum of Se^{75} in spectrometers using photographic detection techniques. They report eleven gamma-rays with energies 0.0247, 0.0662, 0.0808, 0.0968, 0.1212, 0.1362, 0.1988, 0.2652, 0.2801, 0.3050, and 0.4019 Mev. They did not obtain a photoelectron spectrum.

DeBenedetti and McGowan[9] made a search for a metastable state in As^{75}, by means of delayed coincidences, and obtained a negative result in the range 10^{-6} to 10^{-3} sec.

Reports[10] have been made at various times on the decay and transition energies of Se^{75}, as determined

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