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Experiments on the Effect of Atomic Electrons on the Decay Constant of Be^7

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In an attempt to detect a possible influence of the atomic electrons on the radioactive decay constant of Be^7 we have measured $\lambda_{BeO}-\lambda_{Be}$ and found $(-3.0\pm 1.8)10^{-4}\lambda_{Be}$. We also describe a method to measure mean lives of radioactive substances in a time short compared with the mean life.

I. INTRODUCTION*

THE decay constant of a radioactive substance undergoing orbital electron capture is proportional, according to current beta-decay theories, to the electron density at the nucleus $|\psi(0)|^2$. This fact seems to afford a possibility of altering the nuclear decay constant λ by acting on the atomic electrons; e.g., if it were possible to completely strip a nucleus of all its electrons, it clearly could not decay by orbital electron capture and such a radioactive nucleus, as long as it stays stripped, is stable.

In a light element it does not seem impossible to achieve by chemical means an alteration of $\psi(0)$ sufficiently large to affect in a measurable way the decay constant. The most promising nucleus with which to observe this effect is Be^7 which decays by orbital electron capture with a half-period of 52.9 days, the decay being followed by the emission of a 455-kev gamma-ray.[1] In this atom the contribution to $|\psi(0)|^2$ due to the two 1*s* electrons according to Hartree[2] is, in certain units, 106.62; the contribution due to the 2*s* electrons is 4.25, making a total of 110.87. For the ion $Be^{++}|\psi(0)|^2$ according to the same author is 107.96. From these data one would expect that the decay constant of the atom would be 1.027 times greater than that of the ion. A 2.7 percent variation of the decay constant would be easily observable, but, of course, it is not very practical to keep Be^7 in a doubly ionized state for months. Another possibility that we have considered is to have the Be in a metallic state at high temperature and rely on the thermal expansion of the metal to diminish $|\psi(0)|^2$; the effect achievable in this way is, however, too small to be measured.

We finally decided to compare the decay constant of Be^7 with that of Be^7O, hoping that the change in $|\psi(0)|^2$ induced by the different chemical structure might effect λ in a detectable way. Unfortunately, a precise estimate of the variation of $\psi(0)$ in passing from beryllium metal to beryllium oxide is not available. Professor F. Seitz has kindly pointed out to us the following crude estimates: "Consider first an atom of Be gas; according to Hartree's results the ratio of the density of 2*s* electrons to 1*s* electrons near the nucleus of Be in free space is $1.86/55.9=0.033$. Now the radius of the sphere having the same volume as the beryllium atom in the metal is $2.37a_0$ (a_0 is the Bohr radius $=0.531$A). The charge outside this radius in the free atom is $1.041e$, all of which is associated with the 2*s* wave functions. This charge must be packed inside the sphere in going to the metal so that ψ^2 near the nucleus for the 2*s* electron would just about double. Thus the electron density near the nucleus should be raised by about three percent. The more accurate work of Herring and Hill[3] gives closely the same value.

"The spacing between O and Be in BeO is $1.64\text{A}=3.09a_0$. The accepted 'radius' of the oxygen ion (O^{--}) is $1.32\text{A}=2.49a_0$. Now Hartree's treatment of the neutral oxygen atom gives about $0.2e$ outside the sphere having the radius $3.09a_0$ and about $0.4e$ outside the sphere

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* A preliminary account of this work and of the results obtained was given at the 276th and 380th meetings of the Am. Phys. Soc. [Phys. Rev. **71**, 274 (1947); **73**, 743 (1947)] and in MDDC 1098. Work on the same subject has also been reported by R. Bouchez, R. Daudel, P. Daudel, and R. Muxart, J. de phys. et rad. **8**, 336 (1947).

[1] For the half-life see J. E. Hill, Phys. Rev. **57**, 567 (1940) and this paper; for the gamma-ray see K. Siegbahn, Arkiv. Mat., Astr. Fys. **34B**, 6 (1946).

[2] D. R. Hartree and W. Hartree, Proc. Roy. Soc. **149**, 210 (1935). See also W. Hartree, *ibid.* **150**, 9 (1935) for a second calculation taking into account the exchange effect. We have used the first numbers because the comparison for the Be^{++} is given only in the earlier paper.

[3] C. Herring and A. C. Hill, Phys. Rev. **58**, 132 (1940).

of radius $2.49a_0$. Hartree's results are notable in not drawing the electrons sufficiently close to the nucleus, so the true values for the neutral atom are somewhat smaller. However, this decrease is compensated by the fact that the oxygen in BeO is regarded as the double charged negative ion for which the charge outside any given sphere is certain to be larger than for the neutral atom, even if treated on the basis of Hartree's theory.

"Now in a very rudimentary theory, the charge outside either one of the two radii mentioned in the preceding paragraph would become associated with the Be^{++} ion. This charge would enter the $2s$ state and would be contained inside the ionic radius, which is taken as approximately $0.6a_0$ (i.e., the difference between the radius of the oxygen ion, namely, $2.49a_0$, and the Be—O spacing in the oxide, namely, $3.09a_0$). Now in the free beryllium atom the amount of $2s$ electron inside this radius is only $0.032e$. This is obtained from Hartree's wave functions. Hence, according to the rudimentary picture, the amount of $2s$ electron inside the "ionic radius" is increased from $0.032e$ to a value between from $0.2e$ to $0.4e$ in going from the gas to the oxide, that is, by a much larger factor than in going from the gas to the metal.

"This rudimentary picture can be looked upon in other ways. The fact that the oxygen atom contains so much charge outside the ionic radius, or outside the radius corresponding to the Be—O distance for that matter, means that the outer shell electrons on the oxygen ion in the BeO crystal also belong to the Be atom. In other words, the binding is a mixture of ionic and homopolar electron structures. Thus one can say that the result of binding is to add four electrons to the beryllium atom in the shell of total quantum number 2. This could increase the density of ψ^2 for the $2s$ electrons in the vicinity of the nucleus. It will be highly accidental if this increase is the same to within ten percent as that incurred in the metal."

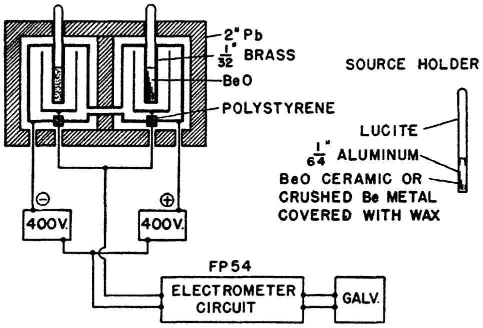

FIG. 1. Schematic diagram of balanced ionization chambers.

In view of the admitted uncertainty of these estimates, we decided to perform the experiment pushing the precision as much as reasonably feasible with the sources at our disposal.

The final result obtained is

$$\lambda_{BeO}-\lambda_{Be}=(-3.0\pm1.8)10^{-4}\lambda_{Be}.$$

It is remarkable that the change in λ is so small and whether this result is caused by an accidental compensation of various effects or has some other reason can be shown only by further experiments using different pairs of compounds or other ways of affecting $\psi(0)$. We plan to perform some of these experiments.

II. EXPERIMENTAL

A. Preparation of the Sources

The Be^7 was prepared by proton bombardment of lithium with the Crocker cyclotron. It is obviously of vital importance that the substance used be radioactively pure because otherwise the results may be falsified by different amounts of radioactive impurities, with half-lives different from Be^7, going into the metal and oxide. Accordingly, the following procedure was adopted: the lithium target was dissolved in dilute hydrochloric acid, 0.1 g of beryllium, and 10 mg each of Cu, Co, and Zn added. Sulfides were precipitated in 0.3 molar hydrochloric acid and discarded. The solution was evaporated and the residue dissolved in 12 molar sodium hydroxide. Hydrogen sulfide was added and the precipitate discarded. The solution was then acidified and the beryllium was precipitated from it by ammonium hydroxide. This operation was repeated several times. Subsequently the beryllium hydroxide was converted to the basic acetate, dissolved in chloroform, the chloroform solution extracted repeatedly with water, and finally the beryllium in the chloroform layer was reconverted to hydroxide.** This hydroxide showed only gamma-ray activity.

** These operations were performed by Dr. R. F. Leininger and Mr. G. Johnson.

The hydroxide was further diluted with normal beryllium and converted to oxide. A fraction of this oxide was converted to a button of solid beryllium metal by members of the Atomic Research Institute of the Iowa State College, to whom we are greatly indebted for their kind cooperation. This button was broken into pieces with a hammer and half of it, in the metallic state, made our sample *M*, the other half, reconverted to oxide, was our sample *C*2. Sample *C*1 was obtained from the part of the oxide that had not been converted to metal. The comparisons occurred among *C*1, *C*2, and *M*. The initial gamma-ray activity of the sources was approximately equivalent to that of 45 micrograms of radium.

TABLE I.

(Mar. 21, 47) Time	Duration, seconds	Sample in Ch 1	Ch 2	p mm sec.$^{-1}$	q mm sec.$^{-1}$
11.50	600	—	—	Background	+0.118
12.02	600	*M*	*C*1	+0.367	
12.12	600	*C*1	*M*		−0.133
12.23	600	*M*	*C*2	+0.047	
12.34	600	*C*2	*M*		+0.175
12.44	600	*M*	*C*1	+0.323	
12.55	600	*C*1	*M*		−0.158
13.06	600	*M*	*C*2	+0.033	
13.16	600	*C*2	*M*		+0.237
...	...	...	...	...	...
...	...	...	...	...	...

B. Radioactivity Measurements

For a comparison of λ_C and λ_M the decay constants of the oxide and the metal, we have used a differential method. The advantages over a separate measurement of λ_C and λ_M are several: it is possible to measure the samples at the same time and by interchanging them minimize systematic errors; the readings on the difference of the activities are always small and with ionization chambers are much more convenient to take than the readings on the activities; the time of observation is used more efficiently and it is practical to use considerably stronger samples than for single readings.

If two samples of metal and oxide have decay constants λ_M and λ_C and their initial activities $M(0)$ and $C(0)$ are approximately equal, we have

$$\delta = M(t) - C(t) = [M(0) - C(0)]^{-\lambda t} + C(0) t \Delta\lambda e^{-\lambda t}, \quad (1)$$

where $\Delta\lambda = \lambda_C - \lambda_M$ and $\lambda = \lambda_M$. We can rewrite (1):

$$\delta e^{\lambda t} = M(0) - C(0) + C(0)(\Delta\lambda/\lambda)(t/\tau), \quad (2)$$

where $\tau = 1/\lambda$ is the mean life of Be^7.

Experimentally we measure $\delta(t)$ and using (2) we obtain from it $\Delta\lambda/\lambda$. In the ideal case that $M(0) - C(0) = 0$, the maximum of δ is obtained at a time $t = \tau$ and the best time for measuring is $t = 2\tau$, i.e., the time at which the ratio between δ and its fluctuation caused by the statistical nature of the radioactive decay is maximum.

The apparatus used is schematically shown in Fig. 1. It consists of two argon-filled ionization chambers made as nearly equal as possible. The pressure in the chambers was 135.6 cm of Hg at 23°C and the density of the gas was kept constant to one part in a thousand. The two chambers were interconnected by a small copper tube to keep their pressures equal. The room in which the apparatus was located, although not temperature-controlled, was well shielded thermally. The collecting electrodes of the two chambers were connected together and to the input of a conventional electrometer circuit using a General Electric type FP-54 plyotron. The circuit was operated by the rate of drift method. The collecting potential for the chambers was taken from a dry cell battery pack delivering potentials of +400 volts and −400 volts. The polarities of the collection voltages were chosen so that the ionization current in one chamber opposed the ionization current in the other chamber. Thus, equal samples placed in the chambers would cause no drift of the galvanometer of the electrometer circuit. The samples were rigidly mounted in aluminum holders that fitted snugly into the central cavity of the ionization chambers, and we checked that possible small accidental differences in the geometry of the arrangement would not affect the results. This was done by verifying that the influence of artificial changes of geometry much larger than the changes to be expected in various runs was negligible. With this differential arrangement very small differences in the activities of two large samples can be measured. Our measuring procedure was as follows: we placed the *M* sample in chamber 1 and the *C* sample in chamber 2; the rate of drift of the galvanometer

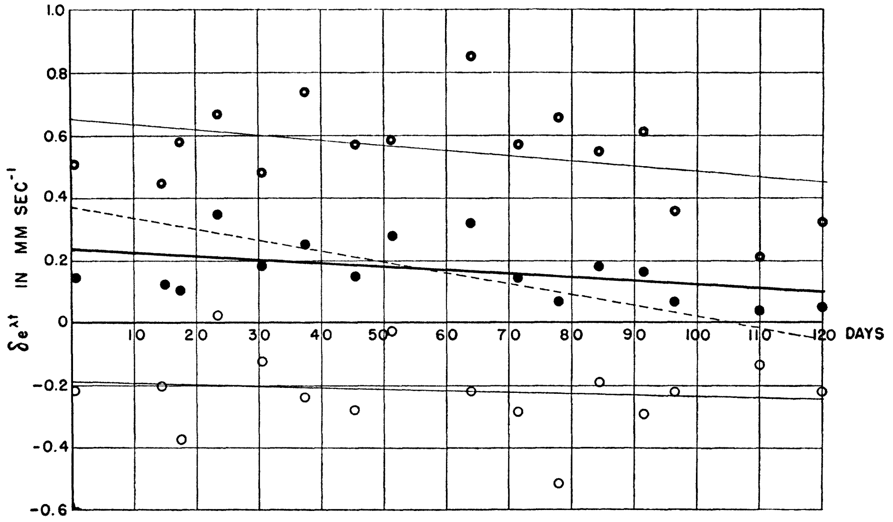

FIG. 2. Observed difference of activities of Be and BeO corrected for decay. $\delta e^{\lambda t}=M-C+M(\Delta\lambda/\lambda)(t/\tau)$ in mm sec.$^{-1}$. (1) Open circles: Be metal—BeO No. 1. (2) Open double circles: Be metal—BeO No. 2. (3) Solid circles: averages of (1) and (2). $M(0)\cong C(0)=270$ mm sec.$^{-1}$. Solid lines are best fits to experimental points. $\Delta\lambda/\lambda=-10^{-3}$ would give dotted straight line.

is then

$$p=MS_1-CS_2, \tag{3}$$

where S_1, S_2 are the sensitivities of chambers 1 and 2, respectively, and are very nearly equal. If we now interchange the samples, we have a rate of drift q

$$q=CS_1-MS_2, \tag{4}$$

from which

$$p+q=(M+C)(S_1-S_2), \tag{5}$$

$$p-q=(M-C)(S_1+S_2)\cong 2\delta S_1. \tag{6}$$

If the apparatus has a spontaneous rate of drift, which in practice, however, was extremely small, it cancels out in forming the expression (6) from the observations.

In our experiment, initially the sample of beryllium metal put in chamber 1 gave 270 mm sec.$^{-1}$ drift of the galvanometer, i.e., $M(0)S_1=270$ mm sec.$^{-1}$; when balanced with a sample of oxide in chamber 2 we obtained $p-q=0.5$ mm sec.$^{-1}$. As an example, observations from an actual run of comparisons between M and C are listed in Table I. Calculations were made by the standard least square procedure from the complete set of observations of that day. We obtained

$$2(M-C1)=0.463\pm0.038 \text{ mm sec.}^{-1},$$
$$2(M-C2)=-0.299\pm0.066 \text{ mm sec.}^{-1}$$

(the errors are standard deviations).

Observations extending over 120 days were finally calculated and plotted in Fig. 2. The value of λ adopted in the calculation is 1.309×10^{-2} day^{-1} corresponding to a half-life of 52.93 days. (For this value see Section IV.)

From the plot of Fig. 2 (all observations have approximately equal precision) we find $\Delta\lambda/\lambda=(-1.38\pm2.5)10^{-4}$ for the comparison M, $C1$; $\Delta\lambda/\lambda=(-4.76\pm3.0)10^{-4}$ for the comparison M, $C2$. If we use all the observations trusting that $C1$ and $C2$ are both free of radioactive impurities, we obtain

$$\Delta\lambda/\lambda=(-3.0\pm1.8)10^{-4}.$$

We must conclude that the $\Delta\lambda/\lambda$ observed is comparable with the error of observation and small compared with what one would expect barring accidental compensations in the change of $\lambda(0)$ between oxide and metal.

III. RADIOACTIVE FLUCTUATIONS

It is interesting to check the fluctuations on the balance of the instrument over a certain time

T. Suppose the instrument were exactly balanced with two sources each giving A mm sec.$^{-1}$ rate of drift, and call $\langle v^2 \rangle$ the average of the square of the deflections obtained in many measurements, each extending over a time T. Call d the deflection of the galvanometer produced by an elementary ionization process; then $A/d=\nu$ is equal to the number of elementary ionization processes produced by one source per second. We have then, according to the laws of fluctuations of radioactive decay,

$$\langle v^2 \rangle_{\mathrm{Av}} = 2\nu T d^2 = 2ATd. \tag{7}$$

These quantities are all directly measurable with the exception of d. However, d can be estimated from the capacity of the chambers and the roughly calculated ionization produced by a 455-kev gamma-ray in our chamber; if all the energy of the gamma-ray is spent in the chamber, we have $d=0.015$ mm and this is clearly an upper limit. In a series of measurements we had $A=270$ mm sec.$^{-1}$, $\langle v^2 \rangle_{\mathrm{Av}}=1800$ mm^2, and $T=600$ sec., from which we obtain using (7) $d=0.006$ in reasonable agreement with the estimate, if one takes into account that only a small fraction of the gamma-ray energy of each quantum is transformed into ionization in the chamber.

IV. A RAPID WAY OF MEASURING LONG HALF-LIVES

Another application of our differential ionization chamber arrangement is the rapid measurement of radioactive periods, provided one has rather strong and radioactively pure sources.

If we balanced at time 0 the substance to be measured with a practically constant source (radium), the unbalance δ at time t is in good approximation for $\lambda t \ll 1$:

$$\delta = A\lambda t, \tag{8}$$

where A is the initial activity of the source. If we can measure A and δ with comparable relative precision, Eq. (8) gives λ.

In the case of Be^7 if $\lambda t = 5\times 10^{-3}$ and A is equivalent to the activity of 100 micrograms of radium, it is easy to determine λ to ± 10 percent with our apparatus, and this result could be bettered for longer life substances. Thus, for example, we have measured $T_{\frac{1}{2}}$ of Be^7 taking observations intermittently over a period of about 5 hours and found 51.3 ± 11 days.

Using the data obtained by intermittent observations over a period of 28 hours we find 52.4 ± 1.3 days. Our best value for $T_{\frac{1}{2}}$ of Be^7, 52.93 ± 0.22 days, was obtained by comparison with a constant radium source extending over a period of 127 days. The standard error is obtained from the consistency of the measurements, but it must be borne in mind that it is possible that the measurement was affected by some slight systematic error.

Similarly, the half-life of Y^{90} was determined in three days and gave 108 days. Co^{60} was also measured intermittently (2 hours total measuring time, 0.6 mg Ra equivalent source) for 8 days and gave 5.08 years. This method could certainly be used also for much longer half-lives, such as radium itself, if one used as balancing substance uranium in equilibrium with its products. It is, however, essential that the substance to be measured be radioactively pure.

This work was done under the auspices of the Atomic Energy Commission.