The Electrostatic Interaction and Low Energy Particles in Alpha-Radioactivity

MELVIN A. PRESTON
The University, Birmingham, England
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In α-decay, after the particle has been emitted, the asymmetrical electric field of the residual nucleus can cause the α-particle to alter its total energy in conjunction with a change in the state of the nucleus. Since the ability of a particle to penetrate the potential barrier increases rapidly with its energy, the number of particles of each possible energy finally escaping will not be in accord with the decay constants predicted by the Geiger-Nuttall law.

It is found that the effect is not important (a quantitative estimate is given), except in the case where the difference between the energies of the two states is less than about 500 kev.

The investigation allows a theoretical study of the experiments in which W. Y. Chang found anomalous fine structure in the α-spectra of Po and Ra. It is found that very improbable assumptions are required to reconcile these experimental results with the theory presented here, and we have been unable to see how any additional effects connected with the nucleus could alter this conclusion.

I. INTRODUCTION

THE existence of groups of low energy α-particles in the decay spectra of polonium[1] and radium[2] has been reported by Chang. Although these groups have weak intensity compared with the main line in each case, their strength is greater than that shown on the Geiger-Nuttall curve by factors up to 10^6 in the case of Po and 10^4 for Ra. This divergence cannot be accounted for within the framework of the standard theory of α-radioactivity on the ground of non-zero angular momenta of the emitted particles, since improbably high nuclear spins would be involved.[1,2] A number of proposed explanations of the phenomenon are listed in Chang's papers. Some of these are concerned with possible mechanisms within the nucleus, but one theory which is discussed at some length in this second paper seems fairly probable and can be treated mathematically without introducing any speculation about the exact nature of nuclear forces, other than that they are of short range. This explanation supposes that after the α-particle has been emitted it can still interact with the residual nucleus and thus, by altering the nuclear state, change its own energy. Since the ease with which a charged particle can penetrate the potential barrier increases very rapidly with the energy of the particle, changes of this energy during the flight of the α-particle will clearly have some effect on the relative probabilities of its emission with the various possible energies.

After the α-particle has left the region where nuclear forces act (i.e., the nucleus), the most important mechanism which would produce such an interaction is the force acting on the particle due to the asymmetry of the motion of electric charges in the residual nucleus. It is the details of such a process that are studied here.* (Bremsstrahlung has been shown to be unimportant by Dancoff.[3])

There are two cases to be considered. We assume always that the parent nucleus is in its ground state. In the normal case, in the absence of the Coulomb barrier, the probabilities of decay to the various states of the product nucleus are of the same order because the energy of the α-particle is considerably greater than the energy differences between nuclear states. However, the energy of the α-particle has such a great effect on the penetrability of the Coulomb potential barrier that the most probable process is the emergence of the particle with its greatest possible energy, leaving the daughter nucleus in its ground state. If we now take into account the interaction mentioned above, the high energy α-particle, after penetrating part of the barrier, can excite the nucleus to a higher state and lose energy itself, finally appearing as a low energy particle. This appears to be the type of process

[1] W. Y. Chang, Phys. Rev. 69, 60 (1946).
[2] W. Y. Chang, Phys. Rev. 70, 632 (1946).

* This possibility was first suggested to the author by Professor Peierls, who in turn attributes it to a remark made some years ago by Professor Gamow.
[3] S. M. Dancoff, Metallurgical Project Report, *Short Range Alphas in Natural Radioactivity.*

envisaged by Chang in his second paper; its probability would have to be greater than that of direct decay to the corresponding excited states. Such a process should occur in the decay of all α-emitters; hence, all should have a low energy group for each excited state of the residual nucleus. However, we shall see that the effect is not, in general, great enough to be distinguished from the direct decay, and is certainly not sufficient to account for the lines observed by Chang.

The second case is one in which, for some reason connected with nuclear structure, the direct decay to the ground state of the daughter nucleus is forbidden. The nucleus is left in an excited state, and the α-particle begins its career with the corresponding low energy. However, because of the interaction, the nucleus can now fall to its ground state, imparting the extra energy to the α-particle instead of emitting a γ-ray. This process will also clearly affect the relative probabilities of emission. With our present knowledge of the constitution of heavy nuclei, it does not seem possible to assess with any certainty the likelihood of the ground state transition being forbidden. It is not forbidden by any of the usual selection rules, such as angular momentum or parity, because these would make equally improbable the transition after emission. Nevertheless the "accidental" vanishing of a usually appreciable factor might make the direct transition improbable. However, we shall see in any case that this process is also incapable of explaining Chang's results.

Indeed, we shall see that there appear to be considerable difficulties attached to any theoretical explanation of Chang's spectra, and the indications are that, whatever the reason for the results, it is not to be found in an effect associated with the polonium or radium nuclei.* In this connection we may also mention that Zajac, Broda, and Feather[4] have re-investigated the γ-radiation from polonium and have found no γ-rays to correspond to the levels suggested by Chang.

* *Note added in proof:* This conclusion has also been indicated by direct experiments on polonium reported by Dr. W. G. Wadey in Phys. Rev. **74**, 1846 (1948).

[4] B. Zajac, E. Broda, and N. Feather, Proc. Phys. Soc. **60**, 501 (1948).

II. GENERAL THEORY

To describe the system formed by the α-particle and the nucleons which comprise the residual nucleus and to allow for the electrical interaction mentioned in the introduction, we employ a many-body wave function, Ψ, which satisfies

$$[-(\hbar^2/2m)\nabla_{\mathbf{r}}^2+V(r)+H_\xi +U(\mathbf{r},\xi)-E]\Psi(\mathbf{r},\xi)=0. \quad (2.1)$$

This is written to describe the relative motion, with origin at the centroid of the residual nucleons. m is the reduced mass (practically that of the α-particle). $\mathbf{r}$ is the position vector of the α-particle, ξ denotes all the coordinates (including spin, etc.) of all the other nucleons, $\nabla_{\mathbf{r}}^2$ denotes the Laplacian operator on $\mathbf{r}$, and H_ξ is the Hamiltonian of the other nucleons. E is the total complex energy, i.e., its imaginary part is $-\frac{1}{2}i\hbar\lambda$, where λ is the total decay constant. $V(r)$ is the potential function for the α-particle which is generally used in theories of α-decay. Outside the nucleus $V=2Ze^2/r$, and inside it forms a well of some shape which we shall not need to specify—in fact we do not even use the assumption that it can be expressed as a function of r inside the nucleus. U is the potential due to the nuclear charge asymmetry. For $r>r_0$ (the nuclear radius),

$$U=\sum_i(2e^2)/(|\mathbf{r}-\mathbf{r}_i|)-2Ze^2/r$$

$$=2e^2\sum_i\sum_{k=1}^{\infty}(r_i^k/r^{k+1})(2/2k+1)^{\frac{1}{2}}P_k(\cos\Theta_i), \quad (2.2)$$

where the sum $\sum_i$ is over all the protons in the product nucleus, Θ_i is the angle between $\mathbf{r}$ and $\mathbf{r}_i$, and P_k is a normalized Legendre function. The form of U for $r<r_0$ is also immaterial to our discussion.

The states of the residual nucleus can then be specified by a complete set of normalized orthogonal eigenfunctions $u_m(\xi)$, and the total wave function Ψ can be expanded as a series in these functions.

$$(H_\xi-E_m^{(N)})u_m(\xi)=0, \quad (2.3a)$$

$$\int u_m u_m^* d\xi=1, \quad (2.3b)$$

$$\Psi=\sum_m \phi_m(\mathbf{r})u_m(\xi). \quad (2.4)$$

Then substituting for Ψ in (2.1), multiplying by u_n^*, and integrating over ξ, we find

$$[-(\hbar^2/2m)\nabla^2+V(r)-(E_n^{(N)}-E)]\phi_n(\mathbf{r}) = -\sum_m \phi_m(\mathbf{r})U_{nm}(\mathbf{r}), \quad (2.5)$$

where

$$U_{nm}(\mathbf{r})=\int u_n^*(\xi)U(\mathbf{r},\xi)u_m(\xi)d\xi. \quad (2.6)$$

Also $E-E_n^{(N)}=E_n$, the complex energy of the nth α-particle group. We take it that the states are enumerated so that each $\phi_n(\mathbf{r})$ corresponds to a fixed angular momentum l_n of the α-particle.** Thus

$$\phi_n(\mathbf{r})=r^{-1}f_n(r)\sum_{M=-l_n}^{l_n} c_M \times P_{l_n}{}^M(\cos\theta)\exp(iM\varphi). \quad (2.7)$$

We now apply the addition theorem for spherical harmonics to the factor $P_k(\cos\Theta_i)$ in (2.2), put this result in (2.6), and find for $U_{nm}(\mathbf{r})$ the following expression:

$$2\sum_{k=1}^{\infty} r^{-(k+1)}\left\{C_{k0}{}^{nm}P_k(\cos\theta) +2\sum_{s=1}^{k} P_k{}^s(\cos\theta)\left(C_{ks}{}^{nm}\cos s\varphi +S_{ks}{}^{nm}\sin s\varphi\right)\right\}, \quad (2.8)$$

where

$$C_{k0}{}^{nm}=(2/2k+1)\int u_n^* e^2\sum_i r_i{}^k P_k(\cos\theta_i)u_m d\xi, \quad (2.9)$$

$$C_{ks}{}^{nm}, S_{ks}{}^{nm}=(2/2k+1)\int u_n^* e^2\sum_i r_i{}^k \times P_k{}^s(\cos\theta_i)\begin{matrix}\cos\\ \sin\end{matrix} s\varphi_i u_m d\xi, \quad (2.10)$$

and $(r_i, \theta_i, \varphi_i)$ are the coordinates of the ith proton in the nucleus. Then, substituting (2.7) in (2.5), multiplying through by $P_{l_n}{}^M(\cos\theta)\sin\theta \times\exp(-iM\varphi)$, integrating over θ and φ for each M, and adding the resulting equations, we get

$$\frac{\hbar^2}{2m}\left(\frac{d^2}{dr^2}f_n-\frac{l_n(l_n+1)}{r^2}f_n\right)-(V(r)-E_n)f_n =2\sum_m\sum_{k=1}^{\infty} f_m a_k{}^{nm}/r^{k+1}. \quad (2.11)$$

The quantities $a_k{}^{nm}$ are each a sum of quantities which depend on integrals of the form

$$\int_{-1}^{1} P_{l_m}{}^M(\mu)P_{l_n}{}^{M'}(\mu)P_k{}^{|M-M'|}(\mu)d\mu.$$

Since these integrals vanish unless $l_m+l_n>k>|l_m-l_n|$, we have the general equations

$$\frac{d^2f_n}{dr^2}+\left\{\frac{2m}{\hbar^2}(E_n-V)-\frac{l_n(l_n+1)}{r^2}\right\}f_n =\sum_{m=0}^{N-1} f_m \sum_{k=|l_m-l_n|}^{l_m+l_n} A_k{}^{nm}/r^{k+1}$$
$$k\neq 0,\ n=0,1,\cdots(N-1). \quad (2.12)$$

N is the total number of nuclear states whose interactions are considered.

In the particular case when one of l_m or l_n is zero, there is only one possible value of k, and the summation over k on the right-hand side of (2.12) reduces to just one term.

$A_k{}^{nm}$ consists of terms of the form $C_{k0}{}^{nm}$, $C_{kM}{}^{nm}$, and $S_{kM}{}^{nm}$ multiplied by constants which depend on the c_M's of (2.7). If we take a statistical average over M of the values of $|C_{k0}{}^{nm}|$, $|C_{kM}{}^{nm}|$, and $|S_{kM}{}^{nm}|$, and call this $|Q_k{}^{nm}|$, we can then treat the problem simply as if M were zero and write

$$A_k{}^{nm}=(2m/\hbar^2)2^{\frac{1}{2}}Q_k{}^{nm}. \quad (2.13)$$

Equations (2.12) are a set of N simultaneous linear second-order equations for the functions f_n. There are thus $2N$ sets of independent solutions, a certain linear combination of which describes our problem. For large r, the equations become independent, and all the f_n's tend to functions proportional to $\exp(\pm ik_nr)$. However, the solutions must represent particles leaving the nucleus; hence we must take for f_n those solutions

** This is true if the parent nucleus has zero spin. Po and Ra are even-even nuclei. If the spin is not zero (2.4) should be replaced by $\Psi=\sum_{m,l} c_{ml}\phi_{ml}u_m$, where each ϕ_{ml} has a fixed angular momentum. However, there will clearly be a later averaging process over the c_{ml} which can affect the results only by an unimportant numerical factor.

which tend to $a_n \exp(ik_n r)$, where a_n is a complex constant. Then the ratios between the quantities $v_n|a_n|^2$ are the relative intensities of the lines in the α-particle spectrum. The condition that all the f_n's should represent outgoing waves at infinity has thus provided N of the $2N$ (complex) conditions needed to determine the particular linear combination of solutions which applies to our problem. The remaining N conditions depend on the nucleus.

The values of the f_n's at r_0 give the relative probabilities of an α-particle of each energy E_n being at the edge of the nucleus, i.e., the relative probabilities of decay if there were no potential barrier. The other conditions could be N homogeneous equations involving these functions; to determine these would require extensive knowledge of intranuclear behavior. However, we shall use our N conditions at infinity, start with the functions $f_n = a_n \exp(ik_n r)$, and integrate the equations inwards. Then the f_n's at r_0 appear as functions of the a_n's; if one set is known the other can be determined. We shall find that a good deal can be discovered without very exact knowledge of the $f_n(r_0)$.

Strictly speaking, of course, our problem is one with given boundary conditions in which we must find the eigenvalues E_n. These eigenvalues are complex; the real part is the energy of the escaping particle and the imaginary part is proportional to the decay constant. Thus the eigenvalues are usually experimentally known, and it is simpler to discuss the problem as above, using this knowledge.

We have mentioned that $f_n(r_0)$ appear as functions of a_n's. So, of course, do df_n/dr at $r = r_0$. If we assume all f_n and df_n/dr given as initial conditions at r_0, we can integrate outwards from r_0 to infinity and find the a_n's. We might therefore attempt to solve the problem by taking "reasonable" initial values, such as the presence originally of only the high energy particles (i.e., $f_0(r_0) = 1$, all other $f_n(r_0) = 0$), and then applying perturbation theory. There are two objections to this procedure. Firstly, the initial values must represent the physical fact that only outgoing waves are present. Slight errors in these values would be equivalent to a small admixture of the solution which at infinity represents an incoming wave. Inside the potential barrier this solution grows exponentially as r is increased, and hence appreciable amounts of outgoing wave might appear in the solution at large r, for relatively small initial errors.

In the second place, the perturbation technique itself turns out to be invalid here. Dancoff[3] has studied this problem by perturbation methods and has paid particular attention to the case in which the energy difference between the interacting states is small ($\lesssim 500$ kev). He found excitation probabilities greater than unity and pointed out that while this may indicate that the effect is appreciable for these energies, it also means that the perturbation theory is unreliable. The present author also originally used a perturbation theory which required that $|f_n| \ll |f_0|$. Although this condition holds at r_0 and at infinity, it was found that at certain intermediate points $|f_n|$ was as much as 100 times greater than $|f_0|$ in some cases.

We have developed a method for integrating outward which avoids these difficulties, but it is more laborious than the inward integration used in this paper, and is no more reliable (except perhaps when the differences between the energies of the states is small).

III. METHOD OF SOLUTION

Consider that set of M functions f_n which satisfy the N-equations (2.12) with the boundary conditions that, at $r = \infty$, $f_j \sim a_j \exp(ik_j r)$ and $f_n = df_n/dr = 0$, if $n \neq j$. Call these functions f_{nj}. For a fixed j, f_{nj} constitute a set of solutions of (2.12); as j runs through all values from 0 to $N-1$, we obtain N linearly independent sets. We form by a combination of these solutions another set of solutions f_n defined by

$$f_n = \sum_{j=0}^{N-1} f_{nj}, \quad n = 0, 1, \cdots (N-1). \quad (3.1)$$

These functions are the functions which provide the solution of our problem. Firstly, they have the correct asymptotic behavior for

$$f_{nn} \sim a_n \exp(ik_n r), \quad (3.2)$$

$$f_{nj} = df_{nj}/dr = 0 \quad \text{if} \quad j \neq n, \quad (3.3)$$

and, therefore, by (3.1), $f_n \sim f_{nn}$. Secondly, the N constants a_n are still at our disposal and can be fixed as discussed in the previous section.

Consider now the set j. Let $E_j=\frac{1}{2}mv_j^2$, $k_j=mv_j/\hbar$, $x=k_jr$, $\kappa_j=4Ze^2/\hbar v_j$,

$$\gamma_{nj}=1/\gamma_{jn}=k_n/k_j. \quad (3.4)$$

Since f_{jj} is the only non-zero function of the set at $x=\infty$, we employ a perturbation method for integrating inwards. Thus, treating $|f_{nj}|\ll|f_{jj}|$ and neglecting f_{nj} on the right-hand side, (2.12) becomes

$$d^2f_{nj}/dx^2+\{\gamma_{nj}^2-\gamma_{nj}\kappa_n/x-l_n(l_n+1)/x^2\}f_{nj}$$

$$=f_{jj}\sum_{k=|l_n-l_j|}^{l_n+l_j} B_k^{nj}/x^{k+1} \quad (3.5)$$

$$=F_{nj}(x)f_{jj}. \quad (3.6)$$

Here $B_k^{nj}=A_k^{nj}k_j^{k-1}$. A_k^{nj} contains in Q_k^{nj} a factor of the order of r_i^k where r_i is a position of a proton in the nucleus. Now k_jr_i is of order 1. Hence B_k^{nj} contains the factor $r_i^kk_j^{k-1}$ which is of order r_i. Even after multiplication by $me^2/\hbar^2$, this is still small. Also x is always greater than unity and is about 10 only a short distance from the nucleus. Thus in the equation satisfied by f_{jj} itself, *viz.*,

$$d^2f_{jj}/dx^2+\{1-\kappa_j/x-l_j(l_j+1)/x^2\}f_{jj}$$

$$=f_{jj}\sum_{k=1}^{2l_j} B_k^{jj}/x^{k+1}, \quad (3.7)$$

the terms on the right-hand side are, in general, negligible compared to $l_j(l_j+1)/x^2$, and may be neglected. (If $l_j=0$, the right-hand side is zero.) However, the solution to be developed below allows for the effect of the right-hand side to be estimated if desired.

We define the confluent hypergeometric function $X_n(x)$ as follows:

$$d^2X_n/dx^2+\{1-\kappa_n/x-l_n(l_n+1)/x^2\}X_n=0, \quad (3.8)$$

$$X_n\sim\exp i(x+\eta_n), \quad \text{as} \quad x\rightarrow\infty. \quad (3.9)$$

The constant η_n is defined in Eq. (4.5).

From (3.7) and (3.2), we see that (except for the small right-hand side of (3.7))

$$f_{jj}=a_jX_j. \quad (3.10)$$

We use this value in the right-hand side of (3.6) and also with

$$f_{nj}(x)=v_{nj}(x)X_n(\gamma_{nj}x). \quad (3.11)$$

Then

$$v_{nj}(\infty)=(dv_{nj}/dx)_\infty=0. \quad (3.12)$$

Using (3.6), (3.11), (3.10), and (3.8) and simplifying, the equation for v_{nj} becomes, if we suppress the suffixes on v, γ, and F,

$$X_n(\gamma x)v''(x)+2\gamma X_n'(\gamma x)v'(x)=a_jF(x)X_j(x). \quad (3.13)$$

The $'$ denotes differentiation with respect to the argument of the function. This can be written

$$(d/dx)\{X_n^2(\gamma x)v'(x)\}=a_jF(x)X_j(x)X_n(\gamma x),$$

and, therefore,

$$v'(x)=a_jX_n^{-2}(\gamma x)\int_\infty^x F(t)X_j(t)X_n(\gamma t)dt$$

and

$$v(x)=a_j\int_\infty^x\left\{X_n^{-2}(\gamma t)\int_\infty^t F(u)X_j(u)\times X_n(\gamma u)du\right\}dt. \quad (3.14)$$

The lower limits are determined by the boundary conditions (3.12). Thus the functions f_{nj} and hence, by (3.1), f_n have been found in terms of certain hypergeometric functions.

If it is desired to take account of the correction to f_{jj} caused by the non-vanishing of the right-hand side of (3.7), a similar procedure can be used giving

$$f_{jj}=a_jX_j\left\{1+\int_\infty^x dtX_j^{-2}(t)\times\int_\infty^t \sum(B_k^{jj}/x^{k+1})X_j^2(u)du\right\}. \quad (3.15)$$

The integral is always a small quantity compared to unity, for B_k^{jj} is of the order 10^{-4} or less and the remainder of the integral is at most 10. (The method of evaluation is indicated in the next section.)

The expression (3.14) can be further simplified by introducing

$$g_n(x)=\int^x X_n^{-2}(t)dt, \quad (3.16)$$

where the additive constant is to have a convenient value, to be chosen later. Then, integrating (3.14) by parts, we find

$$v_{nj}(x)=a_j\gamma_{jn}\int_\infty^x\{g_n(\gamma_{nj}x)-g_n(\gamma_{nj}t)\}\times F_{nj}(t)X_j(t)X_n(\gamma_{nj}t)dt. \quad (3.17)$$

This equation expressed a formal solution of the problem. Its usefulness depends on a number of things. Firstly, we must be satisfied that the perturbation solution is close to the actual solution. Secondly, for (3.17) to be valuable, we must have manageable explicit expressions for the hypergeometric functions which appear. Thirdly, since f_{nj} are found approximately, there is a danger that the sum $\sum_j f_{nj}(=f_n)$ is such that the terms largely cancel, leaving only a remainder of the order of the errors of individual terms. We must ensure that this is not so.

IV. EVALUATION OF THE SOLUTION

The problem has been formulated in terms of a complex energy E which represents the decay process. However, in the numerical calculations which we are about to outline, we ignore the imaginary part of E. This is justified, since the imaginary part is so small that it does not have any appreciable effect on the values of the functions considered in this section.

The first step in the evaluation of (3.17) is to obtain an explicit expression for $X_n(x)$. This function depends on κ_n, which is usually about 50. Thus we want the asymptotic formulae for large κ_n. These have been obtained in a number of papers. We shall use the particular result[5] that, when $l=0$,

$$X=(\cot\alpha)^{\frac{1}{2}}\exp(\omega), \quad (4.1)$$

$$\omega=\kappa(\alpha-\cos\alpha\sin\alpha), \quad (4.2)$$

and

$$\cos^2\alpha=x/\kappa. \quad (4.3)$$

Suffixes have been dropped. (The term of X in $\exp(-\omega)$ which appears inside the barrier is quite negligible for the calculation in this paper.) These equations were developed primarily for the region where $x<\kappa$. However, it can be seen that this restriction is not essential to their derivation, and the expression (4.1) equally represents the hypergeometric function for large x. This may also be checked directly. The exact definition is

$$X=\exp(-\tfrac{1}{2}\kappa\pi)(\kappa\pi)^{-\frac{1}{2}}\times\Gamma(1+\tfrac{1}{2}i\kappa)W_{-\frac{1}{2}i\kappa,\frac{1}{2}}(-2ix), \quad (4.4)$$

where W is Whittaker's function. (Compare Eq. (4.1) with Eqs. (3.18) and (3.2) of reference 5 and the definition of W.[6])

Using the known asymptotic expansion for W (see reference 6, p. 343), it can be checked that, for x tending to infinity and κ large, both (4.1) and (4.4) have the same expression, *viz.*,

$$\exp(ix-\tfrac{1}{2}i\kappa\log x+i\eta)\{1+0(1/x)\},$$

where

$$\eta=\eta_n=\tfrac{1}{4}\pi+\tfrac{1}{2}\kappa_n(\log\kappa_n-1-\log 4). \quad (4.5)$$

Thus when $l_n=0$, we shall use (4.1) for X_n.

When $l_n>0$ it is most convenient to employ a suitable recursion formula to estimate X_n.[7] For example, if $l_n=1$,

$$X_n=C(1/x+\tfrac{1}{2}\kappa_n-d/dx)(\cot\alpha_n)^{\frac{1}{2}}\exp(\omega_n) \doteq C(\tfrac{1}{2}\kappa_n+\tan\alpha_n)(\cot\alpha_n)^{\frac{1}{2}}\exp(\omega_n). \quad (4.6)$$

As $x\to\infty$, $\tan\alpha_n\to -i$. Hence, for the correct infinity behavior, we choose C so that

$$X_n=(1-2i/\kappa_n)^{-1}(1+2\tan\alpha_n/\kappa_n)\times(\cot\alpha_n)^{\frac{1}{2}}\exp(\omega_n). \quad (4.7)$$

Equation (4.6) cannot be used near $x=\kappa$, for there dX/dx is not equal to the derivative of the approximate expression for X, which is actually the first term of a semiconvergent series.[5] However, when x is of the order of κ, the centrifugal term is very small compared with $1-\kappa/x$ (except, of course, in the actual limit), and hence the solutions are almost independent of l_n in this region. Thus even when x is near κ, (4.7) gives X_n with only a slight error, since when α approaches zero, X_n behaves, aside from the constant factor, as if l_n were zero.

The correcting factor in (4.7) is never very different from 1, since, at the nuclear radius, $\tan\alpha_n/\kappa_n\sim 0.05$ and it decreases to zero at the boundary of the potential barrier; then it becomes imaginary and its magnitude changes from zero to about 0.02 as x goes to infinity. Thus, in calculating the integrals (3.17) and

[5] M. A. Preston, Phys. Rev. **71**, 865 (1947).

[6] E. J. Whittaker and G. N. Watson, *Modern Analysis* (Cambridge University Press, London, 1946), p. 340.

[7] L. Infeld, Phys. Rev. **59**, 743 (1941); also see reference 5, Eq. (5.2).

TABLE I. Values of I.

$r\times10^{13}$ cm	I_{n0}^-	I_{n0}^+	I_{0n}^-
8.27	$4.1+9.6i$	0.24	5.7
9.36	$3.4+10.6i$	0.26	6.0
10.0	$2.9+11.2i$	0.27	6.1
11.5	$1.6+12.4i$	0.31	6.2
16.0	$-2.9+15.5i$	0.38	6.3

(3.16), the effect of this term can be represented by a constant mean value which is not very different from one.

If l_n is not too much greater than one, considerations very similar to these will still apply.

Now

$$d/dx(\tfrac{1}{2}\exp(-2\omega))=\exp(-2\omega)\tan\alpha.$$

Therefore, we may define

$$g_n(x)=\tfrac{1}{2}\rho_n'\exp(-2\omega_n), \tag{4.8}$$

where ρ' is the mean value factor, almost unity, which we have just discussed. Then

$$g_n(\gamma_{nj}t)=\tfrac{1}{2}\rho_n'\exp\{-2\gamma_{jn}\kappa_j(\beta_{nj}-\cos\beta_{nj}\sin\beta_{nj})\} \tag{4.9}$$

$$=\tfrac{1}{2}\rho_n'\exp(-2\Omega_{nj}), \tag{4.10}$$

where

$$\cos\beta_{nj}=\gamma_{nj}\cos\alpha_j=\gamma_{nj}(t/\kappa_j)^{\frac{1}{2}}. \tag{4.11}$$

Substituting these explicit forms of g_n, X_n, X_j in the expression (3.17) for v_{nj}, we find

$$v_{nj}(k_jr)=v_{nj}(x)=\tfrac{1}{2}a_j\gamma_{jn}\rho_{nj}(\cot\alpha_0\cot\beta_0)^{\frac{1}{2}}\times\exp(\omega_0-\Omega_0)\sum_k B_k^{nj}\times\{I_{nj}^-(k)-I_{nj}^+(k)\}/x^{k+1}, \tag{4.12}$$

$$v_{jn}(k_nr)=\tfrac{1}{2}a_n\rho_{jn}(\cot\alpha_0\cot\beta_0)^{\frac{1}{2}}\times\exp(\Omega_0-\omega_0)\sum_k B_k^{nj*}\times\{I_{jn}^-(k)-I_{jn}^+(k)\}/x^{k+1}, \tag{4.13}$$

where

$$I_{nj}^+(p-1)=I_{jn}^+(p-1)=\int_{k_jr}^{\infty}\exp(\Omega-\Omega_0+\omega-\omega_0)(x/t)^p\times(\cot\alpha\cot\beta\tan\alpha_0\tan\beta_0)^{\frac{1}{2}}dt, \tag{4.14}$$

$$I_{nj}^-(p-1)=\int_{k_jr}^{\infty}\exp(-(\Omega-\Omega_0)+\omega-\omega_0)(x/t)^p\times(\cot\alpha\cot\beta\tan\alpha_0\tan\beta_0)^{\frac{1}{2}}dt, \tag{4.15}$$

$$I_{jn}^-(p-1)=\int_{k_jr}^{\infty}\exp((\Omega-\Omega_0)-(\omega-\omega_0))(x/t)^p\times(\cot\alpha\cot\beta\tan\alpha_0\tan\beta_0)^{\frac{1}{2}}dt. \tag{4.16}$$

Here we have written Ω, ω, β, α for Ω_{nj}, ω_j, β_{nj}, α_j and have denoted by subscript $_0$ the value of any quantity when $t=x=k_jr$. These integrals are to be found by numerical methods. If $E_n<E_j$, I_{jn}^- as it stands can be evaluated for a given value of k_jr; its integrand is a decreasing exponential. However, I_{nj}^- has an increasing exponential integrand inside the potential barrier, and in the external regions where it is oscillatory, the amplitude of the oscillations is of the same order of magnitude as the value of the integrand near the end of the barrier. However, the integral is convergent since, as $t\rightarrow\infty$, this amplitude goes to zero like t^{-p}. To evaluate I_{nj}^- numerically it is therefore necessary to transform it. This can be done by the substitution of the complex variable

$$u=2i\,\mathrm{arc}\cos\{\gamma_{nj}(t/\kappa_j)^{\frac{1}{2}}\}. \tag{4.17}$$

It can then be seen that in the u-plane the path of integration can be taken as the line parallel to the real axis from $u=i\beta_0$ to $u=\infty+i\beta_0$. On this path, both real and imaginary parts of the integrand have a rapidly decreasing exponential behavior. The numerical work is laborious, but the results are reliable.

To find the numerical value of I_{nj}^+ either the method for I_{jn}^- or that for I_{nj}^- can be used.

V. APPLICATION TO POLONIUM

As an example, we apply the considerations of the previous sections to the case of polonium, where Chang has reported twelve low energy radiations. We consider first the simplified case of a spectrum of two lines—those connected with the ground state and a typical excited state. For the latter, we have taken the line labeled α_9 by Chang. Thus the energies are $E_0=5.303$ and $E_n=E_9=4.111$ Mev. Then, in the notation of the previous sections, we have, ignoring the effect of all states but these two,

$$\begin{aligned} f_0 &= f_{00}(k_0r)+f_{0n}(k_0r), & f_{nn} &= a_n\mathrm{X}_n(k_nr), \\ f_n &= f_{n0}(k_nr)+f_{nn}(k_nr), & f_{n0} &= v_{n0}(k_0r)\mathrm{X}_n(k_nr), \\ f_{00} &= a_0\mathrm{X}_0(k_0r), & f_{0n} &= v_{0n}(k_nr)\mathrm{X}_0(k_0r). \end{aligned}$$

We shall study this case on the assumption that the transition is "dipole,"*** that is $p=2$ and $l_0=0$, $l_n=1$. Then $\mathrm{X}_0=(\cot\alpha)^{\frac{1}{2}}e^{\omega}$, where $\cos^2\alpha=k_0r/\kappa_0$, and $\omega=\kappa_0(\alpha-\cos\alpha\sin\alpha)$. Also $\mathrm{X}_n=\{\kappa_n+2\tan\beta)/(\kappa_n-2i)\}(\cot\beta)^{\frac{1}{2}}e^{\Omega}$, where $\cos^2\beta=k_nr/\kappa_n$, $\Omega=\kappa_n(\beta-\cos\beta\sin\beta)$. Then we have

$$v_{n0}(k_0r)=\tfrac{1}{2}a_0\gamma_{0n}\rho_{n0}(\cot\alpha_0\cot\beta_0)^{\frac{1}{2}}\exp(-(\Omega_0-\omega_0)) \times B_1^{n0}\{I_{n0}^-(1)-I_{n0}^+(1)\}/(k_0r)^2$$

and

$$v_{0n}(k_nr)=\tfrac{1}{2}a_n\rho_{0n}(\cot\alpha_0\cot\beta_0)^{\frac{1}{2}}\exp(\Omega_0-\omega_0) \times B_1^{n0*}\{I_{0n}^-(1)-I_{n0}^+(1)\}/(k_0r)^2.$$

The values of the integrals I have been calculated and are shown in Table I. The meaning factors ρ_{n0} and ρ_{0n} can be seen to lie between 0.95 and 1.05. We shall replace them by unity. We also write

$$B_1^{n0}=A_1^{n0}=(2m/\hbar^2)2^{\frac{1}{2}}Q_1^{n0}=(2^{5/2}me^2/3\hbar^2)R, \quad (5.1)$$

where R is of the same order as, e.g.,

$$C_{10}^{n0}=\int u_n^*\Sigma r_i\cos\theta_i u_0 d\xi.$$

We then find

$$\begin{aligned} f_0(r_0) &= \mathrm{X}_0(a_0+a_n\exp(i\delta)R^*B), \\ f_n(r_0) &= \mathrm{X}_n(a_n\exp(i\delta)+a_0RD), \end{aligned} \quad (5.2)$$

where r_0 is the nuclear radius and a_0, a_n are real. The values of the known quantities in Eqs. (5.2) are shown in Table II. Note that B and D, which represent the effect of the electrostatic interaction, are not sensitive to the value of the nuclear radius. Since the effect is an extra-nuclear one, occurring over distances large compared with r_0, this is a satisfactory result. We may also note the justification of the perturbation methods. If the perturbation calculation is justified, it is necessary that $|RD\mathrm{X}_n|\ll\mathrm{X}_0$ and $|R^*B\mathrm{X}_0|\ll\mathrm{X}_n$. By its definition (5.1) $|R|$ is certainly less than 10^{-12}, the greatest possible distance of a proton from the center of the nucleus; hence these inequalities hold.

TABLE II. Parameters in Eqs. (5.2).

$r_0\times10^{13}$ cm	χ_0	χ_n	B	D
8.27	6.0×10^{13}	5.1×10^{17}	4.1×10^{14}	$(0.4+1.0i)10^7$
9.36	6.6×10^{12}	5.1×10^{16}	3.4×10^{14}	$(0.3+1.0i)10^7$
10.0	2.0×10^{12}	1.5×10^{16}	3.0×10^{14}	$(0.2+1.0i)10^7$

Next, let us consider the effect of assuming for a_0 and a_n the values given by Chang.[1] Since the squares of these quantities are proportional to the respective partial decay constants, we have $a_n/a_0\doteq7\times10^{-3}$. ($\lambda_9=2.8\times10^{-12}$, $\lambda_0=5.9\times10^{-8}$ sec.$^{-1}$.) Now $|DR|<10^{-5}$; thus $|a_0DR|\ll a_n$. Therefore $f_n=a_n\exp(i\delta)\mathrm{X}_n$. Also $|R^*Ba_n|$ is at most of the order of a_0, but since 10^{-12} is probably a high value for $|R|$, the term $a_n\exp(i\delta)R^*B$ does not alter f_0 very much. Hence $|f_n/f_0|\doteq a_n\mathrm{X}_n/a_0\mathrm{X}_0\doteq10^2$. This means that the probability of decay with energy E_n is much greater (10^2) than with energy E_0, *in the absence of the barrier*. At first sight, this might appear to be the explanation of Chang's results; *viz.*, the emission of an α-particle with energy E_0 is a forbidden process as compared to the emission with energy E_n, but a transition from E_n to E_0 takes place outside the nucleus. However, we shall see that these assumptions are in contradiction with the observed absolute intensity of the α-particles.

By the conservation theorem, we have, for the general case of N states,

$$\frac{\partial}{\partial t}\int\sum_0^{N-1}\phi_m\phi_m^*dv=-\frac{\hbar}{2im}\int\sum(\phi_m^*\,\mathrm{grad}\phi_m-\phi_m\,\mathrm{grad}\phi_m^*)\cdot\mathbf{n}d\sigma,$$

where the integrations are over the volume and surface of a sphere of large radius and ϕ_m is defined by Eq. (2.4). Now the energy is complex so that $\sum\phi_m\phi_m^*$ contains a factor $\exp(-\lambda t)$ and the rate of charge of the volume integral is $-\lambda\int\sum\phi_m\phi_m^*dv$. This integral can be split into two parts, one over the region inside the nucleus and one over the region outside, i.e.,

$$\int\sum\phi_m\phi_m^*dv=\sum_m(I_i^m+I_0^m)=\sum I_i^m+\int_{r_0}^{\infty}\sum|f_m|^2dr.$$

*** In γ-ray transition dipole and quadrupole effects are of approximately equal importance; however, in the long-range effects considered here we may expect the dipole to predominate.

However, $|\phi_m|$ decreases very rapidly outside the nucleus, so $\sum I_0^m$ can be neglected. For the surface integral, since the sphere is large, we can use the asymptotic expression for ϕ_m. The result is

$$\lambda = \sum_m v_m |a_m|^2 / \sum_m I_i^m. \tag{5.3}$$

In the case we have been considering where only two levels are concerned,

$$\lambda = v_0 a_0^2/(I_i^0 + I_i^n), \quad \text{since} \quad v_n a_n^2 \ll v_0 a_0^2.$$

We must now consider the integrals I_i^0 and I_i^n which involve the behavior of the wave functions inside the nucleus. It is reasonable to make these functions and their derivatives continuous at the nuclear boundary. We have seen that with Chang's intensities, $|f_n/f_0| \sim 10^2$, and we may also note that f_n'/f_n and f_0'/f_0 are of comparable magnitude. Thus whatever the explicit internal expression of the wave functions, it is clear that $I_i^n \gg I_i^0$. Hence, with Chang's relative intensities

$$\lambda = v_0 a_0^2 / I_i^n = (v_0 a_0^2 / v_n a_n^2)(v_n / \mathcal{I}_i^n), \tag{5.4}$$

when $\mathcal{I}_i^n$ is the internal integral in the standard theory without interaction. (The function f_n differs from X_n, which corresponds to it in the standard theory of radioactivity, only by the factor a_n.) But since $a_0 \gg a_n$, the partial decay constant $\lambda_n \doteq (v_n a_n^2 / v_0 a_0^2)\lambda$. Hence $\lambda_n = v_n / \mathcal{I}_i^n = \lambda_n^0$, where λ_n^0 is the partial decay constant on the standard theory. Thus, when interaction is allowed for, the partial decay constant is practically unchanged from that expected on the standard theory.

In other words, although we may assume suitable conditions to obtain the *relative* intensities found by Chang, the total decay constant, which is determined from the partial one by the ratio $v_0 a_0^2 / v_n a_n^2$, is much less than the experimental value. For example, if we take the nuclear radius 8.3×10^{-13} cm (which on the standard one-body theory fits the experimental data for the intense main line[5]), we find that if $a_n/a_0 \doteq 7 \times 10^{-3}$, then $\lambda \doteq 5 \times 10^{-14}$ sec.$^{-1}$, whereas the experimental value is 6×10^{-8} sec.$^{-1}$. Of course with a larger radius we can increase the value of λ. Taking $r_0 = 11.5 \times 10^{-13}$ cm, we find $\lambda \doteq 1.5 \times 10^{-9}$ sec.$^{-1}$ and with $r_0 \doteq 12.5 \times 10^{-13}$ we find that λ is about the observed value. However, the nuclear radius as a parameter in the one-body model is always less than 10^{-12} cm and, using the law $r_0 = R_0 A^{\frac{1}{3}}$, we would expect about 9×10^{-13} cm for polonium. A radius such as 12.5×10^{-13} cm would imply a most unusual extension of the region where nuclear forces hold.

The above arguments are not essentially altered if we allow for the emission of particles in more than one low energy state which can interact with the nucleus and acquire the energy of the main line. Equations (5.2) are then replaced by

$$f_0/\chi_0 = a_0 + \sum_1^{N-1} a_m e^{i\delta_m} R_m^* B_m,$$

$$f_m/\chi_m = a_0 R_m D_m + a_m e^{i\delta_m}, \quad m = 1, 2, \cdots (N-1).$$

Now as the difference between E_0 and E_m decreases, D_m increases and so presumably does R_m. However, for m great enough, we have as before $|a_0 D_m R_m| \ll a_m$, if we use Chang's intensities. Also B_m decreases with $E_0 - E_m$; hence $|a_m R_m^* B_m|$ remains of the same order as a_0 or less for all m. Thus it still follows that $|f_m/f_0| \gg 1$, at least for m large enough. Thus we can still reduce (5.3) to an equation similar to (5.4), *viz.*,

$$\lambda \doteq v_0 a_0^2 / \sum_M^{N-1} a_m^2 \mathcal{I}_i^m,$$

where M is the lowest value of m for which $|f_m/f_0| \gg 1$ is valid. That is

$$\lambda \doteq v_0 a_0^2 \Bigg/ \left(\sum_M^{N-1} a_m^2 v_m / \lambda_m^0 \right) < (v_0 a_0^2 / v_n a_n^2) \lambda_n^0,$$

where n is some value of $m > M$. Thus, just as before, λ is much less than the experimental value, unless a large radius is taken.

To summarize, we may say that Chang's results can be explained with our model only on the basis of two assumptions: that the direct transition to the ground state is (more or less) forbidden and electrostatic interactions occur after the α-particle has left the nuclear region and that the radius of this region must be much larger ($\sim$25 percent) than it is in the case of other radioactive elements. This latter assumption would involve us in many new difficulties. It is to be noted that any explanation involving strictly nuclear behavior does not affect this point; it could only alter the ratio (f_0/f_n) at r_0. Coupled with the inability of Zajac, Broda, and Feather[4] to find γ-rays which would confirm Chang's results, these theoretical arguments would suggest that the source of the lines observed by Chang was not in the polonium nucleus.

VI. GENERAL DISCUSSION

We may consider what the effect of the electrostatic interaction will be in a general case. Considering again for simplicity the case of only one level in addition to the ground state, Eqs. (5.2) can be solved for a_0 and a_n. The result is

$$\begin{aligned} a_0 &= f_0/X_0 - R^*Bf_n/X_n, \\ a_n e^{i\delta} &= f_n/X_n - DRf_0/X_0. \end{aligned} \tag{6.1}$$

In general the quantities $|f_0|$ and $|f_n|$, which as indicated in Section III determine the probabilities of decay in the absence of the potential barrier, will be of the same order since $E_0 - E_n$ is small compared with the total energy of the escaping particle. In (6.1) the terms f_0/X_0 and f_n/X_n represent the effect of the barrier in the standard theory and the other terms R^*Bf_n/X_n and RDf_0/X_0 introduce the corrections due to the interaction after emission. If we assume $|f_0| \doteq |f_n|$ we see that the interaction alters a_0 by a fractional amount $|R^*BX_0/X_n|$ and alters a_n by the fraction $|RDX_n/X_0|$. To form some idea of the magnitude of these changes we may consider the case of polonium for which we have worked out numerical values. Taking $R = 10^{-12}$, we shall obtain upper limits for the effect. Then $|R^*BX_0/X_n| < 10^{-2}$ and $|RDX_n/X_0| < 10^{-1}$. Since the decay constants involve a_n^2, these correspond to changes of certainly less than 2 percent in λ_0 and 20 percent in λ_n. Thus the upper limit of the effect is a 20 percent change in the decay constant of the state of lower α-particle energy. When it is remembered that the dependence of λ on energy is logarithmic, it will be realized that even this upper limit does not give a very large effect. The above calculations are for $E_0 - E_n = 1.2$ Mev.

As $E_0 - E_n$ decreases, the effect of the interaction becomes more pronounced. In fact as this difference gets small it can be confirmed that the ratio $|R^*BX_0/X_n|$ becomes greater than one (although $|DRX_n/X_0|$ probably decreases slightly). However, when this happens we have clearly violated a condition for the validity of the perturbation assumptions implied in the use of Eq. (3.5). Nevertheless, we are led to suspect that when the energy difference is of the order of 500 kev or less the interaction with the nucleus after emission may appreciably affect the relative intensities of the two α-particle groups. As we have indicated above, it is this region of energy differences to which Dancoff[3] has paid particular attention, and his results also indicate that the effect is probably fairly large. In some of the elements which have a fine structure in the α-spectrum, the lower energy α-particles do not obey the Geiger-Nuttall law very exactly. This has been attributed to a non-zero value of the angular momentum quantum number l. We now see that if the energy levels are separated by less than about 500 kev (e.g., RaC) the partial decay constants may be altered by the electrostatic interaction also. It is not easy to separate these two causes, since there are at present few independent estimates of either l or R, but they may have effects of roughly the same order of magnitude, changing λ_n by a factor of about 2 or 3.

We may also consider briefly the effect when the ground state transition is accidentally more or less forbidden, i.e., $|f_0| \ll |f_n|$. Then (6.1) becomes $a_n \exp(i\delta) = f_n/X_n$ and $a_0 = f_0/X_0 - R^* \times Ba_n \exp(i\delta)$. Thus, if $|f_0|$ is very small a line may appear with intensity $|R^*Ba_n|^2$, which would be less than that expected for energy E_0. On the other hand, if $|f_0|$ is somewhat larger, there may be destructive interference making a_0 vanish, and what would have been a line with abnormally low intensity disappears entirely. There does not appear to be any example of either of these processes in the present experimental data.

It is a pleasure to record my appreciation of many stimulating discussions with Professor R. E. Peierls which have been of the greatest help in this investigation.