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THEORY OF ALPHA DECAY OF SPHEROIDAL NUCLEI

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December 10, 1953

Theory of Alpha Decay of Spheroidal Nuclei

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ABSTRACT

Various effects of spheroidal nuclear distortion on the alpha decay process are considered theoretically.

Differential equations governing alpha decay in the region beyond the maximum nuclear radius are derived. They consist of ordinary radial Schrödinger equations for alpha decay to various nuclear states with the addition of quadrupole interaction terms coupling the various equations.

The significance of wave amplitudes of various angular momentum alpha groups as Fourier components of the total wave function is pointed out, and experimental alpha decay rate data for even-even nuclei are discussed in these terms.

barrier. An important fact to realize, though, is that if such "directed" alpha emission takes place, it necessarily implies the presence of a mixture of different angular momentum waves of alpha emission. Furthermore, the relative amounts of various angular momentum components may change with radial distance through the coupling influence of the intrinsic electric quadrupole moment of the daughter nucleus. The influence of such noncentral electric interactions between the nucleus and alpha particle has been treated theoretically by Preston, and the treatment of coupling in the present paper will be seen to parallel his in many important respects.

Even-even nuclides of the heaviest elements ($\mathbb{Z}\geq 88$) are observed generally to decay by alpha emission to ground and excited states that are interpreted by the unified nuclear model as members of a rotational band sequence with even parities, spins of 0, 2, 4,...and energies $(\mathbb{A}^2/2\mathcal{X})\mathbf{I}(\mathbf{I}+1)$. $\{(\mathbb{A}^2/2\mathcal{X})=$ the rotational quantum energy. The different angular momentum states of the outgoing alpha particle wave necessary for a description in terms of directed emission will involve the above states of the nucleus.

In order to gain from experimental alpha decay rate data a real understanding of the fundamental nature of the process it is necessary first to solve the well-defined quantum mechanical problem of the outgoing waves in the region beyond the range of short range nuclear forces. The separate application of simple barrier penetration formulas to the various alpha groups may give misleading information as to the true magnitudes of the wave functions near the nuclear radius. Preston has stated the problem clearly. One might assume reasonable values for the various alpha

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INTRODUCTION

The dependence of alpha decay rates of even-even nuclides upon decay energy and atomic number has been remarkably well explained by coulombic barrier penetration treatments. Some of these treatments have differed widely in their specific assumptions about the "lifetime in the absence of a barrier," but by the choice of somewhat different values for the nuclear radius in the various cases good agreement with experimental decay rates of even-even nuclides are secured.*

The nucleus is generally assumed to have a spherically symmetric distribution of charge. The nuclear model of A. Bohr and B. R. Mottelson, however, attributes large spheroidal distortions to nuclei lying in regions much removed from closed neutron or proton shells. Such distortions can be present for even-even nuclei and give rise to an intrinsic electric quadrupole moment, $Q_{\rm o}$. Reference 2 will be referred to as A hereafter.

One effect of introducing spheroidal distortion is to bring some nuclear matter out to greater radial distances. Hill and Wheeler³ have suggested that the above effect might lead to increased alpha decay rates by virtue of a thinner potential barrier, and they estimate this effect by calculating the one-dimensional penetration factor through the thinnest part of the

^{*}The nuclides Po210, Po208, and Em212 are exceptions, decaying more slowly by factors of 5-20. They have 126 or less neutrons.

groups near the nucleus, integrate the wave equation outwards to large distances, and compare the magnitudes of the various outgoing waves with the experimental intensities. Preston states that small errors in the initial boundary conditions or in the integration might lead to the spurious appearance of considerable amounts of incoming waves at infinity and thus invalidate the results. Preston chooses, rather, to let the experimental intensities of the N alpha groups provide N restrictive boundary conditions on the waves at large distances and integrates inward toward the nucleus. The phases of the N waves must be chosen so as to satisfy boundary conditions at the nuclear surface.

FORMULATION OF THE EQUATIONS GOVERNING THE EXTERNAL SOLUTION

The use of nuclear wave functions from the strong-coupling approximation of the unified nuclear model permits the alpha particle-recoil nucleus system to be treated as a simple mechanical system and permits a great specialization of the generalized system of alpha decay radial equations of Preston. The strong coupling wave functions for the ground state rotational band in even-even nuclei reduce simply to the rotational wave functions of a symmetric top (A, p. 20), with quantum numbers I, M, and K. I is the total angular momentum quantum number, M, its projection along an axis fixed in space, and K, its projection along the nuclear symmetry axis. For all levels of the ground state band the quantum number K is zero; hence, the wave functions reduce to a particularly simple form, normalized spherical harmonics of even order. 9

$$\psi_{\text{nuc}} = Y_{\text{I}}^{\gamma}(\theta^{\dagger}, \varphi^{\dagger}), \qquad (1)$$

where θ ' and φ ' denote the direction of the nuclear symmetry axis with respect to a spherical polar coordinate system fixed in space. Since the nucleus is assumed to have axial symmetry, the two moments of inertia perpendicular to the symmetry axis will be equal and will be designated \Im . The only part of the strictly nuclear Hamiltonian which concerns this alpha decay formulation is that corresponding to the kinetic energy of the symmetric top with K=0, namely,

$$H_{\text{nuc}} = -\frac{\pi^2}{2^{3}} \left[\frac{\partial}{\partial \theta'} \left(\sin \theta' \frac{\partial}{\partial \theta'} \right) + \frac{1}{\sin^2 \theta'} \frac{\partial^2}{\partial \phi'^2} \right]$$
(2)

The eigenfunctions are those given by (1) with eigenvalues

$$E_{I} = \frac{n^{2}}{23} I(I+1).$$
 (3)

The complete Hamiltonian for the alpha decay problem will also include the usual kinetic and coulombic potential energy terms (center of mass system),

$$H_{\alpha} = -\frac{\hbar^{2}}{2mr^{2}} \left[\frac{\partial}{\partial r} \left(r^{2} \frac{\partial}{\partial r} \right) + \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial}{\partial \theta} \right) + \frac{1}{\sin^{2} \theta} \frac{\partial^{2}}{\partial \phi^{2}} \right] + \frac{zZe^{2}}{r}$$
 (4)

where r, θ , and φ are the relative coordinates of alpha particle and recoil nucleus with respect to the coordinate system fixed in space; e is the electrostatic unit charge; z, the charge of the alpha particle, i.e., 2; Z the charge of the daughter nucleus, and m the reduced mass.

Finally, an interaction term must be included in the Hamiltonian to account for the electric quadrupole interaction.

$$H_{int} = \frac{zQe^2}{2r^3} P_2(\cos \omega)$$
 (5)

where ω is the angle between the nuclear symmetry axis (θ', φ') and the relative position vector (θ, φ) of the alpha particle-recoil nucleus system. $P_2(\cos \omega)$ is the Legendre polynomial of second order. Q is the intrinsic electric quadrupole moment of the daughter nucleus given in A, Equation V.3 for a uniformly charged spheroidal nucleus as

$$Q = \frac{3}{\sqrt{5\pi}} ZR_0^2 \alpha_0 . \tag{6}$$

The nuclear surface to second order in $\alpha_{_{\mbox{O}}}$ is given by

$$R(\eta) = R_0[1 + \alpha_0 P_2(\cos \eta) - \frac{\alpha_0^2}{5}]$$

with η the angle measured from the nuclear symmetry axis.

Thus, the problem is to find the solution of the Schrödinger equation

$$(H_{\text{nuc}} + H_{\alpha} + H_{\text{int}})\psi = E\psi$$
 (7)

that asymptotically approaches at large r the outgoing waves of the various alpha groups in their proper relative intensities and that behaves properly as outgoing waves at the nuclear surface.

Let us consider the equation without the quadrupole interaction first.

$$(H_{\text{nuc}} + H_{\alpha})\psi = E\psi$$

The variables can be separated in the usual manner by substitution of

$$\psi = R(r)S(\theta, \varphi)P(\theta', \varphi').$$

Thus,
$$E = -\frac{\pi^{2}}{2\sqrt{3}P} \left[\frac{\partial}{\partial \theta'} \left(\sin \theta' \frac{\partial}{\partial \theta'} \right) + \frac{1}{\sin^{2}\theta'} \frac{\partial^{2}}{\partial \phi'^{2}} \right] P$$

$$-\frac{\pi^{2}}{2mr^{2}} \frac{1}{S} \left[\frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial}{\partial \theta} \right) + \frac{\partial}{\sin^{2}\theta} \frac{\partial^{2}}{\partial \phi^{2}} \right] S$$

$$-\frac{\pi^{2}}{2mr^{2}} \frac{1}{R} \left[\frac{\partial}{\partial r} \left(r^{2} \frac{\partial}{\partial r} \right) \right] R + \frac{zZe^{2}}{r}$$

The angular eigenfunctions S and P are just spherical surface harmonics, and on substitution of

$$S(\theta, \varphi) = Y^{\mu}_{\ell}(\theta, \varphi)$$

$$P(\theta', \varphi') = Y^{\nu}_{T}(\theta', \varphi')$$

one obtains the radial equation

$$E - \frac{zZe^2}{r} = + \frac{\hbar^2}{23}I(I+1) + \frac{\hbar^2}{2mr^2} \ell(\ell+1) - \frac{\hbar^2}{2mr^2} \frac{1}{R} \left[\frac{\partial}{\partial r} (r^2 \frac{\partial}{\partial r}) \right] R .$$

Any product or linear combination of products of surface harmonics $Y_\ell^\mu(\theta,\varphi)Y_I^\nu(\theta',\varphi') \text{ is a satisfactory eigenfunction of the angular variables.}$ However, in the physical problem restrictions are imposed, since the total angular momentum L and its component M along a fixed axis in space must be conserved (i.e., be the same as in the initial nucleus).

Thus, only particular linear combinations of the products $Y_{\ell}^{\mu}(\theta,\phi)Y_{I}^{\nu}(\theta',\phi') \text{ are allowed. The desired linear combinations are discussed by Blatt and Weisskopf }^{10} \text{ and designated } \mathcal{L}_{\ell}^{M}.$ These functions are related to the surface harmonics in the following way:

$$Y_{L\ell I}^{M} = \sum_{\mu} \sum_{\nu} C_{\ell I}(LM; \mu\nu) Y_{\ell}^{\mu}(\theta, \varphi) Y_{I}^{\nu}(\theta', \varphi'),$$

where the $C_{L}(LM;\mu\nu)$ are Clebsch-Gordan or vector addition coefficients in the notation of Blatt and Weisskopf. The summation is actually a single summation, since the Clebsch-Gordan coefficients vanish unless $\mu + \nu = M$. Thus defined, the Y functions are orthonormal.

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$$\int \int_{L_1 L_1 I_1}^{M_1 *} \int_{L_2 L_2 I_2}^{M_2} \sin \theta d\theta d\phi \sin \theta' d\theta' d\phi'$$

$$= \delta_{L_1 L_2} \delta_{M_1 M_2} \delta_{L_1 L_2} \delta_{I_1 I_2}$$

In this paper only even-even nuclei are considered, so that L=M=0. The $\mathcal U$ functions take the simple form

$$\mathcal{J} \stackrel{\text{O}}{=} \frac{1}{(2j+1)^{1/2}} \sum_{-j \le \mu \le j} Y_{j}^{\mu}(\theta, \varphi) Y_{j}^{-\mu}(\theta', \varphi') \tag{8}$$

Let the solution to the complete equation (7), be expressed by a series expansion in \mathcal{G}° , where the coefficients μ , j are functions of r.

$$\psi = \sum_{j=0}^{\infty} \psi j(r) \mathcal{J}_{ojj}^{o}(\theta, \varphi, \theta', \varphi') \qquad (9)$$

$$j = 0$$

$$j \text{ even}$$

Substituting (9) into (7), multiplying by \mathcal{J}_{oLL}^{o*} (complex conjugate of \mathcal{J}_{oLL}^{o}) and integrating over all angular space one obtains a set of ordinary oLL second order linear differential equations in the μ_L 's, coupled by the quadrupole interaction.

$$-\frac{\pi^{2}}{2mr^{2}}\frac{\partial}{\partial r}\left(r^{2}\frac{\partial}{\partial r}\right)\mu_{L} + \left[\left(\frac{\pi^{2}}{23} + \frac{\pi^{2}}{2mr^{2}}\right)L(L+1) + \frac{2Ze^{2}}{r} - F\right]\mu_{L}$$

$$= -\frac{Qe^{2}}{r^{3}}\sum_{\substack{j=0\\j \text{ even}}}^{\infty} \mu_{j}\int_{\text{oLL}}^{\sqrt{o^{*}}} P_{2}(\cos \omega) \mathcal{J}_{\text{ojj}}^{o} d\Omega d\Omega'. \qquad (10)$$

Making the substitution $u_L = W_L/r$ and rearranging one obtains,

$$\frac{d^{2}w_{L}}{dr^{2}} - \left[\frac{2m}{\pi^{2}} \left(\frac{2Ze^{2}}{r} - E\right) + \left(\frac{m}{3} + \frac{1}{r^{2}}\right)L(L+1)\right] w_{L}$$

$$= \frac{2mQe^{2}}{\pi^{2}r^{3}} \sum_{\substack{j=0\\ j \text{ even}}} w_{j} \int \mathcal{Y}_{oLL}^{\delta} P_{2}(\cos \omega) \mathcal{Y}_{ojj}^{\circ} d\Omega d\Omega'.$$
(11)

The integrals on the right hand side of (11) are equal to the Racah coefficients 11 (LLOO $|P_2(\cos\omega)|$ jj00). They vanish here if L and j differ by more than two units. The case L = 0, j = 0 also vanishes. From the general formula of Racah it may be deduced that the nonvanishing coefficients in (11) are given as follows:

$$(\mu\mu\phi\phi)|P_{2}(\cos\omega)|\mu\mu\phi\phi) = \frac{\mu(\mu+1)}{(2\mu+3)(2\mu-1)}$$
(12a)

$$(\nu\nu00|P_{2}(\cos \omega)|\nu+2,\nu+2,00) = \frac{3}{2} \frac{(\nu+2)(\nu+1)}{(2\nu+3)(2\nu+5)^{1/2}(2\nu+1)^{1/2}}$$

The explicit equations for the first four waves are

$$w_0'' - \left[\frac{4mZe^2}{\hbar^2r} - \frac{2mE}{\hbar^2}\right] w_0 = \frac{2mQe^2}{\hbar^2r^3} \left[\frac{w_2}{\sqrt{5}}\right]$$
 (13a)

$$w_{2} = \frac{\left[\frac{4mZe^{2}}{\pi^{2}r} - \frac{2mE}{\pi^{2}} + 6\left(\frac{m}{\sqrt{r}} + \frac{1}{r^{2}}\right)\right]}{\pi^{2}} w_{2} = \frac{2mQe^{2}}{\pi^{2}r^{3}} \left[\frac{1}{\sqrt{5}}w_{0} + \frac{2}{7}w_{2} + \frac{6}{7\sqrt{5}}w_{4}\right]$$
(13b)

$$w_{4}" - \left[\frac{4mZe^{2}}{\hbar^{2}r} - \frac{2mE}{\hbar^{2}} + 20\left(\frac{m}{3} + \frac{1}{r^{2}}\right)\right]w_{4} = \frac{2MQe^{2}}{\hbar^{2}r^{3}}\left[\frac{6}{7\sqrt{5}}w_{2} + \frac{20}{77}w_{4} + \frac{15}{11\sqrt{13}}w_{6}\right] (13c)$$

$$w_{6}" - \left[\frac{4mZe^{2}}{\hbar^{2}r} - \frac{2mE}{\hbar^{2}} + 42\left(\frac{m}{3} + \frac{1}{r^{2}}\right)\right]w_{6} = \frac{2mQe^{2}}{\hbar^{2}r^{3}} \left[\frac{15}{11\sqrt{13}}w_{4} + \frac{14}{55}w_{6}\right]$$
(13d)

RELATION OF WAVE AMPLITUDES TO A CORRELATION FUNCTION INVOLVING THE ANGULAR VARIABLES

If one were to plot a correlation probability function between the nuclear orientation and the angular position of the alpha particle at some particular distance just beyond the distance of maximum nuclear radius, one would surely expect to find a correlation favoring location of the alpha particle at those angles where it is nearest to the spheroidal nuclear surface. For prolate distortion the correlation function in angle ω (cf. equation 5) should be a maximum at $\omega = 0$ and π . For oblate distortion the correlation should favor $\omega = \pi/2$.

Leaving aside for the moment the estimation of the precise form of the correlation function, let us consider the relation between the correlation probability function and the amplitudes of the various alpha waves. Letting the correlation probability function be $|f(\cos \omega)|^2$, there is defined an angular wave function $f(\cos \omega)$ involving the four angular variables θ , φ , θ^i , φ^i . This function can be expanded in the orthonormal set of functions $\mathcal{G}_{0,j}^0$ discussed earlier. The symmetry of $f(\cos \omega)$ about $\omega = \pi/2$ insures that the odd j terms vanish in the expansion.

Let
$$f(\cos \omega) = \sum_{\substack{j=0\\j \text{ even}}} b_j \mathcal{V}_{0jj}^{0}. \qquad (14)$$

The coefficients b_k are determined in the usual manner by multiplying both sides of the equation by y_{okk}^{\star} and integrating over all angular space.

$$b_{k} = \int \mathcal{Y}^{\delta}_{okk} f(\cos \omega) d\Omega d\Omega^{\dagger} . \qquad (15)$$

To evaluate the coefficients expand $f(\cos \omega)$ in a series of Legendre polynomials in $\cos \omega$.

$$f(\cos \omega) = \sum_{i=0}^{\infty} a_i P_i(\cos \omega)$$

$$i = 0$$

$$i \text{ even}$$
(16)

and

$$a_{n} = \frac{2n+1}{2} \int_{0}^{\pi} f(\cos \omega) P_{n}(\cos \omega) \sin \omega d\omega \qquad (17)$$

Substituting (16) into (15)

$$b_{k} = \sum_{i=0}^{\infty} a_{i} \int y_{0kk}^{*} P_{i}(\cos w) dy dy.$$

$$i = 0$$

$$i = \text{even}$$
(18)

Since $\int_{000}^{0} = 1/4\pi$, (18) is equivalent to the following:

$$b_{k} = 4\pi \sum_{i=0}^{\infty} a_{i} \int y_{okk}^{\circ} P_{i}(\cos \omega) y_{ooo}^{\circ} d\Omega d\Omega'$$

$$i = 0$$

$$i = even$$

The integrals are now simply equal to Racah coefficients, and all terms in the summation vanish except i = k.

$$b_k = 4\pi a_k (kk00 | P_k (\cos \omega) | 00 00)$$
 (19)

These particular coefficients can be shown from Racah's general formula 11 to be equal to $(2k \pm 1)^{-1/2}$. Hence the expansion coefficients b_k are related uniquely to the coefficients a_k by the simple expression

$$b_k = 4\pi/(2k + 1)^{-1/2} a_k$$
 (20)

Combining (20) and (17),

$$b_{k} = 2\pi (2k + 1)^{1/2} \int_{0}^{\pi} f(\cos \omega) P_{k}(\cos \omega) \sin \omega d\omega \qquad (21)$$

A TRANSFORMATION OF THE ALPHA DECAY PROBLEM TO A SPHERICAL POLAR COORDINATE SYSTEM FIXED IN THE NUCLEUS

Solutions of the radial equations (13) may be found for all space where the radial distance exceeds the maximum radial excursion of nuclear matter (or more strictly speaking, the maximum distance at which the short range nuclear forces on the alpha particle are of importance). A serious problem seems to be presented in bringing the solutions in to all parts of the nuclear surface for the imposition of boundary conditions, since the boundary is defined not by r alone, but depends on all five variables in the problem.

An interesting possibility is suggested by the unique dependence of the expansion coefficients a_k and b_k of the previous section on one another, as given by (20). One might formulate the alpha decay problem in two variables r and ω such that the problem is completely equivalent to the five variable problem given by (7).

In this alternative formulation one relates the angular functions by equating (14) and (16).

$$\sum_{j=0}^{\infty} b_{j} \mathcal{J}_{0jj}^{0} = f(\cos \omega) = \sum_{j=0}^{\infty} a_{j} P_{j}(\cos \omega) . \qquad (22)$$

Substituting from (20) gives

$$\sum_{j=0}^{\infty} 4\pi (2j+1)^{-1/2} a_{j} \int_{0}^{0} = \sum_{i=0}^{\infty} a_{i} P_{i}(\cos \omega) . \qquad (23)$$

Since (23) is true for any values a_j , the following must be identities:

$$\mathcal{L}_{\text{ojj}}^{\circ} = \frac{1}{(8\pi^{2})^{1/2}} \cdot \left(\frac{2j+1}{2}\right)^{1/2} P_{j}(\cos \omega) = \frac{1}{(8\pi^{2})^{1/2}} S_{j}(\mu) \tag{24}$$

$$\begin{split} &S_{j}(\mu) = \left(\frac{2j+1}{2}\right)^{1/2} P_{j}(\mu) \text{ are orthonormal functions on the interval -1 to +1,} \\ &\text{as } \int_{-1}^{1} S_{j} S_{k} d\mu = \delta_{jk} \text{ , where } \mu = \cos \omega. \end{split}$$

These identities can also readily be derived from the addition theorem of spherical harmonics 12 and (8). The terms in the Hamiltonian of (7) which represent rotational kinetic energy must be replaced by suitable operators on ω that will give the same angular eigenvalues, i.e.,

$$(\hbar^2/2 \mathcal{F} + h^2/2mr)L(L + 1)$$
.

The operator

$$H_{\omega} = -(\hbar^2/2 \Re + \hbar^2/2 mr^2) \frac{\partial}{\partial \omega} (sin \omega \frac{\partial}{\partial \omega})$$

satisfies this requirement with $(8\pi^2)^{-1/2} S_L(\mu)$ the corresponding eigenfunctions. The operator H_{ω} is interesting in that it represents the kinetic energy operator of a symmetric top (K=0) with what might be called a "reduced moment of inertia" equal to the product of \mathcal{T} and mr^2 divided by their sum. The rest of the Hamiltonian in (7) may be left unchanged. The four angular coordinates of the original formulation have been transformed

to a system of four coordinates, one of which is ω , the angle between the θ, φ and θ', φ' directions, and the other three of which might be the three Eulerian angles specifying the spatial orientation of the system as a whole. (The system as a whole will in general be an asymmetric top with two of the moments of inertia functions of ω .) (This transformation to relative rotational coordinates is expected to yield such simplification only when total angular momentum is zero.)

The derivation of radial equations from the two dimensional Schrödinger equation proceeds analogously to that of the five dimensional equation and leads to the same radial equations (13).

In the two dimensional problem the integrals appearing in equations (10) and (11) may be simplified by substitution of the identity (25) and transformation of the differential volume element to the "relative and total" rotational coordinate system. The integrand is a function only of the relative rotational coordinate ω and is independent of the Eulerian angles pertaining to rotation as a whole. Thus, integration over the three Eulerian angles just gives a constant $8\pi^2$, which divides out with the normalization factor in (24). Thus,

$$I_{L_{\hat{\mathbf{j}}}} = \int \mathcal{Y}_{\text{oLL}}^{\text{o*}} P_{2}(\cos \omega) \mathcal{Y}_{\text{ojj}}^{\text{o}} d\Omega d\Omega' = \int_{0}^{\pi} S_{L}(\cos \omega) P_{2}(\cos \omega) S_{j}(\cos \omega) \sin \omega d\omega$$

$$= \frac{(2L+1)^{1/2}(2j+1)^{1/2}}{2} \int_{0}^{\pi} P_{L}(\cos \omega) P_{2}(\cos \omega) P_{j}(\cos \omega) \sin \omega d\omega . \qquad (25)$$

These integrals over triple products of Legendre polynomials have been evaluated numerically for specific cases by Shortley and Fried¹³ and a general formula given by Racah. Formulas (12) of this paper have been cross checked with Racah's above-mentioned formula. The integrals are also simply related to Clebsch-Gordan coefficients (by Racah's equation (50') and Blatt and Weisskopfs' equation (5.8) p. 791 as follows:

$$I_{Lj} = \frac{(2L+1)^{1/2}(2j+1)^{1/2}}{5} \cdot [C_{Lj}(20;00)]^2$$
 (26)

BOUNDARY CONDITION CONSIDERATIONS

Valuable light might indeed be shed upon the fundamental role of the nucleus in alpha decay if one could integrate inward equations (13) with boundary conditions based on experimental alpha group intensities, carrying the solutions up to all parts of the nuclear surface. The magnitude and angular variation of the solution at the nuclear surface might in turn suggest a nuclear model giving an internal solution to match this external one.

Experimental alpha group intensities establish half the necessary boundary conditions at large distance. 15 Assuming trial values for the phases one could integrate the imaginary component of the solution inward tryingtdifferent phase angles of the waves at large r until the phases were found which made the imaginary component become vanishingly small within the barrier region. With these phases the total necessary boundary conditions except for an ambiguity in sign, discussed below, would be known, and the real components of the waves could be integrated in to $R_{\rm max}$.

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Then one might in principle (considering the two dimensional analogous problem) join the solution smoothly to a solution in a spheroidal coordinate system fixed in the nucleus and carry the solution on to the spheroidal nuclear surface.

A simpler approximate procedure might be merely to bring the wave from R_{max} to the nuclear surface by considering the wave to behave roughly as given by the one-dimensional solutions at fixed angle ω .

It should be pointed out that the trial and error method for establishing the proper phase boundary conditions does not give a single unique set of phases. The condition for acceptibility was that the imaginary components tend to zero in the barrier region. Insofar as the different L-waves may tend to zero with positive or negative signs an ambiguity is present. The several possible cases would need to be considered separately, each leading to different behavior of the solution at the nucleus. Only one of these cases will represent the actual physical behavior.

DISCUSSION

The correlation probability function with single peaks at ω -0 and π is composed of Fourier components of like sign, while the function with a single peak at $\pi/2$ has Fourier components of alternating sign.

Assuming the qualitative correctness of the Hill and Wheeler³ proposal that alpha decay preferentially seeks the thinnest portion of the barrier and that the alpha wave is of roughly equal amplitude at all parts of the nuclear surface, it follows that the solutions of (13) would near the nucleus be all of like sign for prolate distortion and of alternately opposite sign for oblate distortion. The sign of the quadrupole coupling

terms in (13) will be seen in either case to give an increased dissipation of the waves when the solution is peaked at the thinnest part of the barrier. Such merely corresponds to the fact that the quadrupole potential is a maximum in the direction of maximum radial excursion of the nucleus. The quadrupole potential by its coupling action will generally tend to cause the peaks of the wave function near the nucleus to migrate toward angles $\pi/2$ removed from the initial peaks. The general behavior of individual L-waves during such migration might qualitatively be appreciated from a simple example. Consider the correlation function to consist of Dirac δ -functions at two angles

$$f(\omega) = \frac{1}{2}\delta(\omega) + \frac{1}{2}\delta(\pi - \omega)$$
 (27)

Expanding in normalized Legendre functions $S_n(\cos \omega)$ [cf. equation (24)] and evaluating coefficients $f(\omega) = \sum_{i} C_i S_i(\cos \omega)$ (28)

$$C_{n} = \int_{0}^{\pi} \frac{1}{2} [\delta(\omega) + \delta(\pi - \omega)] S_{n}(\cos \eta) \sin \eta d\eta$$

$$C_{n} = \begin{cases} 0, \text{ odd} \\ S_{n}(\cos \omega), \text{ neven} \end{cases}$$
 (29)

Figure 1 shows the variation of the first three \textbf{C}_n 's as a function of $\ \omega.$

Table I gives the ratio of ℓ = 2 and ℓ = 4 waves to ℓ = 0 at the nucleus as calculated from experimental data* assuming no quadrupole coupling effects. The approximate expression for the WKB penetration factor P when differentiated gives $d\log_{10}P/dE = 0.854(Z-2)E^{-3/2}$ (where Z is the atomic number of the alpha emitter and E is the alpha energy in Mev). The energy difference effect was calculated from this formula, and the centrifugal barrier effect was calculated using formulas derived by Preston. The centrifugal reduction factors of the penetrability are calculated to be nearly the same for all alpha emitters of Table I and equal on the average 0.64 for ℓ = 2 and 0.23 for ℓ = 4.**

Table A

| Alpha Emitter | Energy of 4+ State of Daughter (kev) | Alpha Group Abundance (% of total alpha) |
|---|--|--|
| Ra ²²⁴ Th ²²⁶ Th ²²⁸ Th ² 30 | region ~280-884 297 253 (210) | <0.1 2.4 0.2 ((0.8) |
| U ²³⁰ U ²³² U ²³⁴ Pu ²³⁸ | 230 189 (170) 146 | ~0.5 0.3 (0.5) |
| Pu ²³⁰ Pu ²⁴⁰ | 151 | 0.1 |

Intensities in parentheses are from gamma measurements; all other, from alpha spectroscopy.

Table B

| Alpha Emitter | Energy of 2+ State (kev) | Alpha Group Abundance (% of total alpha) | |
|-------------------|-----------------------------|--|--|
| Pu ²³⁴ | ~47 | 15 ± 3 | |
| Cf ²⁴⁶ | ~43 | 12 | |

Energies and intensities determined from conversion electron coincidence counting in nuclear emulsions.

^{*}Data are taken from the Table of Isotopes (Hollander, Perlman, and Seaborg, Revs. Modern Phys. 25, 469 (1953)) except for the following (privately communicated by Asaro, Stephens, and Perlman for Table A; privately communicated by Dunlavey for Table B).

Devaney, who obtains 0.37 and 0.0368 for ℓ = 2 and ℓ = 4 groups, respectively.

Table I

| pha Emitter Apparent Relative Wave Amplitudes at l.l x 10-12 cm (neglecting quadrupole coupling effects) | | Wave Amplitudes at x 10-12 cm | Zg /E 1/2 |
|--|---------------------------------|----------------------------------|--------------------------------------|
| | w2 : w0 | w ₄ : w ₀ | |
| 224 Ra Ra Ra | 1.2 1.4 | | 5.53 6.27 |
| Th 226 Th 228 Th 230 Th 232 | 1.2 1.3 1.2 1.2 | 1.8 0.64 (1.4) | 8.14 9.56 10.7 11.9 |
| U ² 30 U ² 32 U ² 34 U ² 36 U ² 38 | 1.1 1.2 1.1 1.2 1.1 | 0.7 0.5 (0.7) | 10.8 11.8 13.1 12.7 13.4 |
| Pu234 Pu236 Pu238 Pu240 Pu | 0.7 0.8 1.0 1.0 | 0.22 0.25 | 13.4 13.7 13.7 13.9 |
| Cm ²¹ +2 Cm ²¹ +1+ | 1.0 1.0 | 0.11 | 14.2 14.3 |
| Cf ²⁴⁶ | 0. 6 | . | 14.6 |

The most striking behavior seen from Table I is the sharp decrease 17 in the 4:0 ratio with increasing Z, while the 2:0 ratio remains nearly constant (decreasing slightly). While the explanation for this behavior may also imply a change in relative amplitudes at the nucleus, the general trend of the 4:0 and 2:0 ratios is at least partly explained simply in terms of the external quadrupole coupling effects. It is seen from Figure 1 that the operation of the coupling terms in equations (13) on simple peaked angular distributions is such that the various L-waves (except L = 0)

actually go through nodes. The intrinsic quadrupole moment is increasing with mass number in the heavy region (cf. Ford 18) and perhaps to sufficient magnitude to bring the ℓ = 4 wave nearly to its first node. Over this region the ℓ = 2 wave will only be decreased slightly relative to ℓ = 2, its first node being further removed from the initial conditions.

This explanation in terms of quadrupole coupling on the external solution would predict that for nuclei of still greater quadrupole moments than the heaviest in Table I the ℓ = 4 alpha group should go to zero and then become more abundant again. The low 2:0 ratio in Cf ²⁴⁶ is perhaps evidence that the 2:0 node is being approached.

In the absence of lifetime measurements or coulomb excitation cross sections directly determining intrinsic quadrupole moments in the heavy region, let us deduce an approximate dependence of Q_0 on atomic number, mass number, and the energy of the first excited state. From equations (V.7) (VI.1) and (II.6a) of Reference A it can be determined that

$$|Q_0| \approx \text{(const.)}. \quad ZE_2^{-1/2}A^{-1/6}$$
 (30)

where Z is the atomic number, E2 the energy of the the first excited state and A, the mass number.

From lifetime data in the 160-176 mass region as given in Table XXVII of A the constant in (30) has the value of about 2.9 (E_2 being expressed in kev). As a first guess then, one might take (for mass numbers near 230) the expression

$$|Q_0| \approx 2.9/230^{1/6} ZE_2^{-1/2} = 1.2ZE_2^{-1/2}$$
 (31)

where Q_0 is in $10^{-2\frac{1}{4}}$ cm² and E_2 in kev.

The last column of Table I gives the values of the parameter $\mathbb{Z} \mathbb{E}_2^{-1/2}$, which should be approximately proportional to the intrinsic quadrupole moment. Of course, the coupling effect will depend not only upon the quadrupole moment but upon the energies and energy differences of the alpha groups. The sign of the quadrupole moment is a matter for speculation.

Superimposed on the general decrease are erratic variations. The great differences between various thorium and uranium isotopes differing by only a pair of neutrons suggests that specific nuclear effects depending on nucleon configurations may play a significant role. One observes from Table I that along with the high-low alternation of the \mathbf{w}_4 : \mathbf{w}_0 ratio in thorium and uranium isotopes there is an opposing high-low alternation of the \mathbf{w}_2 : \mathbf{w}_0 , but the significance is obscure. It should be pointed out that Asaro, Stephens and Perlman¹⁹ have found an additional alpha decay group in Th²²⁸ going to a spin one, odd parity state of energy slightly lower than the 4+ state. A negative parity state cannot belong to the rotation-vibration-spectrum based on the even parity ground state in the strong surface coupling model but must represent a different particle configuration. The additional complexity of the situation in Th²²⁸ sounds a note of caution regarding any detailed interpretation of the values of Table I.

From considerations set forth earlier in this paper, one might hope to relate the relative values of the ℓ -waves at a particular radial distance to a simple angular correlation function between alpha position and nuclear orientation. One might assume, for example, that there is equal probability of alpha emission from all parts of the nuclear surface.

n. 1

Simple considerations of the dissipation of alpha waves in the barrier region would then lead one to expect very sharply peaked correlations at the angles of maximum radial excursion of nuclear matter. A delta function distribution like (27) would lead to the following initial relative values for positive and negative deformations:

Table II

Initial Fourier Components of Delta Function Correlations

Wave Positive (prolate) Negative (oblate)

| | $\mathbf{w} = 0, \pi$ | | $\omega = \pi/2$ |
|------------------|-----------------------|-------------|---|
| \mathbf{w}_{0} | 1 | gg a so a g | 1 |
| Sw | 2.2 | | -1.1 |
| $w^{j_{4}}$ | 3.0 | | 1.1 2. 1.1 1.1 1.1 1.1 1.1 1.1 1.1 1.1 1.1 1 |
| | | | |

For distributions of finite width the higher components will be decreased, but for even half the distortions given by Ford 18 for this region the distributions would still be narrow enough that the relative amplitudes of $w_{\rm h}$ and $w_{\rm 2}$ would probably not be appreciably less than their delta function values. The ratios in Table I for radium and thorium isotopes would be expected to approach nearest to the actual ratios at the nucleus, since the neglected quadrupole coupling effects will be smallest for them. (The quadrupole coupling would be expected to decrease the ratios from initial peaked values.)

The fact that the ratios exceed in several cases the maximum values expected with oblate distortion is a point favoring prolate distortion in these heavy nuclei. The approach to the limiting values of Table II for

prolate distortion is in no case very close. Thus, the assumption of equal wave amplitude at all parts of the nuclear surface may not correspond to reality. Indeed, it seems plausible that internal nuclear factors might lead to a node in the alpha wave at $\omega=0$, giving a zero amplitude at the nuclear surface at the angle of thinnest barrier. Perhaps the alpha emission probability is proportional to the probability that the four nucleons of the most loosely bound neutron and proton pairs are at a certain position on the nuclear surface. That is, the alpha wave function at the nuclear surface might be roughly proportional to a product of the nuclear wave functions of its constituent nucleons. In the Bohr-Mottelson strong coupling model all nucleon wave functions in the spheroidal well except those with quantum number $\Omega=1/2$ will have nodes along the nuclear symmetry axis.

The radial correlation function near the nucleus may then be sharply peaked but at some angle greater than $\omega=0$. It is also possible that multiple-peaked correlation functions may result.

Other evidence supporting the idea of general occurrence of the axial node at the nuclear surface comes from the studies of absolute alpha decay rates. Various alpha decay correlations of even-even nuclei using the conventional spherical nucleus formulas have shown the decay rates to be consistent with effective nuclear radii from ordinary radius formulas, the three nuclei Po²⁰⁸, Po²¹⁰, and Em²¹² constituting the only exceptions, being slower than expected by a factor of ~20 for the poloniums and ~5 for Em²¹². Hill and Wheeler³ suggested that the explanation for these exceptions might be simply in their having less quadrupole distortion and hence thicker barriers than the other nuclei. Carrying this argument out

in detail would lead one to expect that the various alpha decay correlations should have shown the effective nuclear radius to increase rapidly going from radium isotopes to curium, since the decreasing first excited states imply greatly increasing quadrupole distortion. That this is not the case could be evidence for the general occurence of an axial node, preventing the alpha decay process from taking full advantage of the thinner barrier in the axial direction.

The reason for the hindrance of Po^{208} , Po^{210} , and Em^{212} is more likely due to a relatively large change in nuclear shape between initial and final nuclei. Such hindrance effects analogous to the Franck-Condon principle in molecular spectra have been discussed by both Hill and Wheeler³ and by Bohr and Mottelson.² Po^{210} , having just two protons beyond a closed shell must have an oblate shape, while its alpha decay daughter Pb^{206} with two neutron holes in the closed shell structure must have a prolate shape. Similarly Em^{212} would be probably of oblate shape and Pb^{204} of prolate shape. Po^{208} having two protons and two neutron holes would be of intermediate shape between Em^{212} and Pb^{204} .

It does not seem worth-while pursuing much further any speculations based on the ratios of Table I, where the quadrupole coupling terms in alpha decay have been ignored. The coupling terms may be important if quadrupole moments of the order of the estimate (31) obtain. For Cm^{242} alpha decay (31) gives a moment of $|Q_0| \approx 17 \times 10^{-24} \text{cm}^2$, and the various quantities of equations (13) take on the following values (with unit distance 10^{-13} cm):

$$4mZe^2/\hbar^2 = 51.0$$
 $2mE/\hbar^2 = 1.179$ $m/\sqrt{3} = 0.0014$ $2m|Q|e^2/\hbar^2 = 460$.

It was found for these numerical values that the perturbation methods derived by Preston⁵ are not applicable, because of the large coupling and small energy differences between states. Other methods are being investigated.

Of the greatest importance to the application of the thoery outlined here would be lifetime measurements for the E2 gamma transitions of even-even nuclei, measurements of cross sections for coulomb excitation, and measurements of spectroscopic quadrupole moments of odd nuclei, even the sign allone being of considerable importance.

Finally, it is to be emphasized that despite the use of Bohr-Mottelson strong coupling nuclear wave functions the quadrupole coupling effect in alpha decay as expressed in equations (13) is not dependent upon the validity of their nuclear model. The derivation could have proceeded from simple consideration of the E2 transition probabilities between the nuclear states. The coupling of different alpha decay groups is an inevitable consequence of the electromagnetic transition field just as is the internal conversion process for gamma rays. The magnitude of the coupling is directly related to the gamma transition probability. The effect thus would be of most importance for fast transitions. Lifetime measurements in the heavy rare earth region have showed that many E2 transitions are of this especially fast nature. If the E2 transitions connecting excited states of even nuclei in the heavy region are also of this fast nature, the possible special importance of quadrupole coupling in alpha decay follows.

Also the phenomenon of interference of different alpha waves near the nucleus to produce peaked correlation functions is not specially dependent upon nuclear models with stable spheroidal distortions. If, for example, the angular momentum in the "rotational" excited states were carried by a single proton in a spherical nucleus, the correlation function would relate the angular positions of the proton and the alpha particle. Similarly, if the angular momentum were thought to reside in a group of nucleons in a spherical well, the correlation function could be appropriately defined. However, despite the fact that the coupled alpha decay description of this paper could be generalized to nuclear models other than those giving actual spheroidal distortion, the great usefulness of the strong surface coupling model in visualizing and formulating the alpha decay treatment is clearly evident. The systematic decrease with atomic number of the alpha transition rate to the 4+ state is not easily explained with assumption of a spherical nuclear shape, but the stable spheroidal shape allows the simple explanation discussed earlier.

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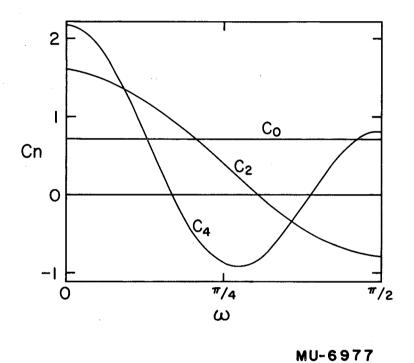


Fig. 1. Fourrier Components of a delta function distribution. f (θ) = 1/2 $\delta(\omega)$ + 1/2 $\delta(\pi-\omega)$.