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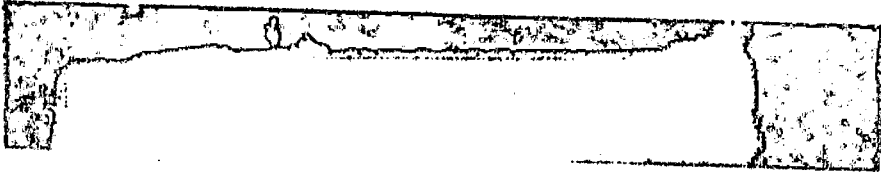
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THEORETICAL STUDIES OF THE ALPHA DECAY OF  $U^{233}$

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THEORETICAL STUDIES OF THE ALPHA DECAY OF  $U^{233}$ \*

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February 1959

ABSTRACT

The alpha decay of a deformed odd-mass nucleus,  $U^{233}$ , is treated by the use of numerical integration on an IBM-650 computer. The results of this treatment are compared with the theory of Bohr, Fröman, and Mottelson.

Approximate analytic methods are developed for predicting the intensities of the higher members of the ground rotational band.

A comparison is made between the numerical integration and the experiments of Roberts, Dabbs, and Parker, in which they examine the angular distribution of alpha particles from aligned  $U^{233}$  nuclei.

The results of the numerical integration of  $U^{233}$  are presented in matrices analogous to those of Fröman, and numerical values of the functions are given for selected values of "r".

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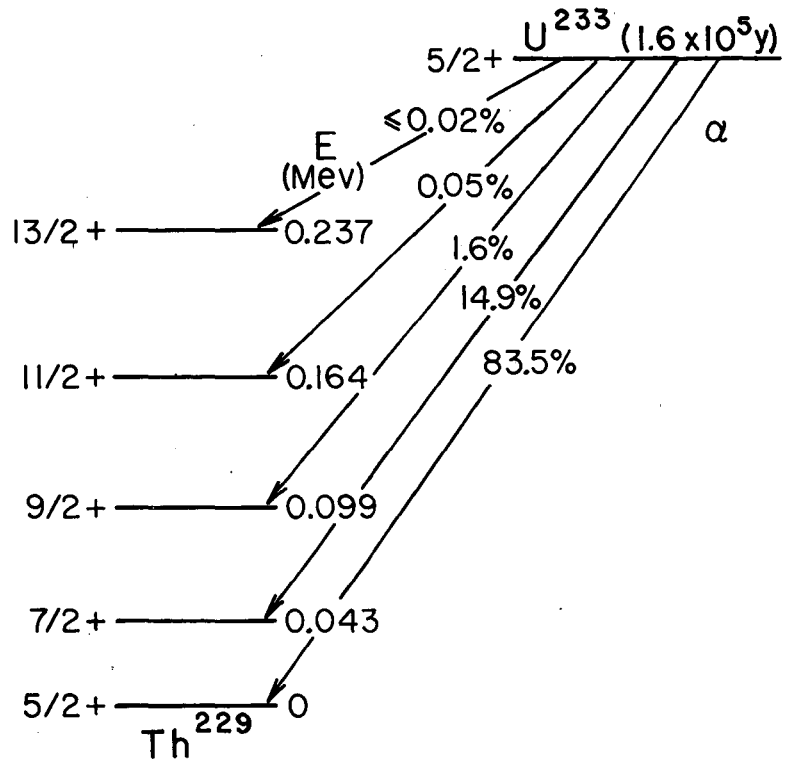
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## I. INTRODUCTION

The theory of Bohr-Fröman-Mottelson (B.F.M.) makes definite predictions concerning the amplitudes of alpha partial waves at the nuclear surface, in the case of deformed nuclei.<sup>1</sup> It was decided to test the validity of their prediction in the case of the alpha decay of  $U^{233}$  by carrying out extensive numerical integrations of the alpha wave equation including the nuclear quadrupole interaction. The relative intensities of the alpha particles to the low-lying states of  $Th^{229}$  have been measured;<sup>2</sup> there is a good deal of confidence in the spin assignments of these levels, and there are estimates of the nuclear quadrupole moment.<sup>3,4</sup> We use the estimates for the quadrupole moment of  $U^{233}$ , as there are none available for  $Th^{229}$ . We expect the quadrupole moment of  $Th^{229}$  to be roughly the same as that of  $U^{233}$ . This information is summarized in Fig. 1.

The study of  $U^{233}$  is of interest for reasons other than the comparison with B.F.M. At the time the problem was undertaken, alpha intensities had been reported for higher members of the ground rotational band,<sup>5</sup> intensities which differed considerably from B.F.M. predictions. Since that time, however, the gamma rays following the alpha decay have been examined, and the large intensity of high-energy gamma rays indicates that the higher levels populated by the alpha decay are not all members of the ground rotational band.<sup>6</sup>

Roberts, Dabbs, and Parker<sup>7</sup> have aligned  $U^{233}$  nuclei at low temperatures in single crystals of  $Rb(UO_2)(NO_3)_3$  and examined the anisotropy of emitted alpha particles. They interpret their results as indicating that the  $l = 2$  wave is out of phase with the  $l = 0$  alpha partial wave in the alpha group to ground. Their experiment also puts limits on the amount of the  $l = 2$  wave which populates the ground state of  $Th^{229}$ .



MU-16869

Fig. 1. Alpha decay of  $U^{233}$ .

We might hope that either the intensity limits of Roberts, Dabbs, and Parker or the boundary conditions at the nuclear surface of B.F.M. will eliminate one of the phase choices for the  $\ell = 2$  partial wave relative to the  $\ell = 0$ .

We have also developed approximate analytic methods to predict intensities of the alpha decay populating the  $11/2$  and  $13/2$  members of the  $\text{Th}^{229}$  ground state rotational band.

## II. FORMULATION OF THE ALPHA-DECAY PROBLEM

The problem of alpha decay in the region of uranium is complicated, as contrasted to the region of lead, by the existence of large quadrupole moments which interact with the escaping alpha particle. Favored alpha decay, i.e., decay between parent and daughter states having the same nucleonic wave functions and hence the same  $K$  ( $K$  is the projection of the nuclear spin on the nuclear symmetry axis), has been treated in this region of large quadrupole moments by several authors, both by numerical integrations<sup>8,9,10</sup> and by analytic approximations<sup>11</sup> for the case of even-even nuclei. The quantum mechanical treatment of an odd-even nucleus is quite similar to that of the even-even nucleus.<sup>12</sup> We start from Schroedinger's equation

$$H \psi = E \psi \quad (1)$$

Here we have  $H = T + V + H_{\text{nuc}}$  where  $T$  is the kinetic energy of the system,  $V$  its potential energy, and  $H_{\text{nuc}}$  the Hamiltonian for the internal energy of the recoil nucleus. We expand the potential  $V$  in spherical coordinates. Making the usual multipole expansion, we obtain

$$V = 2e \sum_{p=1}^Z \sum_{\lambda=0}^{\infty} \frac{e_p r_p^\lambda}{r^{\lambda+1}} P_\lambda(\cos \gamma) \quad (2)$$

where  $\underline{r}$  gives the position of the alpha particle;  $\underline{r}_p$  gives the position of the  $p$ th proton in the daughter nucleus, and  $\gamma$  is the angle between  $\underline{r}$  and  $\underline{r}_p$



in the system of the recoil nucleus. In our treatment of the problem, we include the central and quadrupole terms of the potential. We next construct a solution of Schroedinger's equation of the form

$$\psi = \sum_{l', I_f', T'} r^{-1} U_{l', I_f'}(r) Y_{l', I_f', T'}^{IM}(\theta, \varphi, \chi_i) \quad (3)$$

as the first step in the solution. Here

$$Y_{l', I_f', T'}^{IM}(\theta, \varphi, \chi_i) = \sum_{m'} \left( \left\langle l', I_f', m', M-m' \mid IM \right\rangle Y_{l', m'}(\theta, \varphi) \right. \\ \left. \times \Phi_{I_f', T'}^{M-m'}(\chi_i) \right) \quad (4)$$

$l'$  is the angular momentum of the alpha particle, and  $m'$  is the component of its angular momentum on a space-fixed axis,  $I_f'$  and  $I$  are the final and initial nuclear angular momenta, and  $M-m'$  and  $M$  are projections of  $I_f'$  and  $I$  on the space-fixed axis, and  $T'$  represents all other quantum numbers specifying the nuclear state. Here  $\Phi_{I_f', T'}^{M-m'}(\chi_i)$  describes the intrinsic state of the daughter nucleus and the bracketed symbol is a Clebsch-Gordan coefficient. The orthogonality condition on the  $Y_{l', I_f', T'}^{I, M}$  function is

$$\int Y_{l', I_f', T'}^{IM} Y_{l, I_f, T}^{IM} \sin \theta d\theta d\varphi d\chi_i = \delta_{ll'} \delta_{I_f I_f'} \delta_{TT'} \quad (5)$$

We next substitute  $\psi$  into Schroedinger's equation, multiply by

$$Y_{l, I_f, T}^{I, M}$$

and then integrate over all variables but  $r$ . We do this for each value of  $l, I_f$  of interest in the daughter nucleus and we are left with a set of coupled ordinary differential equations of the form

$$\begin{aligned}
& \frac{-\hbar^2}{2M} \frac{d^2 U_{\ell I_f}}{dr^2} + \left[ \frac{\hbar^2}{2Mr^2} \ell(\ell+1) + E_{I_f T} - E \right] U_{\ell I_f} \\
& + 2Ze^2 U_{\ell I_f} / r + \sum_{\ell' I_f'} U_{\ell' I_f'} \left\langle Y_{\ell I_f T}^{IM*} \left| \frac{Q_0 e^2}{r^3} P_2(\cos \gamma) \right| Y_{\ell' I_f' T'}^{IM} \right\rangle = 0
\end{aligned} \tag{6}$$

All terms but the last summation describe the interaction of two charged particles. The last term gives a mixing of states, due to the perturbation induced by the nuclear quadrupole moment.

### III. A TEST OF THE B.F.M. HYPOTHESES

The hypotheses of B.F.M. may be stated as follows: any alpha partial wave has a projection of its angular momentum on the nuclear-symmetry axis which is equal to  $K_f \pm K_i$  at the nuclear surface.  $K_f$  and  $K_i$  are the projections of the spins of the final and initial nuclear states on the nuclear-symmetry axis. For  $(K_f + K_i) > \ell$  there is but one permissible value of  $m_\ell$ , which is zero in the case of favored alpha decay. Looking at the B.F.M. hypothesis in an  $| I_f, \ell \rangle$  representation, one sees that a given  $\ell$  wave will be apportioned among the states that it populates in proportion to the Clebsch-Gordan coefficient  $\langle I_i, \ell, K, 0 | I_f, K \rangle$  at the nuclear surface. This hypothesis will be referred to as B.F.M.-1. Furthermore, B.F.M. makes the approximation that the relative intensities of alpha decay to the various levels from population by a given  $\ell$  wave will be given by the square of the Clebsch-Gordan coefficient  $\langle I_i, \ell, K, 0 | I_f, K \rangle$  times the barrier penetration factor for the particular alpha energy. This approximation, which we shall refer to as B.F.M.-2, would be exact only in the limit of infinite moment of inertia. In the case of favored alpha decay of even-even nuclei, a given alpha partial wave populates just one level of the daughter nucleus and so affords us no test of the B.F.M. hypotheses.

In the numerical work to be described, B.F.M.-1 and B.F.M.-2 are tested separately for the first time. In the case of odd-even nuclei, we

may utilize B.F.M.-1 to set boundary conditions at the nuclear surface for solutions of the alpha wave equations and then compare the alpha intensities from numerical integration with the experimentally observed intensities. This gives us a test of B.F.M.-1, though our calculations did not include  $l = 4$  contributions. In the region of  $U^{233}$  the  $l = 4$  contribution should not be very significant.

#### IV. NUMERICAL INTEGRATION AND BOUNDARY CONDITIONS

To return to the specific problem of  $U^{233}$ , the equations describing the alpha particles as they leave the nucleus are:

$$\frac{d^2 u_0}{d r^2} + \left[ .929 - \frac{48.78}{r} \right] u_0 - \frac{1}{r^3} \left[ 101 u_1 - 117 u_2 + 69.3 u_3 \right] = 0 \quad (7a)$$

$$\frac{d^2 u_1}{d r^2} + \left[ .929 - \frac{48.78}{r} - \frac{6}{r^2} \right] u_1 - \frac{1}{r^3} \left[ 101 u_0 + 38.71 u_1 - 76.0 u_2 - 26.4 u_3 \right] = 0 \quad (7b)$$

$$\frac{d^2 u_2}{d r^2} + \left[ .921 - \frac{48.78}{r} - \frac{6}{r^2} \right] u_2 - \frac{1}{r^3} \left[ 117 u_0 - 76.0 u_1 - 2.58 u_2 - 76.3 u_3 \right] = 0 \quad (7c)$$

$$\frac{d^2 u_3}{d r^2} + \left[ .910 - \frac{48.78}{r} - \frac{6}{r^2} \right] u_3 - \frac{1}{r^3} \left[ 69.3 u_0 - 26.4 u_1 - 76.4 u_2 - 18.1 u_3 \right] = 0 \quad (7d)$$

where  $r$  is in units of  $10^{-13}$  cm;  $\frac{u_0}{r}$  is the radial function of the  $l = 0$  wave populating the  $5/2$  state of  $\text{Th}^{229r}$ , and  $\frac{u_1}{r}$ ,  $\frac{u_2}{r}$ , and  $\frac{u_3}{r}$  are the radial functions

of the  $l = 2$  wave populating the  $5/2$ ,  $7/2$ , and  $9/2$  levels of  $\text{Th}^{229}$ , respectively. This set of equations includes only the  $l = 0$  and  $l = 2$  alpha partial waves. In this treatment we use a value of 13.2 barns for  $Q_0$ , and the alpha-decay energies are 4.900, 4.857, and 4.801 Mev.

We note that as  $r$  approaches  $\infty$ , the coupling term, which has a  $1/r^3$  dependence, becomes negligible. When we can ignore this term, the equations are decoupled, and their solutions are given by linear combinations of regular ( $F_l$ ) and irregular ( $G_l$ ) Coulomb functions. Because the alpha particles are outgoing waves, the solution must be of the form  $Ce^{ikx'}$  and not have any component of the form  $e^{-ikx'}$ , where

$$x' = \rho - \eta \ln 2\rho - \frac{l\pi}{2} + \sigma_l \quad (8)$$

The notation is that of the general usage in Coulomb functions.<sup>13</sup> We note that as  $r$  approaches  $\infty$ ,  $F_l$  approaches  $\sin x'$ , and  $G_l$  approaches  $\cos x'$ . Therefore, at large distances, our solutions must be of the form  $(A + iB)(G_l + iF_l)$ , where  $A$  and  $B$  are real constants. Pennington and Preston show in detail how the solutions approach this asymptotic form.<sup>10</sup>

The determination of the radial wave functions was accomplished by numerical integration on an IBM-650 computer in the region where the coupling could not be neglected. As we have four second-order differential equations, there are eight boundary conditions which must be applied. We begin by looking at the imaginary part of the solution.

The procedure adopted was to give one of the  $u$ 's an amplitude of one at a sphere of radius  $9.0 \times 10^{-13}$  cm (the nuclear surface) and the other three were given amplitudes of zero. The nonzero function was started off as a regular Coulomb function, and the other three were kept at zero by conditions used on the derivatives. Carrying out the numerical integration for this set of boundary conditions, we obtain one set of solutions. When  $u_1 = 1$  at the nuclear surface, at infinity we have  $u_j = A_{1j} F_j + B_{1j} G_j$ . The coefficients  $A_{1j}$  and  $B_{1j}$  are obtained by fitting the numerical values from the computer program to linear combinations of Coulomb functions at large distances, in this case  $8.5 \times 10^{-12}$  cm. By separately setting each of the four  $u_1$ 's equal to one, we obtain four independent solutions of the differential equations. The imaginary part of the solution of the physical

problem will be some linear combination of these four solutions, i.e., our solutions of physical interest have the form

$$\Im(u_i) = \sum_{j=0}^3 \alpha_j (A_{ji} F_i + B_{ji} G_i), \quad (9)$$

where  $\alpha_j$  is some real number. We make use of the experimental intensities of alpha particles populating the various levels by noting that the intensity is

$$I_i = \left( \sum_{j=0}^3 \alpha_j A_{ji} \right)^2 + \left( \sum_{j=0}^3 \alpha_j B_{ji} \right)^2 \quad (10)$$

The experimental intensities give us three boundary conditions, a trivial condition of over-all normalization and two relative intensities. For the final boundary condition, we make use of B.F.M.-1 to obtain the ratio of amplitudes of the  $\ell = 2$  wave at the nuclear surface populating the 5/2 and 7/2 states of  $\text{Th}^{229}$  for the real part of the solution. Finally, as a test of B.F.M.-1, we may examine the relative amplitudes at the nuclear surface of the real part of the  $\ell = 2$  wave populating the 5/2 and 9/2 levels of  $\text{Th}^{229}$ . If the B.F.M.-1 hypothesis is valid, all five conditions will be satisfied. We have calculated the real part of the wave function by using our knowledge of the asymptotic form of real and imaginary components of the wave function to obtain

$$\Re(u_i) = \sum_{j=0}^3 \alpha_j (A_{ji} G_i - B_{ji} F_i) \quad (11)$$

at  $r = 8.5 \times 10^{-12}$  cm. We integrate inward numerically.  $A_{ji}$  and  $B_{ji}$  will be the same constants as were obtained from the imaginary part of the solutions, and the set of four  $\alpha_j$  values that comes closest to satisfying the three intensity conditions and the two constraints put on the real part of the solution by B.F.M.-1 is used.

## V. RESULTS OF NUMERICAL INTEGRATION

We found a set of  $\alpha_j$  values which approximately satisfies the conditions mentioned previously for the  $l = 2$  wave both in phase and out of phase with the  $l = 0$  wave at the nuclear surface. The conditions were not satisfied exactly in either case but were satisfied approximately in both cases. The results for the amplitudes of the real parts of the wave functions on a sphere in the nuclear surface region are given in Table I.

Table I

Relative D-Wave Amplitudes at $r = 9.0 \times 10^{-13}$ cm.			
$I_f$	B.F.M.-1 prediction	$l = 2$ in phase	$l = 2$ out of phase
5/2	-0.86	-0.86	-0.88
7/2	1	1	1
9/2	-0.59	-0.55	-0.51

Another way of stating B.F.M.-1 is as follows: in an  $|\ell, m_\ell\rangle$  representation only the component having  $m_\ell = 0$  will be present in favored alpha decay. We transformed the wave functions to an  $|\ell, m_\ell\rangle$  representation and found the  $m_\ell = 0$  component of the  $l = 2$  wave to be some two orders of magnitude larger than the other  $m_\ell$  components. The deviations from perfect agreement with B.F.M.-1 may be attributed to slight inaccuracies in the reported alpha intensities and incorrect choice of quadrupole moment in this calculation, or, finally, to the neglect of alpha particles with angular momenta greater than  $l = 2$ . Our calculations support the validity of B.F.M.-1.

From our wave functions we may calculate the phase shifting of the alpha partial waves caused by the nuclear quadrupole moment. This information will be of interest in the case of an odd-even nucleus, because it enters into calculations of angular distributions from angular correlation experiments and nuclear alignment and nuclear polarization experiments. The phase shifting of the  $i$ th alpha partial wave by the nuclear quadrupole moment is given by the relation

$$\theta_i = \tan^{-1} \left( \frac{\sum_{j=0}^3 \alpha_j B_{ji}}{\sum_{j=0}^3 \alpha_j A_{ji}} \right) \quad (12)$$

where  $\theta_i$  is given in radians.

The calculated phase shifts are given in Table II in degrees.

Table II

Phase shifts caused by nuclear quadrupole moment		
$I_f, l$	$l = 0$ and $l = 2$ in phase	$l = 0$ and $l = 2$ out of phase
5/2,0	- 1.1°	+ 1.94°
5/2,2	- 14°	+ 0.87°
7/2,2	- 5.3°	- 0.179°
9/2,2	- 2.8°	+ 7.16°

These phase shifts may be compared with those calculated for even-even alpha emitters in this region,  $\approx -3^\circ$  for the  $l = 2$  wave when it is in phase with the  $l = 0$  wave. In nuclear-alignment experiments, only the difference in phase shift of the  $|5/2,2\rangle$  and  $|5/2,0\rangle$  is of direct interest. The interference term contributing to anisotropy has a factor  $\cos(\theta_0 - \theta_2)$ . The Coulomb phase difference between S and D waves here is about  $-7^\circ$  and, correcting for the quadrupole interaction, we have  $\approx -10^\circ$  for the  $l = 0$  partial wave in phase with the  $l = 2$  wave, and  $\approx -6^\circ$  for the  $l = 0$  partial wave out of phase with the  $l = 2$  wave.

Next let us examine our numerical integration calculations to test the B.F.M.-2 approximation. From B.F.M.-2, one may predict the amount of  $l = 2$  wave and the amount of  $l = 0$  wave populating the  $5/2$  state of  $\text{Th}^{229}$  independent of nuclear-alignment experiments. Using data from neighboring even-even nuclei, B.F.M. conclude that 81% of the ground-state intensity is due to the  $l = 0$  wave, and 19% comes from the  $l = 2$  alpha wave.<sup>1</sup> In the numerical calculation, our modified prediction is that only 75% of the ground-state intensity is due to the  $l = 0$  wave, and 25% is due to the  $l = 2$

wave. This conclusion holds for both choices of phase of the  $l = 2$  wave, under the constraint that B.F.M.-1 and the intensity conditions hold. This discrepancy suggested that one might find some other approximation that is superior to B.F.M.-2 but simpler than numerical integration to predict unseen intensities of odd-even nuclei and partial wave amplitudes at the nuclear surface for even-even nuclei.

## VI. AN APPROXIMATE TREATMENT OF ALPHA DECAY

An approximate method that was developed<sup>14</sup> and was used to treat  $U^{233}$  will be described briefly.

We see that the solutions of the set of equations (7) would be regular or irregular Coulomb functions were it not for the quadrupole moment, that is, the right hand side of the equations would vanish if  $Q_0$  were zero. From an examination of the series expansion of the W.K.B. integrand, we surmise that the radial wave functions of the alpha partial waves might be well represented by functions of the form

$$(\alpha_{l, I_f} + r^{-3/2} \beta_{l, I_f}) G_{l, I_f}(r) \quad (13)$$

in the region of the nuclear surface. Here  $\alpha_{l, I_f}$  and  $\beta_{l, I_f}$  are parameters fixed over all  $r$  values, and  $G_{l, I_f}$  is the irregular Coulomb function. It is clear that  $\beta_{l, I_f}/r^{3/2}$  approaches zero as  $r$  approaches  $\infty$ , so one may identify  $\sum_l \alpha_{l, I_f}$  as the square root of the quotient of the alpha partial-wave intensity and its velocity.

## VII. APPLICATION OF APPROXIMATE METHOD TO $U^{233}$

To determine the values of the coefficients  $\alpha_{l, I_f}$  and  $\beta_{l, I_f}$ , we apply B.F.M.-1 to obtain two conditions, i.e., the partition of the  $l = 2$  alpha wave between the  $5/2$ ,  $7/2$ , and  $9/2$  states at the nuclear surface. We then substitute the analytic approximation into the differential equations



and demand that the equations be satisfied exactly at some arbitrary intermediate distance ( $2.0 \times 10^{-12}$  cm) to obtain four more conditions. The distance at which we demand that the approximate solutions satisfy the differential equations is somewhat arbitrary; the reasons for choosing  $2.0 \times 10^{-12}$  cm are mainly pragmatic — using this value, we found that we could get best agreement with results of detailed numerical integrations for both  $\text{Cm}^{242}$  and  $\text{U}^{233}$ , but it is to be emphasized that the results are not very sensitive to the distance chosen. The over-all normalization gives us a seventh condition. To obtain the final condition we may do one of two things: (a) we can use the ratio of the  $l = 2$  wave to the  $l = 0$  wave at the nuclear surface obtained from a neighboring even-even nucleus,  $\text{U}^{232}$  or  $\text{U}^{234}$ , or (b) we can use the ratio of any two experimental intensities, bearing in mind that the observed alpha intensity to any level I is equal to  $\sum_l |\alpha_{l,I}|^2$ . Using (b) to obtain the final condition, we are then able to check the approximation with the third experimental intensity and with the amount of  $l = 2$  wave calculated to populate the  $5/2$  state in the numerical integration. We compare the intensity predictions of this treatment with the predictions of B.F.M. We may compare several things in the following manner. We may use B.F.M.-1 as a boundary condition at the nuclear surface and then use B.F.M.-2 and the analytic method described here to calculate intensities at infinity. We may also use the results of the numerical integration to provide boundary conditions at the nuclear surface. We shall adjust the B.F.M. intensity predictions by the use of the relative intensities of the  $5/2$  and  $7/2$  states. The comparisons are found in Table III. The agreement with experiment is fairly good for  $l = 2$  alpha partial waves, using the B.F.M.-2 approximation; however, this method does not take into account different phase choices for the alpha partial waves. If we consider the relatively weaker  $l = 4$  wave populating states of spins  $11/2$  and  $13/2$ , the terms in the radial equations due to the nonvanishing nuclear quadrupole moment become more important in an intensity prediction of alpha decay, and the predictions may vary considerably, depending on the choice of partial wave phases. It is for this application that we feel that the approximate method described here has a considerable advantage over the B.F.M.-2 approximation.

Table III

Test of B.F.M.-2					
$I_f$	Experimental intensity	Boundary conditions at nuclear surfaces			
		B.F.M.-1		Numerical integration	
		B.F.M.-2	Analytic approx.	B.F.M.-2	Analytic approx.
5/2	100	100	100	100	100
7/2	17.9	17.9	17.9	17.9	17.9
9/2	1.9	2.54	2.35	2.2	1.85

A calculation was made by the use of the analytic approximation including the  $l = 4$  partial wave in the alpha decay of  $U^{233}$  to form  $Th^{229}$ . We can then predict the alpha intensities populating the 11/2 and 13/2 states of  $Th^{229}$  that are members of the ground-state rotational band. If we neglect the  $l = 6$  contributions and apply the data on relative amplitudes of alpha partial waves from the neighboring even-even nuclides, we obtain the intensity predictions for four phase choices. We compare these with B.F.M.-2 and experimental observation in Table IV. The experimental values used here are from recent work by Ruiz and Asaro.<sup>6</sup>

Table IV

Relative Intensity Predictions							
$I_f$	Relative phase					B.F.M.	Experi- mental <sup>6</sup>
	$l = 0$	+	+	+	+		
	$l = 2$	+	+	-	-		
	$l = 4$	+	-	+	-		
5/2		100	100	100	100	100	100
7/2		16	18.3	16.8	15.5	13	17.9
9/2		2.46	3.01	2.70	2.37	1.8	1.9
11/2		0.036	0.286	0.180	0.016	0.2	.06 ± .03
13/2		0.007	0.020	0.012	0.0035		≤ .02

Our intensity predictions do not include the effect of the phase shifting due to the quadrupole interaction; i.e., the wave functions at large distances are of the form  $(A_{\ell, I_f} + i B_{\ell, I_f}) (G_{\ell, I_f} + i F_{\ell, I_f})$  and the true intensity is  $\sum_{\ell} [ |A_{\ell, I_f}|^2 + |B_{\ell, I_f}|^2 ]$ . In this approximation, we have taken the intensity to a given level as being  $\sum_{\ell} |A_{\ell, I_f}|^2$ . To estimate the correction, we may calculate phase shifts for a neighboring even-even isotope,  $U^{234}$ , and using the equation  $I_{\ell, I_f} = |A_{\ell, I_f}|^2 + |B_{\ell, I_f}|^2$ , when  $I_{\ell, I_f}$  is the intensity of the  $\ell$ th partial wave populating  $I_f$ , and noting that  $\theta_{\ell} = \tan^{-1} \frac{B_{\ell}}{A_{\ell}}$ , estimate  $B_{\ell}$  from the value of  $A_{\ell}$ , which we have calculated;  $\theta_{\ell}$  is the phase shift and is given for the various sets of phases in Table V, for the 1<sup>st</sup> and 4<sup>th</sup> sets of phase choices. The 2<sup>nd</sup> and 3<sup>rd</sup> sets of phase seen to be ruled out by the experimental value of the 11/2 intensity. The phase shifts are calculated using a method previously developed.<sup>14</sup>

Table V

Phase Shifts Calculated From $U^{234}$ Data			
Phase shift	Relative phase		
	$\ell = 0$	+	+
	$\ell = 2$	+	-
	$\ell = 4$	+	-
$\theta_{\ell=0}$		- .04	+ .04
$\theta_{\ell=2}$		- .11	+ .03
$\theta_{\ell=4}$		- .20	- .22

Using these data, we modify the predicted intensities; only the 11/2 and 13/2 levels are changed perceptibly.

Table VI

Modified Intensity Predictions					
$I_f$	Relative phase		B.F.M.		Experimental
	+	+			
	+	-			
	+	-			
5/2	100	100	100		100
7/2	16	15.5	13		17.9
9/2	2.46		2.37		1.9
11/2	0.037	0.017	0.2		$0.0 \pm .03$
13/2	0.007	0.0037			$\leq .02$

The phase shift corrections here are only approximate, since phase shifts for individual  $U^{233}$  groups will not be identical to those in  $U^{234}$ , especially for the relatively weak  $l = 4$  groups.

#### VIII. COMPARISON WITH NUCLEAR-ALIGNMENT EXPERIMENT

Some experimental data are available on the relative phases of the  $l = 0$  and  $l = 2$  alpha partial waves. Roberts, Dabbs, and Parker have aligned  $U^{233}$  nuclei in a single crystal of  $Rb(UO_2)(NO_3)_3$  and have obtained an angular distribution of alpha particles.<sup>7</sup> They have interpreted their results as indicating that the  $l = 2$  partial wave populating the ground state of  $Th^{229}$  is out of phase with the  $l = 0$  wave. To arrive at this conclusion, they make the assumption that the quadrupole coupling constant,

$$q = \frac{3 e Q_{\text{spec.}}}{4I(2I - 1)} \left\langle \frac{\partial^2 V(0)}{\partial z^2} \right\rangle \quad (14)$$

is negative. Here  $e$  is the electronic unit of charge,  $Q_{\text{spec.}}$  is the spectroscopic value of the nuclear quadrupole moment and

$$\left\langle \frac{\partial^2 V(0)}{\partial z^2} \right\rangle$$

is the gradient of the electronic field evaluated at the surface of  $U^{233}$ .

The calculations of Eisenstein and Pryce<sup>15</sup> are interpreted by Roberts et al.<sup>7</sup> as indicating that

$$\left\langle \frac{\partial^2 v(0)}{\partial z^2} \right\rangle$$

is positive in the  $UO_2^{++}$  ion. As this conclusion is not entirely proved and since it creates difficulties for a comprehensive interpretation of alpha decay of deformed nuclei generally, we also consider the possibility of the  $l = 2$  wave in phase with the  $l = 0$ . If we define the percent of  $l = 2$  admixture in the population of the  $5/2$  state as  $100 \delta^2/(1+\delta^2)$ ; Roberts et al. show from the measurements that they have made that

$$\frac{0.795 \delta^2 + 4.145 \delta + 0.226}{1 + 0.835 \delta^2} \cdot \frac{q}{k} = 0.0625 \pm .0025^\circ K; \text{ where } K \text{ is} \quad (15)$$

the Boltzmann constant.

Using the values of  $\delta$  which we obtained in the numerical integration of  $U^{233}$ , we may then calculate a value for  $q$ . If the  $l = 2$  wave is in phase with the  $l = 0$  wave, we have  $\delta = 0.577$ ; if the  $l = 2$  wave is out of phase with the  $l = 0$  wave, we have  $\delta = -0.577$ . For the  $l = 2$  wave in phase we calculate  $\frac{q}{k} = 0.0277^\circ K$ ; for the  $l = 2$  wave out of phase, we calculate  $\frac{q}{k} = -0.0418^\circ K$ . Roberts et al. give a value for  $|\frac{q}{k}|$  of  $0.0388 \pm .0086^\circ K$  from specific heat measurements, but the sign is not determined in these measurements. Roberts et al. argue that the sign of  $q$  is negative in a manner analogous to Bleaney et al. for  $Np^{237}$ .<sup>16</sup> From paramagnetic-resonance measurements, Bleaney shows that the magnetic moment of  $Np^{237}$  and the quadrupole coupling constant of  $Rb(NpO_2)(NO_3)_3$  must have opposite signs. Bleaney suggests that the magnetic moment,  $\mu$ , is positive and  $q$  is negative on theoretical grounds. Our value calculated for the  $l = 2$  wave out of phase with the  $l = 0$  is well within the limits of error of their measurement, and the value for the  $l = 2$  wave in phase with the  $l = 0$  seems to be outside the limits of error.

We will be able to make a definite phase choice only when more experimental data become available. Either a high-precision determination of the populations of the  $11/2$  and  $13/2$  levels of  $Th^{229}$  by alpha decay, or a measurement of the sign of

$$\left\langle \frac{\partial^2 v(0)}{\partial z^2} \right\rangle$$

will definitely determine the relative phases of the  $\ell = 0$  and the  $\ell = 2$  partial waves.

### IX. RESULTS OF THE NUMERICAL INTEGRATION OF $U^{233}$

The results of the numerical integrations of  $U^{233}$  may be expressed in several ways. In analogy with Fröman, we give matrices through which one may convert amplitudes of partial waves at the nuclear surface of amplitudes at infinity which are (intensity/velocity)<sup>1/2</sup>.

Let  $a_t$  be a column vector giving the amplitudes of partial waves at the nuclear surface, where  $t$  denotes indices  $\ell$  and  $I_f$ . We may relate this to a column vector  $b_t$ , which gives the amplitudes of the partial waves at infinity, by an equation of the form  $b_t = \sum_{t'} k_{t,t'} a_{t'}$ . We then factor  $k_{t,t'}$  into two matrices,

$$k_{t,t'} = \sum_{t''} [ G_{t''}^{-1} (R) \delta_{t,t''} ] k'_{t'',t'} \quad (16)$$

in the case of the real (irregular) components, and in the case of the imaginary parts,

$$k_{t,t'} = \sum_{t''} [ F_{t''}^{-1} (R) \delta_{t,t''} ] k'_{t'',t'} \quad (17)$$

In both cases the Coulomb functions are evaluated at the nuclear radius, in our work chosen to be  $9.0 \times 10^{-13}$  cm. The matrices  $k'_{t,t'}$  are similar to those given by Fröman<sup>9</sup> and by Rasmussen and Hansen<sup>11</sup> and are a convenient way of displaying the detailed effects of the quadrupole interaction. The matrices  $k'_{t,t'}$  become simple unit matrices  $\delta_{t,t'}$  in the limit of zero nuclear quadrupole moment. It should be pointed out that these matrices apply to a spherical surface at the nucleus whereas Fröman's matrices are given for a spheroidal nuclear surface.

From the imaginary part of the numerical integration we obtain the matrix  $k'_{t,t'}$  (Table VII).

Table VII

		$k'_{t,t'}$ Matrix From Imaginary Part of Numerical Integration							
		$l' = 0$		$l' = 2$		$l' = 2$		$l' = 2$	
		$I_f' = 5/2$		$I_f' = 5/2$		$I_f' = 7/2$		$I_f' = 9/2$	
$l = 0$	$I_f = 5/2$	1.000	-0.010i	0.150	-0.030i	-0.180	+0.043i	0.118	-0.030i
$l = 2$	$I_f = 5/2$	0.154i	-0.0195i	0.886	-0.010i	-0.111	+0.022i	-0.054	+0.0049i
$l = 2$	$I_f = 7/2$	-0.173	+0.027i	-0.106	+0.013i	0.849	-0.0353i	-0.110	+0.022i
$l = 2$	$I_f = 9/2$	0.096	-0.0067i	-0.021	+0.0024i	-0.099	+0.0076i	0.854	+0.0006i

Table VIII shows the matrix  $k'_{t,t'}$  obtained from the real part of the numerical integration.

Table VIII

		$k'_{t,t'}$ Matrix From Real Part of Numerical Integration							
		$l' = 0$		$l' = 2$		$l' = 2$		$l' = 2$	
		$I_f' = 5/2$		$I_f' = 5/2$		$I_f' = 7/2$		$I_f' = 9/2$	
$l = 0$	$I_f = 5/2$	1.000	+0.013i	-0.166	-0.016i	0.1953	+0.013i	-0.1024	-0.0037i
$l = 2$	$I_f = 5/2$	-0.1948	-0.022i	0.9053	+0.012i	0.116	+0.0068i	0.059	+0.0058i
$l = 2$	$I_f = 7/2$	0.242i	+0.022i	0.128	+0.0052i	1.007	-0.027i	0.126	+0.0005i
$l = 2$	$I_f = 9/2$	-0.143	-0.048i	0.057	+0.032i	0.093	+0.015i	0.911	+0.019i

Finally we give in Tables IX and X numerical values of the radial wave functions  $u_{I_f, \ell}$ , for several values of  $r$ .

#### X. SUMMARY

We believe this detailed numerical integration of the alpha-decay wave equation for  $U^{233}$  shows the essential validity of the Bohr-Fröman-Mottelson hypothesis (B.F.M.-1) that for favored alpha decay there is zero projection of alpha angular momentum on the nuclear-symmetry axis while the alpha is near the surface. The approximation (B.F.M.-2) that the projection remains zero near the classical turning point is shown to be a fairly good approximation for the relatively abundant  $\ell = 2$  wave but a very poor approximation for the weak  $\ell = 4$  wave. The analytical approximation based on modified Coulomb functions is shown to give results nearer those of the numerical integration than does the B.F.M.-2 approximation. The extra phase shifts due to the quadrupole interaction were derived, and the shifts most significant to the interpretation of nuclear-alignment experiments were shown to be negligibly small.

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Table IX

Real part of alpha wave functions for $U^{233}$ decay with $l = 0$ and $l = 2$ in phase				
$rx10^{-13}$ cm	$  5/2, 0 >$	$  5/2, 2 >$	$  7/2, 2 >$	$  9/2, 2 >$
9.0	1.52(17) <sup>a</sup>	1.26(17) <sup>a</sup>	-1.46(17) <sup>a</sup>	8.08(16) <sup>a</sup>
9.2	9.89(16)	8.23(16)	-9.47(16)	5.22(16)
9.6	4.28(16)	3.54(16)	-4.07(16)	2.24(16)
10.0	1.89(16)	1.55(16)	-1.79(16)	9.81(15)
10.4	8.55(15)	6.99(15)	-8.02(15)	4.39(15)
11.0	2.70(15)	2.18(15)	-2.51(15)	1.37(15)
12.0	4.32(14)	3.45(14)	-3.95(14)	2.14(14)
14.0	1.49(13)	1.16(13)	-1.32(13)	7.05(12)
16.0	7.08(11)	5.39(11)	-6.11(11)	3.23(11)
18.0	4.42(10)	3.30(10)	-3.72(10)	1.94(10)
20.0	3.48 (9)	2.55 (9)	-2.86 (9)	1.47 (9)
25.0	1.40 (7)	9.87 (6)	-1.09 (7)	5.42 (6)
30.0	1.51 (5)	1.03 (5)	-1.11 (5)	5.35 (5)
35.0	3.74 (3)	2.49 (3)	-2.60 (3)	1.20 (3)
40.0	1.98 (2)	1.29 (2)	-1.29 (2)	5.68 (1)
45.0	2.29 (1)	1.45 (1)	-1.38 (1)	5.61 (0)
50.0	6.51 (0)	3.99 (0)	-3.54 (0)	1.30 (0)
55.0	2.17 (0)	1.38 (0)	-1.36 (0)	5.39(-1)
60.0	-2.65 (0)	-1.51 (0)	-1.03 (0)	-2.25(-1)
65.0	-8.95(-1)	-6.58(-1)	8.61(-1)	-4.01(-1)
70.0	2.62 (0)	1.61 (0)	-1.40 (0)	4.39(-1)
75.0	-2.66 (0)	-1.55 (0)	1.09 (0)	-2.16(-1)
80.0	2.14 (0)	1.19 (0)	-6.18(-1)	2.31(-3)

<sup>a</sup>Number in parentheses indicates power of ten by which preceding number is to be multiplied.

Table X

Real part of alpha functions for $U^{233}$ decay with $l=0$ and $l=2$ out of phase				
$r \times 10^{-13}$ cm	$  5/2, 0 \rangle$	$  5/2, 2 \rangle$	$  7/2, 2 \rangle$	$  9/2, 2 \rangle$
9.0	5.65(17) <sup>a</sup>	-6.76(17) <sup>a</sup>	7.68(17) <sup>a</sup>	-3.92(17) <sup>a</sup>
9.2	3.81(17)	-4.45(17)	5.05(17)	-2.58(17)
9.6	1.75(17)	-1.95(17)	2.22(17)	-1.13(17)
10.0	8.16(16)	-8.78(16)	9.98(16)	-5.08(16)
10.4	3.87(16)	-4.02(16)	4.57(16)	-2.32(16)
11.0	1.30(16)	-1.30(16)	1.47(16)	-7.45(15)
12.0	2.26(15)	-2.12(15)	2.41(15)	-1.22(15)
14.0	8.79(13)	-7.58(13)	8.59(13)	-4.31(13)
16.0	4.55(12)	-3.71(12)	4.18(12)	-2.08(12)
18.0	3.02(11)	-2.36(11)	2.64(11)	-1.31(11)
20.0	2.48(10)	-1.88(10)	2.10(10)	-1.03(10)
25.0	1.08(8)	-7.67(7)	8.42(7)	-4.04(7)
30.0	1.22(6)	-8.33(5)	8.97(5)	-4.18(5)
35.0	3.13(4)	-2.07(4)	2.17(4)	-9.79(3)
40.0	1.70(3)	-1.09(3)	1.11(3)	-4.79(2)
45.0	2.01(2)	-1.26(2)	1.21(2)	-4.92(1)
50.0	5.83(1)	-3.56(1)	3.22(1)	-1.19(1)
55.0	1.70(1)	-1.10(1)	1.14(1)	-4.34(0)
60.0	-2.50(1)	1.42(1)	-1.01(1)	3.29(0)
65.0	-6.96(0)	5.22(0)	-7.30(0)	3.25(0)
70.0	2.35(1)	-1.44(1)	1.29(1)	-4.27(0)
75.0	-2.47(1)	1.43(1)	-1.05(1)	2.62(0)
80.0	2.05(1)	-1.14(1)	6.45(0)	-7.69(-1)

<sup>a</sup>Number in parentheses indicates power of ten by which preceding number is to be multiplied.

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