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# NUMERICAL SOLUTIONS OF THE CURIUM-242 ALPHA-DECAY WAVE FUNCTIONS

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October , 1957

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## ABSTRACT

Numerical integration of the previously derived alpha decay wave equation, including electric quadrupole coupling terms, was carried out for $Cm^{242}$ including $\ell = 0, 2, 4$, and 6 partial waves. Eight integrations were carried inward in spherical polar coordinates on an IBM-650 computer with different initial conditions, such that a complete set of eight linearly independent solutions to the system of coupled equations was generated. Eight different linear combinations of this base set were found which satisfy the boundary conditions imposed by experimental $Cm^{242}$ alpha group intensities. Wave amplitudes on a spherical surface near the nucleus are given for all eight cases, and the radial variation throughout the barrier region is given for two cases. The matrix formalism of Fröman is employed in another presentation of the results, and a comparison is made with the analogous Fröman matrix. By using a modified Fröman matrix together with our results, the alpha wave distributions are calculated for a spheroidal nuclear interaction surface. A discussion is made of conditions for the existence of solutions satisfying boundary conditions imposed by alpha group intensities, and the quadrupole phase shift problem is considered.

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## INTRODUCTION

For nuclei with mass numbers greater than ~220 there seems to be a stable spheroidal deformation, which gives rise to rotational bands and other phenomena treated in the Bohr-Mottelson model.[1] The even-even nuclei exhibit even-parity rotational bands based on the 0+ ground state and consisting of levels with 2+, 4+, 6+, etc. assignments. Successive levels are connected by especially large E2 transition matrix elements. It is these large electric quadrupole interactions that complicate the solution of the Schrödinger equation governing alpha decay. General treatments of the alpha-decay problem with noncentral interactions have been made by Preston[2] and by Perlman and Rasmussen.[3,4] More specialized treatment of the alpha decay of even-even spheroidal nuclei have been made by several authors,[5] and we shall not here repeat any detailed introduction to the spheroidal nuclear alpha-decay problem.

## THE WAVE EQUATION

It is convenient to consider the alpha-particle wave function with respect to a spherical polar coordinate system $(r,\theta)$ with polar axis the nuclear symmetry axis of the spheroidal nucleus. The azimuthal angle has no significance, since the only final nuclear states to be considered here have no angular-momentum component along the symmetry axis, i.e., $K = 0$. With the daughter nucleus possessing an intrinsic quadrupole moment, $Q_o$, and a finite rotational moment of inertia, $\mathfrak{J}$, the wave equation is not separable. If only the states of the ground rotational band are considered as final states, then we may express the alpha-wave function by the expansion

$$\psi(r,\theta,\varphi) = \sum_{0\leq\ell\leq N} r^{-1} w_\ell(r) \sqrt{\frac{2\ell+1}{4\pi}}\; P_\ell(\cos\theta), \tag{1}$$

where the $P_\ell$ are Legendre functions. In this paper we shall neglect the odd-$\ell$ terms, since the odd-spin members of the rotational band usually lie rather high in energy.

Using the expansion (1), one can reduce the three-dimensional wave equation to a set of coupled ordinary differential equations in the expansion functions $w_\ell(r)$. The first four equations have been explicitly given before,[6] and are as follows:

$$w_0'' - \left(\frac{4mZe^2}{\hbar^2 r} - \frac{2mE_0}{\hbar^2}\right) w_0 = \frac{2mQ_0e^2}{\hbar^2 r^3}\,\frac{w_2}{\sqrt{5}}, \tag{2a}$$

$$w_2'' - \left(\frac{4mZe^2}{\hbar^2 r} - \frac{2mE_2}{\hbar^2} + \frac{6}{r^2}\right) w_2 = \frac{2mQ_0e^2}{\hbar^2 r^3}\left(\frac{w_0}{\sqrt{5}} + \frac{2w_2}{7} + \frac{6w_4}{7\sqrt{5}}\right), \tag{2b}$$

$$w_4'' - \left(\frac{4mZe^2}{\hbar^2 r} - \frac{2mE_4}{\hbar^2} + \frac{20}{r^2}\right) w_4 = \frac{2mQ_0e^2}{\hbar^2 r^3}\left(\frac{6w_2}{7\sqrt{5}} + \frac{20w_4}{77} + \frac{15w_6}{11\sqrt{13}}\right), \tag{2c}$$

$$w_6'' - \left(\frac{4mZe^2}{\hbar^2 r} - \frac{2mE_6}{\hbar^2} + \frac{42}{r^2}\right) w_6 = \frac{2mQ_0e^2}{\hbar^2 r^3}\left(\frac{15}{11\sqrt{13}}\, w_4 + \frac{14w_6}{55}\right) + \ldots, \tag{2d}$$

where Z is the charge of the daughter nucleus, m is the reduced mass, $E_\ell$ is the total decay energy of the $\ell$-wave alpha, and $Q_0$ is the intrinsic nuclear quadrupole moment.

These equations are of a convenient form for numerical solution by digital computer methods. The results in this paper were obtained with the aid of an IBM-650 computer.

## BOUNDARY CONDITIONS

The solutions in this paper are those of the inward-integration type, where boundary conditions are taken from the experimental alpha-group intensities, and the final information sought is the wave function near the nuclear surface.

The solutions at large enough distance that the quadrupole terms become negligible will be taken as of the form of outgoing Coulomb waves,[7]

but with the phase factor $e^{i\delta_\ell}$ left to be determined:

$$w_\ell \longrightarrow A_\ell(G_\ell + iF_\ell)\, e^{i\delta_\ell}. \tag{3}$$

For the system of four second-order equations (2) it is necessary to impose eight boundary conditions. Four of these conditions come from the experimental intensities $I_\ell$.

$$v_\ell A_\ell^2 = I_\ell\ , \tag{4}$$

with $v_\ell$ the velocity of the $\ell$-wave particle at infinity.

Since the alpha particle behind the potential barrier exists in a quasi-stationary state, its wave function in the nuclear surface region may be considered almost purely real. Thus, the four additional boundary conditions can be obtained to good approximation by imposing the condition that the imaginary part of each partial wave vanish at the nuclear surface. In actual fact we have required the imaginary components to vanish on a spherical surface near the nucleus, but the real parts of the solutions finally obtained were found to be quite insensitive to the location of the surface on which these boundary conditions were imposed.

There is an ambiguity remaining in the specification of the solution, associated with the phase (or sign) of the partial waves (see Ref. 5).

## EXPERIMENTAL DATA

The following experimental values[8] from $Cm^{242}$ alpha decay were used:

| | |
|---|---|
| $E_o = 6.253$ Mev | $I_o = 73.7\%$ |
| $E_2 = E_o - 0.0441$ Mev | $I_2 = 26.3\%$ |
| $E_4 = E_o - 0.1460$ Mev | $I_4 = 0.035\%$ |
| $E_6 = E_o - 0.3037$ Mev | $I_6 = 0.006\%$ |
| | $Q_o = +9 \times 10^{-24}\ cm^2$ |

## NUMERICAL METHODS

The set of differential equations (2) was integrated by applying an iterative numerical method to a set of approximating difference equations. The arithmetic was performed on the University of California Radiation Laboratory digital IBM-650 computer. The basic equation used relates the (n-1)th second

difference $\Delta^2 y_{n-1}$ to the second derivative $y''_n$ (as obtained from the appropriate Eqs. (2)) and its second difference $\Delta^2 y''_{n-1}$:

$$\Delta^2 y_{n-1} = h^2[y''_n + (1/12)\,\Delta^2 y''_{n-1}], \tag{5}$$

where $\Delta^2 y_{n-1}$ is defined as $\Delta^2 y_{n-1} = y_{n+1} - 2y_n + y_{n-1}$, and where h is the interval in the independent variable, $\rho$. This equation requires three starting values at equally spaced intervals of $\rho$. In this section we make frequent use of the dimensionless distance parameter, $\rho$, which is equal to the product of distance and $k_o$, the wave number of the ground-state alpha group at infinity. For the $Cm^{242}$ problem we have $k_o = 1.08508 \times 10^{13}$ $cm^{-1}$. Hence, one unit in $\rho$ is equivalent to a distance of 0.921 fermi ($10^{-13}$ cm).

It was hoped that inward integration from $\rho = 75$ would yield solutions accurate to the order of a percent near the nuclear surface. The solutions on which the final results are based were obtained with an interval of $h = 0.5$ (units of $\rho$). This interval is coarser than desirable near the nucleus, as evidenced by the size of the first error term of difference formula (5), namely $(1/240)\,\Delta^6 y_n$. However, integrations on two solutions at an interval of $h = 0.1$ showed only a 1% difference at $\rho = 14.5$ from the values of the functions obtained by use of the coarser interval. All four components of the solution for the coarse interval were larger by 1% than the $h = 0.1$ values; at $\rho = 11.5$ the errors were between 1 and 2%. The relative values of the four components are not much altered by integration with the finer interval. Therefore, it was felt unnecessary to rerun all the other linearly independent solutions at smaller interval.

Eight separate inward integrations of the four coupled second-order linear differential equations from a large distance were run with different initial conditions. Eight linearly independent solutions were obtained, constituting a complete set. Any general solution is expressible as a linear combination of these solutions. The boundary conditions consisted in setting the amplitude and first derivative of one partial wave equal to the corresponding regular ($F_i$) or irregular ($G_i$) Coulomb function (calculated by the Riccati II approximation formulas) (Ref. Fröberg),[7] with all other partial-wave amplitudes and first derivatives equal to zero. The matching to pure Coulomb functions at $\rho = 75$ amounts to solution of an approximate set of equations of the form of (2) at distances $\rho < 75$, but where all the quadrupole potential terms are "turned off" at distances greater than $\rho = 75$.

Pennington[9] has developed a perturbation method to treat the coupling effects on out to infinity. His work involves numerical integration of the alpha wave equation in spheroidal coordinates for many heavy element nuclei taking into account $\ell = 0$, 2, and 4 groups only. His general approach in constructing solutions from linear combinations of a basic linearly independent set is similar to ours.

From these eight linearly independent solutions of Eqs. (2) as base vectors, the complex solutions that satisfied the asymptotic boundary conditions (4) and whose imaginary parts vanished on a sphere of radius $\rho = 12$ were found algebraically. The solutions of eight simultaneous algebraic equations, four of them quadratic, were required, and these solutions were obtained by use of the IBM-650 computer.

As an example of the extent to which quadrupole terms affect the base vector solutions, the base vector derived by setting $w_i = \delta_{io} G_o$ at $\rho = 75$ has the following values at $\rho = 15$:

$$w_0 = 1.646\text{x}10^{10},\ w_2 = 2.420\text{x}10^{9},\ w_4 = 1.338\text{x}10^{8},\ \text{and}\ w_6 = 3.607\text{x}10^{6}.$$

In the absence of quadrupole terms $w_2$, $w_4$, and $w_6$ would remain identically zero for all $\rho$.

As an example of a solution (linear combination of base vectors) satisfying the intensity boundary conditions, we give at $\rho = 15$ the partial wave amplitudes of one of the eight solutions:

$$w_0 = 1.831\text{x}10^{10},\ w_2 = 1.891\text{x}10^{10},\ w_4 = 3.837\text{x}10^{9},\ \text{and}\ w_6 = 3.629\text{x}10^{9}.$$

This solution (for all $\rho$) is formed from the eight base vectors by expansion coefficients of 1.000, 0.5973, 0.01709, and 0.009112 for the $G_o \neq 0$, $G_2 \neq 0$, etc. vectors and -0.009372, -0.03615, -0.01373, and -0.0006825 for the $F_i \neq 0$ vectors. With these expansion coefficients the real part of the general outgoing wave solution is obtained. By exchanging corresponding expansion coefficients for $G_i \neq 0$ and $F_i \neq 0$ solutions and changing the sign of one, the imaginary part of the general solution is constructed, but it is not expected to be accurate within the barrier when constructed from inward-integrated solutions, since it will involve small differences of large numbers.

## RESULTS

There are several ways in which the numerical results may be presented. First, we give partial wave amplitudes (real part) as a function of distance for two of the eight cases satisfying the empirical $Cm^{242}$ alpha group intensities. These solutions are given in Table I, and the normalization is such that the S-wave goes over asymptotically into an oscillatory function with unit amplitude.

Second, there is presented in Table II the relative wave amplitude values on the spherical surface ( $\rho = 11.0$; i.e. $r = 10.1 \times 10^{-13}$ cm). For comparison are given the relative values that would be calculated if the quadrupole moment were zero (column 4) and the values that would be calculated if both the quadrupole moment and centrifugal barrier effects were ignored (column 3). In comparing column 4 with the eight numerical cases it is noted that inclusion of quadrupole terms seriously affects only the $\ell = 4$ partial wave. This result is readily understandable from the structure of the coupled Eqs. (2a,b,c,d), where coupling terms will have greatest effect on a partial wave of small amplitude coupled to one of large amplitude.

Examination of column 4 suggests that the Legendre expansion is not converging rapidly and that the $\ell = 8$ group and possibly higher rotational groups are not negligible.

### Comparison with Fröman's Treatment

Fröman[10] in a recent comprehensive treatment of alpha decay has derived analytical expressions for alpha decay of spheroidal nuclei. With these expressions he has calculated numerical values for matrix elements of a matrix $k_{\ell\ell'}(B)$ which by multiplication carries over the Legendre expansion coefficients, $a_\ell$, of the alpha wave function on the spheroidal nuclear surface into coefficients, $b_\ell$, related to the alpha group intensities, i.e. $b_\ell = \sum_{\ell'} k_{\ell\ell'}(B) a_{\ell'}$. $|b_\ell|$ is the reciprocal of the product of the hindrance factor and the centrifugal barrier reduction factor.

Table I. Alpha wave solutions for $Cm^{242}$ decay

| $\rho$ $(=k_o r)^a$ | Case 1 | | | | Case 2 | | | |
|---|---|---|---|---|---|---|---|---|
| | $w_0$ | $w_2$ | $w_4$ | $w_6$ | $w_0$ | $w_2$ | $w_4$ | $w_6$ |
| 11 | 1.18(13)[b] | 1.34(13) | 3.76(12) | 3.42(12) | 1.18(13) | 1.33(13) | 3.13(12) | -2.40(12) |
| 11.5 | 4.82(12) | 5.41(12) | 1.45(12) | 1.33(12) | 4.82(12) | 5.40(12) | 1.22(12) | -9.52(11) |
| 12 | 2.03(12) | 2.25(12) | 5.76(11) | 5.31(11) | 2.03(12) | 2.24(12) | 4.88(11) | -3.88(11) |
| 12.5 | 8.78(11) | 9.59(11) | 2.35(11) | 2.18(11) | 8.78(11) | 9.57(11) | 2.01(11) | -1.62(11) |
| 13 | 3.88(11) | 4.19(11) | 9.87(10) | 9.18(10) | 3.88(11) | 4.18(11) | 8.47(10) | -6.93(10) |
| 15 | 1.83(10) | 1.89(10) | 3.84 (9) | 3.63 (9) | 1.83(10) | 1.89(10) | 3.37 (9) | -2.88 (9) |
| 20 | 2.92 (7) | 2.76 (7) | 4.14 (6) | 3.99 (6) | 2.92 (7) | 2.76 (7) | 3.79 (6) | -3.41 (6) |
| 25 | 1.70 (5) | 1.51 (5) | 1.76 (4) | 1.68 (4) | 1.70 (5) | 1.51 (5) | 1.65 (4) | -1.50 (4) |
| 30 | 2.78 (3) | 2.31 (3) | 2.13 (2) | 2.01 (2) | 2.78 (3) | 2.31 (3) | 2.04 (2) | -1.81 (2) |
| 35 | 1.11 (2) | 8.65 (1) | 6.31 (0) | 5.64 (0) | 1.11 (2) | 8.65 (1) | 6.10 (0) | -5.29 (0) |
| 40 | 1.09 (1) | 7.83 (0) | 4.25(-1) | 3.60(-1) | 1.09 (1) | 7.83 (0) | 4.16(-1) | -3.44(-1) |
| 45 | 2.98 (0) | 1.95 (0) | 7.08(-2) | 5.53(-2) | 2.98 (0) | 1.95 (0) | 7.01(-2) | -5.41(-2) |
| 50 | 7.22(-1) | 5.97(-1) | 3.15(-2) | 2.03(-2) | 7.22(-1) | 5.97(-1) | 3.13(-2) | -2.01(-2) |
| 55 | -1.54 (0) | -8.45(-1) | -3.81(-3) | -7.25(-4) | -1.54 (0) | -8.45(-1) | -4.22(-3) | -1.32(-3) |

a. $k_o = 1.0850_8 \times 10^{13}\ cm^{-1}$.

b. Number in parenthesis is power of ten multiplying preceding number.

Table II. Theoretical partial wave amplitudes near the nucleus showing effects of quadrupole coupling ($Cm^{242}$)

| Final state E(kev) | Iπ | Relative wave amplitude assigning $Q_o$=0 and L=0(abs. value) $\sqrt{C_\ell}$ in Fröman's[10] notation | Relative wave amplitude with L effects but Q=0 (abs. value) $b_\ell$ in Fröman's notation | Partial-wave ratios (complete calculation) (at ρ = 11) 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 0 | 0+ | (1.0) | (1.0) | (1.0) | (1.0) | (1.0) | (1.0) | (1.0) | (1.0) | (1.0) | (1.0) |
| 44 | 2+ | 0.77 | 0.97 | 1.14 | 1.14 | 1.11 | 1.11 | -1.08 | -1.05 | -1.04 | -1.07 |
| 146 | 4+ | 0.051 | $0.11_2$ | 0.32 | 0.27 | 0.102 | 0.159 | -0.40 | -0.087 | -0.17 | -0.32 |
| 304 | 6+ | 0.053 | 0.29 | 0.29 | -.204 | -.24 | .25 | -0.41 | +0.35 | -0.36 | +0.29 |
| 514 | 8+ | 0.014 | 0.26 | | | | | | | | |
| 605 | 1- | 0.045 | 0.049 | | | | | | | | |
| 935 | 0+ | 0.22 | 0.22 | | | | | | | | |
| 1030 | 2+(K=2) | 0.15 | 0.19 | | | | | | | | |
| Magnitude of $w_o$ ($\times 10^{-12}$) (at ρ = 11) | | | | 11.76 | 11.75 | 11.74 | 11.74 | 8.17 | 8.19 | 8.19 | 8.17 |

The argument B is a function of the deformation of the surface and the intrinsic quadrupole moment. Fröman's matrix $k_{\ell,\ell'}(B)$ might be equated to a product of two similar matrices. By multiplication one carries the spheroidal expansion vector $a_\ell$ over to a vector $a'_\ell$ giving expansion coefficients on a spherical surface of the same mean radius as the spheroid. The second matrix carries the $a'_\ell$ vector over to the asymptotic vector $b_\ell$. The off-diagonal elements of the first matrix arise as a consequence of the transformation between two different surfaces, and those in the second arise as a consequence of the electric quadrupole interaction. Numerical values for each of these matrices may be taken from the values on page 41 of Fröman's paper but using arguments $B_1$ and $B_2$ calculated in the following manner. $B_1$ for the former matrix is calculated from Fröman's (VI-9) by setting $q_o$ equal to zero. $B_2$ is calculated by use of (VI-9) by leaving out the term unity in the final factor and inserting the value of $q_o$ defined by Fröman's (VI-2).

It is this second matrix which may be directly derived from the numerical wave functions of the present work. Linear combinations of our eight linearly independent solutions are found which represent the irregular Coulomb function of a single partial wave at $\rho = 11$. The expansion coefficients are found by solution of eight simultaneous linear equations, and these expansion coefficients, when renormalized to account for different penetrabilities of the different partial waves due to the centrifugal potential and to nuclear rotational energies, constitute the matrix elements of the second matrix described in the preceding paragraph. Unlike the matrix derived by Fröman's approximation our matrix is not symmetric and the matrix elements are generally complex. This complex nature reflects the phase shifts on the Coulomb waves arising from quadrupole coupling terms in the wave equation.

The matrix applies to the surface of radius $r = 10.12 \times 10^{-13}$ cm ($\rho = 11.0$), which is representative of alpha particle interaction radii and would satisfy the expression:

$$r = (1.23\ A^{1/3} + 2.5) \times 10^{-13}\ \text{cm.}$$

Our matrix is then as follows:

Quadrupole interaction matrix

| $\ell = 0$ | $\ell = 2$ | $\ell = 4$ | $\ell = 6$ |
|---|---|---|---|
| 1.015+0.0116i | -0.1674-0.0176i | 0.01166+0.00217i | -0.0005093-0.000130i |
| -0.2107-0.0456i | 0.9542-0.00158i | -0.1195-0.00592i | 0.007260+0.000679i |
| 0.02114+0.0135i | -0.1899-0.0595i | 0.9191-0.00360i | -0.1008-0.00187i |
| -0.001089-0.00216i | 0.01885+0.0187i | -0.2052-0.0893i | 0.9086-0.0241i |

For the parameters $r = 10.12 \times 10^{-13}$ cm and $Q_o = 9.0 \times 10^{-24}$ cm$^2$ of our matrix one may calculate that the appropriate argument of the analogous Fröman matrix is B = -0.455. The matrix resulting from interpolation (7 point) of Fröman's numerical matrix elements for this argument is given below for comparison with the preceding matrix.

Fröman matrix for B = -0.455

| $\ell = 0$ | $\ell = 2$ | $\ell = 4$ | $\ell = 6$ |
|---|---|---|---|
| 1.019 | -0.193 | 0.014 | -0.0005 |
| -0.193 | 0.908 | -0.158 | 0.014 |
| 0.014 | -0.158 | 0.917 | -0.155 |
| -0.0005 | 0.014 | -0.155 | 0.917 |

To facilitate comparison the overall normalization of our complex matrix was carried out to make the $\ell = \ell' = 0$ diagonal components nearly equal. The normalization to 1.015, compared to Fröman's 1.019, was arbitrarily made slightly less from qualitative consideration of the sign of the expected deviation from the Fröman approximation. Comparing the two matrices one sees a fairly close correspondence of the real components. The imaginary components are usually considerably smaller than the real components except for elements

far from the diagonal. The asymmetry of the matrix is of the sense that an off-diagonal element in the upper-right-hand half is always smaller than its counterpart in the lower-left-hand half.

In the Fröman approximation the quadrupole coupling terms (off-diagonal terms) in the alpha decay wave equation are considered to operate only over the inner part of the barrier and the diagonal terms for each partial wave in this region are considered the same; the effects of the difference in diagonal terms (centrifugal potential and nuclear rotational energy) are considered to arise from the outer region of the barrier. The asymmetry of the matrix derived from our numerical solutions may be said to arise as a consequence of the operation of off-diagonal terms over the same radial region where diagonal terms significantly differ for the different partial waves. The imaginary components of the matrix elements arise principally as a consequence of quadrupole terms, diagonal and off-diagonal, in the region of the classical turning points. Fröman's approximation neglects quadrupole terms in the turning point region.

## Wave Functions on the Spheroidal Nuclear Surface

Since the nuclear surface of the alpha decay daughter of $Cm^{242}$ is supposedly spheroidal, it is of interest to determine Legendre expansion coefficients for the wave function on the spheroidal surface. In principle we could take a spheroidal section through the numerical wave functions defined by our spherical expansion coefficients, but this procedure is not very practicable. Instead, we chose to multiply our relative spherical expansion coefficient vectors (Table II) by a modified Fröman inverse matrix. The deformation of the spheroid of mean radius $\rho = 11.0$ was chosen from calculation of a nuclear matter spheroidal surface for mean radius $r = 1.23 \times 10^{-13} A^{1/3}$ cm and $Q_o = +9.0 \times 10^{-24}$ $cm^2$ and then adding a constant distance $2.5 \times 10^{-13}$ cm to take into account the finite range of the nuclear interaction and "radius of the alpha particle." This procedure led to a deformation parameter $\beta_2$ of +0.159. Using the modification of Fröman's Eq. (VI-9) described in the preceding section we then calculated the argument $B = +2.00$ for the Fröman matrix which by matrix multiplication transforms the distribution vector on the

spheroidal surface into the corresponding vector on the spherical surface of the same mean radius. We desire the inverse matrix to convert to the spheroidal distribution vector, and this matrix is just the Fröman matrix with argument B = -2.00. Multiplying the spherical surface amplitudes of cases 1 to 4 from Table II we get the spheroidal distribution vectors given in Table III. The transformation will give rise to components of $\ell = 8$ and higher, but these are not listed in Table III, since with the arbitrary exclusion of $\ell = 8$ partial waves from our work, the proper values of the $\ell = 8$ coefficients are quite uncertain. Cases 5 to 8 were not transformed, since indirect evidence discussed later gives information on the probable $\ell = 0$, $\ell = 2$ phase difference which excludes cases 5 to 8 as actual possibilities.

Table III. Expansion coefficients of wave functions on the nuclear surface

| | Case | | | |
|---|---|---|---|---|
| $\ell$ | I | II | III | IV |
| 0 | 0.482 | 0.5022 | 0.483 | 0.465 |
| 2 | 0.246 | 0.1723 | 0.235 | 0.303 |
| 4 | -0.273 | -0.0257 | -0.152 | -0.390 |
| 6 | 0.264 | -0.1950 | -0.136 | 0.315 |

The wave function variations over the spheroid obtained with the expansion coefficients of Table III fall into two groups. Cases 1 and 4 are quite similar to one another, showing maxima of roughly equal height at $\theta = 0^\circ$ and $\sim 55^\circ$ and changing sign near $90^\circ$. Cases 2 and 3 are similar to one another and show a main peak around $30\text{-}35^\circ$ with a minimum at $0^\circ$. Solutions 2 and 3 also both show slight peaking at $90^\circ$ with minima at $60^\circ$ and $75^\circ$, respectively. Inspection of column 4 of Table II leads us to believe that inclusion of the $\ell = 8$ partial wave could significantly alter the details of the distributions and, of course, would lead to twice as many possible cases. The distributions of cases 2 and 3 bear a strong qualitative similarity to the distribution (case I) derived by Rasmussen and Segall,[5] except that the single maximum in this earlier work lay near $50^\circ$. The differences from this earlier work are due to inclusion

in the present work of the $\ell \neq 6$ wave and to use of a deformation about half as great as was used before. The inclusion of the $\ell = 6$ wave has given rise to the two distinct alternative patterns within cases 1 to 4.

Let us now turn to the question of deciding on the physically correct case of the eight possible solutions. Angular correlation experiments with alpha particles may give results dependent on phase differences. The different cases correspond to different phase differences between the various alpha groups, but since the various groups in even-even alpha emission are all of different energy, there can be no interference terms in angular distribution experiments. However, in the alpha decay of odd-mass nuclei there may be more than one partial wave in single alpha groups, and there may thus be interference terms in angular distribution experiments. From the sign of the interference terms one may decide between the two possible phase differences of two interfering partial waves.

Bohr, Fröman, and Mottelson[11] have shown good correlations between relative alpha-group intensities in even-even nuclei and in "favored" decay groups in odd-mass nuclei. (See also Ref. 3 for discussion of favored alpha decay.) We may make further use of the analogy by proposing that the phase differences found in angular distribution experiments on favored alpha groups in odd-mass nuclei should be the same as in the neighboring even-even nuclei.

The two best and most readily interpretable experiments are the alpha 60-kev gamma angular correlation in $Am^{241}$ by Krohn, Novey, and Raboy[12] and the alpha angular distribution from aligned $U^{233}$ nuclei by Dabbs, Roberts, and Parker.[13] The main part of the anisotropy in each experiment is thought to arise from an S-wave D-wave interference term in the main alpha group ($\Delta I = 0$). The $Am^{241}$ result can only be interpreted as indicating the correctness of one of cases 1 to 4 and rejecting cases 5 to 8 (i.e., there is a constructive S-D interference along the nuclear symmetry axis). From the lack of any sharp variation in $\ell = 2$ group-hindrance factors for even-even alpha emitters between mass 233 and 241 we should expect the S-D phase difference to be of the same sense for $U^{233}$ as for $Am^{241}$. The theoretical interpretation of the $U^{233}$ anisotropy can give information on the S-D phase if the sign of the quadrupole coupling constant for uranium in rubidium uranyl nitrate is known. We would like to suggest that the sign of the quadrupole coupling constant in

rubidium uranyl nitrate is probably negative. In this crystal the linear uranyl ($UO_2^{++}$) ions are aligned along the c-axis of the crystal. Eisenstein and Pryce[14] have proposed that a strong hybridization of atomic orbitals 6d and 7s in uranium occurs for the σ-bonding orbitals, and they state that this hybridization will give rise to a "column of charge" along the uranyl bond axis. If this axial column of negative charge characterized the bonding orbitals at all distances from the uranium nucleus, then the positive quadrupole moment of the nucleus would give rise to a positive quadrupole coupling constant, i.e., magnetic substates $m_I = \pm I = \pm 5/2$ would lie lowest in energy. However, the 6d and 7s orbitals have three and six radial nodes, respectively, and a hybrid orbital will thus exhibit alternating concentrations of charge at the poles and in equatorial rings as the radial distance changes. The quadrupole coupling constant will be most dependent on the electronic charge distribution near the nucleus. For the bonding 6d-7s hybrid the innermost interference region will have an equatorial ring of charge. An evaluation of the radial integral for the 6d-7s interference term in the quadrupole coupling expression, using unscreened, nonrelativistic electron wave functions, and also using screened relativistic wave functions,[15] did yield a negative contribution to the coupling constant.[16] The 6d component alone yields a positive contribution, so the sign of the quadrupole coupling constant will depend on the degree of s-d hybridization. If, as we believe, it is negative, the $Am^{241}$ and $U^{233}$ experiments are consistent with each other and support the choice of one of our cases 1 to 4.

Deciding among the cases 1 to 4 is not possible with present information. In principle the needed information can be obtained from a study of the D-G and G-I interference terms in alpha angular-distribution experiments. In practice the information will be difficult to obtain because of the relative weakness of G and I waves for favored alpha transitions.

Let us summarize the method of transforming the vector describing the alpha particle wave function on the spheroidal nuclear surface to the vector $b_\ell$ whose components are partial wave amplitudes ($w_\ell = r\psi_\ell$) at infinity. One multiplies successively by the modified Fröman matrix (whose argument B is calculated setting $q_o$ to zero), by our complex quadrupole interaction matrix

given earlier in this section, and finally by a diagonal matrix $k_{\ell\ell'} = G_\ell^{-1}(kr)\,\delta_{\ell\ell'}$, where r is the mean radius of the nuclear spheroidal surface, and $G_\ell$ is the irregular Coulomb function.

## CONSIDERATIONS REGARDING EXISTENCE OF SOLUTIONS TO THE COUPLED ALPHA-DECAY EQUATIONS

If there are no noncentral interactions, the wave equation separates exactly, and satisfaction of the boundary condition that imaginary components of all partial waves vanish at the nuclear surface can be achieved for any choice of relative group intensities simply by adjusting the phase of each partial wave separately. However, for the coupled equations here it seems on brief consideration that some combinations of relative intensities may be mathematically excluded by the nonexistence of solutions satisfying all the boundary conditions. For example, suppose we seek solutions to Eq. (2) satisfying both the nuclear and the asymptotic boundary conditions of Eq. (4) with all intensities vanishing except for the $\ell = 0$ group. Thus, only two of our eight base vector solutions (i.e., those derived from $G_o \neq 0$ and from $F_o \neq 0$ at large distance) can be used. Yet at the nuclear surface, because of coupling, these base vectors will have components in $w_2$, $w_4$, $w_6$, etc. as well as in $w_0$. It does not seem likely that the imaginary parts of all these components can be made to vanish by any linear combination of just two base vector solutions.

An example of forbidden intensity combinations was furnished when we attempted to calculate solutions with our $Cm^{242}$ base vectors as if $Cm^{242}$ had the hindrance factors of ionium ($Th^{230}$), which experimentally are 1.1 ($\ell = 2$), 12 ($\ell = 4$), and 8200 ($\ell = 6$). No solutions satisfying the boundary conditions existed, but when a larger intensity of the $\ell = 6$ group was tried (hindrance factor 820), all eight solutions were found.

In actual fact the quadrupole moment appropriate for $Th^{230}$ decay is surely considerably less than that for $Cm^{242}$ decay, and with lower quadrupole coupling we know that the existence conditions would impose less restrictive limits on the intensities of weak groups. Surely the 8200 hindrance factor for $\ell = 6$ would give allowed solutions if the $Th^{230}$ problem were solved properly with a small $Q_o$. In a reverse application of the existence conditions

one could, for a given set of experimental hindrance factors, set an upper limit on $Q_o$ as that value above which solutions to the problem vanish.

## CONCERNING PHASE SHIFTS DUE TO QUADRUPOLE COUPLING

Aside from the importance of including quadrupole coupling to derive alpha surface distributions from relative intensity data, quadrupole coupling may be of some importance in affecting the interpretation of alpha angular-distribution experiments with spheroidal nuclei. The phase of alpha waves cannot affect any experimental measurements on the even-even alpha emitters, since each alpha group has only one associated angular momentum (consequence of zero spin of parent). However, odd nuclei may be expected often to have two or more partial waves associated with an individual alpha group. This mixture can give rise to interference terms in the alpha angular distributions, and the expressions for these interference terms involve the phases through a cosine factor having as argument the phase difference of the two partial waves. The phase difference has usually been calculated from the difference of arguments in the general asymptotic expression for outgoing Coulomb waves,

$$G_\ell + iF_\ell \longrightarrow \exp[i(\rho - \eta \log 2\rho - \frac{\ell\pi}{2} + \sigma_\ell)], \qquad (6)$$

where $\sigma_\ell = \arg \Gamma (i\eta + \ell + 1)$ and $\eta$ and $\rho$ are the standard Coulomb function arguments. Where noncentral interactions operate on the alpha particle after it leaves the nuclear surface, additional phase shifts may be introduced. To determine precisely how much additional shift would require study of the particular cases; however, we may get a rough estimate of the magnitude of phase shift effects to be expected in favored decay of odd-mass neighbors of $Cm^{242}$ by examining the phase shifts in the $Cm^{242}$ solutions discussed above.

At the end of the section on "Numerical Methods" the eight expansion coefficients for the case I $Cm^{242}$ solution were given. The extra phase shifts due to quadrupole coupling are easily calculated by simply taking the arc tangent of the ratio of the expansion coefficients for corresponding $F_i \neq 0$ and $G_i \neq 0$ solutions. The extra phase shifts (in radians) are $\delta_o = -0.009$, $\delta_2 = -0.060$, $\delta_4 = -0.68$, and $\delta_6 = -0.075$. As a first approximation we might take these phase shifts for analysis of angular correlation data involving

favored alpha-decay groups of odd-mass neighbors of $Cm^{242}$. From the asymptotic expression (6) one can derive the phase-difference formula given by Devons and Goldfarb,[17] and according to this formula the D-wave lags the S-wave by $7.3^{\circ}$ at infinity. When the additional phase lags due to the quadrupole interaction are taken into account, the D-wave may lag the S-wave by $10.2^{\circ}$ at infinity. The size of the theoretical S-D interference term should thus be reduced by about one percent from the values previously derived, neglecting quadrupole coupling. This correction does not seem very serious.

If one should perform an experiment in which the D-G interference term enters, then a more serious correction due to quadrupole phase shifts is to be expected, for the estimates above indicate an additional $36^{\circ}$ lag between D- and G-waves. In the $Cm^{242}$ problem the G-wave is subject to the greatest phase shifts, since it is a weak group coupled to a relatively intense $\ell = 2$ group. It should be emphasized that the above numerical values of the additional phase shifts apply only to case I. The phase shifts will be different for the different possible cases.

We can expect, regarding the noncentral interaction phase shifts, that (a) only the large collective E2 couplings will be of significance; (b) the shifts will be larger, the larger the intrinsic quadrupole moment; (c) the shifts will be largest for low-intensity groups coupled to high-intensity groups; and (d) the coupling may introduce phase shifts of either sign depending on the relative signs of the coupled partial waves in the classical turning-point region.

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   With this general comment we also wish to point out an error in the general formulas for the noncentral interaction as given in Ref. 3. The quantity $\lambda!$ should be deleted from Eqs. (29.3), (29.7), (29.8), and (29.14).
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