## Beta-Gamma Directional Correlations in Re<sup>186</sup> and Tm<sup>170+</sup>

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The beta-gamma directional correlations have been measured for the (934-keV beta) (137-keV gamma) cascade in Re<sup>186</sup> and the (883-keV beta) (84-keV gamma) cascade in Tm<sup>170</sup>. The coefficient  $A_2$  varies from 0.035 at a beta energy of 220 keV to 0.095 at a beta energy of 830 keV in the case of  $Re^{186}$  and varies from -0.055 at a beta energy of 330 keV to -0.090 at a beta energy of 740 keV in the case of Tm<sup>170</sup>. Interpretation of the decays in terms of limitations on the first forbidden beta decay matrix elements is discussed. For Re<sup>186</sup> the directional correlation and the spectral shape correction can be fitted for values of the parameter [ $\zeta_1$ ] greater than 2. Rough agreement with the measurement of the beta-circularly polarized gamma correlation is also found. For Tm<sup>170</sup> the directional correlation and an allowed spectral shape can be fitted for  $\zeta_1 < -2$  and  $\zeta_1 > 20$ .

#### INTRODUCTION

HE decay scheme of Re<sup>186</sup> is shown in Fig. 1.<sup>1</sup> The 934-keV beta transition with a  $\log ft$  product of 8.0 is interpreted as being first forbidden with a spin change of 1. Porter  $et \ al.^2$  observed a spectrum shape correction factor that increases about 18% from 150 to 900 keV. This indicates the 934-keV beta group has a spectrum shape intermediate between the allowed and unique shape.

The decay scheme of Tm<sup>170</sup> is shown in Fig. 2.<sup>3</sup> The 883-keV beta transition has a  $\log ft$  product of 9.3 and is believed to be first forbidden with spin change 1. Several investigators<sup>4-6</sup> have concluded that the spectrum for the 883-keV beta group has the allowed shape.

Preliminary results for the (934-keV beta) (137-keV gamma) directional correlation in Re<sup>186</sup> and the (883 keV beta) (84 keV gamma) directional correlation in Tm<sup>170</sup> have been previously reported.<sup>7,8</sup> In this report the final interpretation of the data for these two transitions is presented.

For both of these transitions the spin sequence is  $1^{-}(\beta)2^{+}(\gamma)0^{+}$ . In each case comparison with theory has yielded ranges for the nuclear matrix element ratios which produce a simultaneous fit of the directional correlation results and the experimental shape factor.

The (934-keV beta) (137-keV gamma) directional correlation in Re<sup>186</sup> has been previously determined by Novey et al.<sup>9</sup> The (883-keV beta) (84-keV gamma)

25, D. C.).
 <sup>2</sup> F. T. Porter, M. S. Freedman, T. B. Novey, and F. Wagner, Jr., Phys. Rev. 103, 921 (1956).
 <sup>3</sup> Nuclear Deta Starts Not! Acad Sci. Not! Res. Council.

<sup>3</sup>Nuclear Data Sheets, Natl. Acad. Sci.–Natl. Res. Council, NRC 60-1-80 (Office of Printing and Publishing, National Re-search Council–National Academy of Sciences, Washington <sup>4</sup> R. Richmond and H. Rose, Phil. Mag. 43, 367 (1952).
<sup>5</sup> A. V. Pohm and W. E. Lewis, Phys. Rev. 95, 1523 (1954).
<sup>6</sup> G. Bertolini, E. Lazzarini, and M. Mandelli-Bettoni, Nuovo

Cimento 6, 1106 (1957). <sup>7</sup>L. D. Wyly, E. T. Patronis, Jr., C. H. Braden, and Harry Dulaney, Bull. Am. Phys. Soc. 6, 451 (1961).

<sup>8</sup> L. D. Wyly, Harry Dulaney, and C. H. Braden, Bull. Am.

Phys. Soc. 7, 353 (1962). <sup>9</sup> T. B. Novey, M. S. Freedman, F. T. Porter, and F. Wagner, Jr., Phys. Rev. 103, 942 (1956).

directional correlation in Tm<sup>170</sup> has been measured by several investigators<sup>6,10</sup> and the interpretation discussed.<sup>11</sup> However, several subsequent developments in the field of first forbidden beta decay make a reconsideration of these two beta transitions desirable. In the interpretation of previous work on these transitions the  $B_{ij}$  matrix element has been assumed zero except that Novey et al.9 considered the effect of a small admixture of the  $B_{ij}$  matrix element. Recent experiments<sup>12</sup> indicate that in many cases the  $B_{ij}$  matrix element may be large compared to the other matrix elements. Also the use of Coulomb correction factors by Kotani<sup>13,14</sup> introduces greater accuracy into the theoretical expressions by eliminating the commonly used approximation  $(\alpha Z)^2 \ll 1$ .

In the present work the theory of first-forbidden beta decay as formulated by Kotani<sup>14</sup> is used. In the case of spin change 1, the theoretical expressions for the various observables such as the beta spectrum shape factor and the beta-gamma directional correlation are given by



Fig. 1. Decay scheme of Re<sup>186.1</sup> The electron capture branch ( $\approx 4\%$ ) has been omitted.

<sup>10</sup> T. B. Novey, Phys. Rev. 78, 66 (1950); H. Rose, Phil. Mag<sup>•</sup> 43, 1146 (1952)

<sup>11</sup> J. Fujita, M. Morita and M. Yamada, Progr. Theoret. Phys. (Kyoto) 11, 219 (1954).
 <sup>12</sup> Harry Dulaney, Jr., C. H. Braden, and L. D. Wyly, Phys. Rev. 117, 1092 (1960); H. J. Fischbeck and R. G. Wilkinson, *ibid*. 120, 1762 (1960); P. M. Alexander and R. M. Steffen, *ibid*. 124, 155 (1961).

150 (1961).

<sup>13</sup> T. Kotani and M. Ross, Phys. Rev. 113, 622 (1959).
 <sup>14</sup> T. Kotani, Phys. Rev. 114, 795 (1959).

<sup>†</sup> Supported in part by a grant from the National Science Foundation (G6337).

<sup>&</sup>lt;sup>1</sup>Nuclear Data Sheets, Natl. Acad. Sci.-Natl. Res. Council, NRC 59-5-136 (Office of Printing and Publishing, National Research Council-National Academy of Sciences, Washington

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302

332

368

415



Kotani in terms of the following nuclear matrix element parameters:

$$\zeta_{1} = -(\xi - W_{0}/3)u - iC_{V} \int \alpha/C_{A} \int B_{ij} - (\xi + W_{0}/3)x,$$
$$u = i \int \boldsymbol{\sigma} \times \mathbf{r} / \int B_{ij},$$
$$x = -C_{V} \int \mathbf{r}/C_{A} \int B_{ij},$$

where  $W_0$  is the total end-point energy of the betas in  $mc^2$  units and  $\xi$  is a dimensionless expansion parameter defined by Kotani. In the analysis of beta-ray experiments, an attempt is made to determine what values of the parameters are consistent with the data.

In the present work the analysis of the data has been performed using a method that was developed in connection with work on Eu<sup>152</sup> and Eu<sup>154,15</sup> In this method no approximations beyond those made by Kotani are used.

### EXPERIMENTAL PROCEDURE AND RESULTS

The Re<sup>186</sup> and Tm<sup>170</sup> were purchased from the Oak Ridge National Laboratory. Two different rhenium irradiations were used. After decay of the 17-h Re<sup>188</sup> no other contaminant was observed in the rhenium. No contaminant was observed in the thulium. Sources were made by evaporation of the respective solutions on 0.25 mil Mylar and grounded with a thin Aquadag line.



<sup>15</sup> Harry Dulaney, C. H. Braden, and L. D. Wyly, Phys. Rev. **125**, 1620 (1962).

TABLE I. Exp. gamma) direction $P_2(\cos\theta)$ in the of even order L R is the number of R/C is the ratio Mean beta energy (keV)	erimental results for nal correlation in F expansion of the dir egendre polynomials of real beta-gamma c of real beta-gamma o	the (934-keV be $e^{186}$ . $A_2$ is the ectional correla s where $A_4$ is a bincidences at 90 coincidences to a P	eta) (137-keV coefficient of tion in terms assumed zero. D and 270 deg. accidentals.
energy (keV)	$A_2$	R	R/C
223	$0.034 \pm 0.007$	12 700	12.7
240	$0.020 \pm 0.007$	12 000	12.0

11 600

11 100

11 800

14 300

19 300

11.6

11.0

5.9

7.6

5.9

 $0.037 \pm 0.007$ 

 $0.036 \pm 0.007$ 

 $0.043 \pm 0.007$ 

 $0.046 \pm 0.007$ 

 $0.054 \pm 0.007$ 

523  $0.073 \pm 0.005$ 29 100 2.6554  $0.071 \pm 0.005$ 41 800 2.9 592 32 400  $0.074 \pm 0.005$ 2.4630 0.082 + 0.005 $32\ 400\ 23\ 200$ 2.4 $0.086 \pm 0.005$ 2.3 668 705  $0.086 {\pm} 0.006$ 15 000 1.8 $0.083 {\pm} 0.010$ 74314 000  $1.3 \\ 2.2$ 832  $0.097 \pm 0.009$ 16 000 All sources were thin enough to be transparent. Several

All sources were thin enough to be transparent. Several different sources of each radioactive material were used during the course of the directional correlation measurements.

The coincidence circuit was of the fast-slow type<sup>16</sup> with a resolving time of approximately  $10^{-7}$  sec. The gamma detector used a  $1\frac{1}{2}$ -in. diameter by 1-in.-high NaI(Tl) crystal. The beta detector used a 1-in. diameter by 1-mm (or 3-mm) anthracene crystal which along with the source were mounted inside a thin-wall aluminum vacuum chamber. The counting geometry is indicated in Fig. 3. The cylindrical vacuum chamber was 3 in. in diameter by  $6\frac{1}{2}$  in. high. The beta counter was contained in a cylindrical sidearm of 2 in. i.d. The directional correlation experiments were performed with the source 4.4 cm from the beta detector and 4.9 cm from the gamma detector. The beta counter was calibrated using the conversion electron lines in Bi<sup>207</sup> and Cs<sup>137</sup>. Measurements were made at angles of 90, 180, and 270 deg between the beta and gamma counters.

The Re<sup>186</sup> was run with the single-channel analyzer in the gamma channel set on the 137-keV gamma with a window width of approximately 12 keV. The reported mean beta energies are believed accurate to  $\pm 10$  keV; the lower energy calibration (below 500 keV) assumes linearity of the amplifier coarse gain control. Many checks of the amplifier gain verify this assumption but due to the errors in the original calibration an additional error of perhaps 5 keV may be introduced. The beta spectrum as displayed on the 20-channel analyzer was divided so as to obtain approximately 30-keV increments; the particular values may be obtained by inspection of Table I. The beta counting rate was checked at 15 min, or 30 min, intervals depending on the time cycle used in a particular run. A shift of greater than 3% in the beta singles rate during a given count was

<sup>&</sup>lt;sup>16</sup> F. K. McGowan, Phys. Rev. 79, 404 (1950).

considered excessive and the data were excluded and repeated. In most cases the shift in the beta singles rate was less than 1%. The coincidence count was normalized to the gamma singles rate. The experimental results are listed in Table I. The indicated errors are probable errors due to statistical effects.  $A_2$  is the usual coefficient of  $P_2(\cos\theta)$  in the expansion of the directional correlation in terms of even-order Legendre polynomials where we assume  $A_4 = 0$ . No gamma-gamma coincidences were observed when a 0.5-cm Lucite absorber was inserted between the source and beta detector.

Correction for the finite angular resolution of the detectors was made according to the method of Rose<sup>17</sup> with the aid of tables of Stanford and Rivers.<sup>18</sup> The modified directional correlation coefficient,  $A_2^*$ , varies slowly with energy and no attempt has been made, in the interpretation of the data, to attribute significance to subtle dependences of this coefficient on the energy. Accordingly, no corrections have been made for effects which would slightly distort the beta spectrum. Such effects include the finite energy resolution of the beta detector, backscattering of betas from the detector crystal, and finite thicknesses of source and backing. Previous measurements on decays with known anisotropic and isotropic directional correlations do not suggest any serious difficulties due to these effects. Coincidence Fermi plots have been made in some cases to ascertain that there are no gross effects which distort the beta spectrum. Our experience with the present experimental method indicates that the most serious problems are concerned with interference from unwanted cascades, including gamma-gamma cascades. The present decays were quite favorable in this respect. The present results are in excellent agreement with those of Novey et al.,9 as may be seen in Fig. 4 where the circles are four typical points chosen from their data.

TABLE II. Experimental results for the (883-keV beta) (84-keV gamma) directional correlation in  $Tm^{170}$ .  $A_2$  is the coefficient of  $P_2(\cos\theta)$  in the expansion of the directional correlation in terms of even order Legendre polynomials where  $A_4$  is assumed zero. R is the number of real beta-gamma coincidences at 90 and 270 deg. R/C is the ratio of the real beta-gamma coincidences to accidentals.

Mean beta energy (keV)	$A_2$	R	R/C
334	$-0.0552 \pm 0.008$	19 100	1.9
365	$-0.0564 \pm 0.006$	19 300	1.9
397	$-0.0521 \pm 0.006$	18 200	2.0
428	$-0.0546 \pm 0.007$	17 300	2.0
523	$-0.0727 \pm 0.008$	15 300	1.1
554	$-0.1005 \pm 0.006$	23 900	1.4
592	$-0.0795 \pm 0.006$	20 500	1.3
630	$-0.0945 \pm 0.007$	17 700	1.4
668	$-0.0773 \pm 0.009$	$14\ 400$	1.3
705	$-0.0828 \pm 0.009$	12 300	1.2
743	$-0.0955 \pm 0.010$	8 800	1.0



FIG. 4. Experimental results for the (934-keV beta) (137-keV gamma) directional correlation in Re<sup>186</sup> and the (883-keV beta) (84-keV gamma) directional correlation in Tm<sup>170</sup>. The circles are typical points taken from the experimental results of Novey et al.<sup>6</sup> The crosses are typical points taken from the experimental results of Bertolini et al.

The Tm<sup>170</sup> was run with the 84-keV gamma peak centered in the single channel analyzer with a window width of approximately 8 keV. The general procedure was the same as that used for the Re<sup>186</sup> except standard counting times were increased to 30 min or 1 h. Experimental results are given in Table II. Results are in excellent agreement with those of Bertolini et al.<sup>6</sup> as evidenced by the crosses in Fig. 3 which are typical points taken from their data. Earlier measurements suggested a somewhat stronger anisotropy.<sup>10</sup>

The finite lifetimes of the first excited states in Os<sup>186</sup> and Yb<sup>170</sup> make it highly desirable to investigate a possible attenuation effect on the directional correlation. Unfortunately, data bearing on this point are not available. Work is proceeding at this laboratory on the observation of beta-gamma correlations in both solid and liquid sources as a function of delay time for delay times of the order of  $10^{-9}$  sec or less.

### INTERPRETATION OF DATA FOR Re<sup>186</sup>

For the 934-keV beta transition of Re<sup>186</sup>, the directional correlation and shape factor data<sup>2</sup> were characterized by the following conditions:

I. The modified beta-gamma directional correlation coefficient,  $A_2^* = 0.064 \pm 0.010$  at a beta energy, W = 1.5.  $A_2^* = WA_2/p^2\lambda_2$ , where  $A_2$  is the usual coefficient of  $P_2(\cos\theta)$  in the expansion of the directional correlation, W is the total electron energy in  $mc^2$  units, p is the electron momentum in mc units and  $\lambda_2$  is a Coulomb correction factor.18

II.  $A_2^* = 0.064 \pm 0.005$  at W = 2.5.

III. The ratio of the beta spectrum shape correction factor at W = 2.566 to that at W = 1.391, S = C(2.566)/ $C(1.391) = 1.145 \pm 0.065.$ 

A recent measurement of the beta-circularly polarized gamma correlation in Re<sup>186</sup> has been reported.<sup>19</sup> We have

 <sup>&</sup>lt;sup>17</sup> M. E. Rose, Phys. Rev. 91, 60 (1953).
 <sup>18</sup> A. L. Stanford and W. K. Rivers, Rev. Sci. Instr. 30, 719 (1959).

<sup>&</sup>lt;sup>19</sup> J. P. Deutsch and P. Lipnik, Ann. Soc. Sci. Bruxelles 74, 195 (1961); M. Delabaye, J. Deutsch, and P. Lipnik, ibid. 75, 171 (1962). We greatly appreciate communications from J. Deutsch and M. Delabaye relative to their data. In particular, we are advised that the algebraic sign of the polarization parameter is positive.



FIG. 5. Plots of Re<sup>186</sup> constraining conditions for  $\zeta_1 = 2$  for the following experimental values: Ia.  $A_2^* = 0.054$  at W = 1.5; Ib.  $A_2^* = 0.074$  at W = 1.5; IIa.  $A_2^* = 0.059$  at W = 2.5; IIb.  $A_2^* = 0.069$  at W = 2.5; IIIa. S = 1.08; IIIb. S = 1.21; IVa.  $\omega = 0.27$  at  $\cos\theta = -0.924$  and W = 1.62; IVb.  $\omega = 0.9$  at  $\cos\theta = -0.924$  and W = 1.62.

endeavored to incorporate this data into the analysis by use of the constraining condition:

IV. The beta-circularly polarized 137-keV gamma correlation coefficient,  $\omega$ ,<sup>14</sup> for a beta energy of 1.62 and  $\cos\theta = -0.924$ , where  $\theta$  is the angle between the beta and gamma, is given by  $\omega = 0.59 \pm 0.32$ .<sup>19</sup>

We found it essential to utilize data at this value of  $\cos\theta$  rather than at  $\cos\theta = -1$  because the data at the latter angle indicates an  $\omega$  that is too large to be consistent with the spectral shape and directional correlation data. Originally we had interpreted  $\omega$  to be of the magnitude quoted here, but negative. In that case we were unable to reconcile the polarization data with the other measurements.

Since the procedure used to obtain suitable parameter sets from the constraining conditions has been described previously,<sup>15</sup> only a brief outline of the method will be given here. When equated to the appropriate theoretical expression each error limit of a given constraining condition yields a conic section in the *x*-*u* plane for a par-



FIG. 6. Plots of Re<sup>186</sup> constraining conditions for  $\zeta_1 = -4$  for the following experimental values: Ia.  $A_2^* = 0.054$  at W = 1.5; Ib.  $A_2^* = 0.074$  at W = 1.5; IIa.  $A_2^* = 0.059$  at W = 2.5; IIb.  $A_2^* = 0.069$  at W = 2.5; IIIa. S = 1.08; IIIb. S = 1.21; IVa.  $\omega = 0.27$  at  $\cos\theta = -0.924$  and W = 1.62.



FIG. 7. Theoretical curves for modified beta-gamma directional correlation coefficient  $A_2^*$  as a function of the beta energy, W, for Re<sup>186</sup> for the following sets of parameters: (a)  $\zeta_1=2$ , x=0.28, u=-0.17; (b)  $\zeta_1=2$ , x=2.3, u=3.04; (c)  $\zeta_1=-4$ , x=1.1, u=0.11; (d)  $\zeta_1=10$ , x=-1.6, u=-0.5; (e)  $\zeta_1=-50$ , x=10.5, u=2.5. The experimental values for  $A_2^*$  which were calculated from the values for  $A_2$  in Table I are also shown.

ticular value of  $\zeta_1$ . Parameter sets satisfying a given constraining condition are represented by points within the area bounded by the two conic sections resulting from the constraining condition. In Figs. 5 and 6 the conic sections produced by the four constraining conditions are plotted for  $\zeta_1=2$  and  $\zeta_1=-4$ , respectively. In Fig. 5 the entire field below the lower branch of the hyperbola corresponding to  $\omega=0.27$  is consistent with the constraint IV. In Fig. 6 the entire field below the lower branch and above the upper branch of the hyperbola corresponding to  $\omega=0.27$  is consistent with the constraint IV. Evidently, the polarization data has not



FIG. 8. Theoretical curves for the normalized beta spectrum shape correction factor,  $C_N(W)$ , as a function of the beta energy, W, for Re<sup>186</sup> for the following sets of parameters: (a)  $\zeta_1=2$ , x=0.28, u=-0.17; (b)  $\zeta_1=2, x=2.3, u=3.04$ ; (c)  $\zeta_1=-4, x=1.1, u=0.11$ ; (e)  $\zeta_1=-50, x=10.5, u=2.5$ . The theoretical curve for the unique shape is also shown. The four sets of error limits simulate the experimental results of Porter *et al.*<sup>2</sup> The theoretical curve for parameter set (d) is also within the error limits.

delimited acceptable matrix element parameters much beyond the limitations imposed by the spectral shape and directional correlation measurements. This was found to be generally true for other values of  $\zeta_1$ .

From Figs. 5 and 6 the following parameter sets are selected for further consideration:

(a)  $\zeta_1 = 2$ , x = 0.28, u = -0.17; (b)  $\zeta_1 = 2$ , x = 2.3, u = 3.04; (c)  $\zeta_1 = -4$ , x = 1.1, u = 0.11.

Sets (a) and (c) satisfy the constraining conditions while set (b) does not quite satisfy the conditions on the spectral shape and the directional correlation and lies well outside the condition on the polarization. From plots for other values of  $\zeta_1$  not shown here the following additional sets of parameters which satisfy all the con-



FIG. 9. Theoretical curves for the beta-circularly polarized gamma correlation coefficient,  $\omega$ , as a function of  $\cos\theta$ , for a beta energy of W = 1.62, for  $\mathbb{R}^{186}$  and the following sets of parameters: (a)  $\zeta_1 = 2$ , x = 0.28, u = -0.17; (c)  $\zeta_1 = -4$ , x = 1.1, u = 0.11; (e)  $\zeta_1 = -50$ , x = 10.5, u = 2.5. The curve for the parameter set (d) is similar to that for set (e).

straining conditions have been selected to typify the results over the range of the parameter  $\zeta_1$  investigated, viz.  $|\zeta_1| \leq 50$ .

(d)  $\zeta_1 = 10$ , x = -1.60, u = -0.50;

(e) 
$$\zeta_1 = -50, \quad x = 10.5, \quad u = 2.5.$$

The parameter sets determined from the constraining conditions must be checked to see if they produce satisfactory agreement with the experimental data over the entire energy range. In Fig. 7 the theoretical curves for  $A_2^*$  vs energy for each of the parameter sets is plotted and compared with the experimental data. In Fig. 8 the theoretical curves for the normalized shape correction factor,  $C_N(W)$ , vs energy are plotted for each of the parameter sets. The four sets of error limits shown in Fig. 8 are chosen to simulate the experimental results of Porter *et al.*<sup>2</sup> Both the theoretical curves and the experimental points have been normalized at



FIG. 10. Plots of the Tm<sup>170</sup> beta-gamma directional correlation constraining conditions for  $\zeta_1 = -4$  for the following experimental values: Ia.  $A_2^* = -0.060$  at W = 1.6; Ib.  $A_2^* = -0.080$  at W = 1.6; IIa.  $A_2^* = -0.050$  at W = 2.5; IIb.  $A_2^* = -0.070$  at W = 2.5.

W=2.0. Parameter sets (a), (c), (d), and (e) produce theoretical curves for  $A_2^*$  and  $C_N(W)$  that satisfactorily fit the experimental data over the entire energy range. However, set (b) produces theoretical curves for  $A_2^*$ and  $C_N(W)$  that are in gross disagreement with the experimental data.

In Fig. 9 theoretical curves for  $\omega$  vs cos $\theta$  are plotted for several parameter sets. Our interpretation of the experimental results<sup>19</sup> are also shown. For parameter sets (a), (c), (d), and (e) there is satisfactory agreement with the rather wide limits placed on  $\omega$  by the data, except the value of  $\omega$  for  $\cos\theta = -1$  is too large to be consistent with the combination of the spectral shape and directional correlation data. Parameter set (b)



FIG. 11. Plots of the Tm<sup>170</sup> beta-gamma directional correlation constraining conditions for  $\zeta_1=4$  for the following experimental values: Ia.  $A_2^*=-0.060$  at W=1.6; Ib.  $A_2^*=-0.080$  at W=1.6; IIa.  $A_2^*=-0.050$  at W=2.5; IIb.  $A_2^*=-0.070$  at W=2.5.



FIG. 12. Plots of Tm<sup>170</sup> shape factor constraining equations for  $\zeta_1 = -4$  for values of  $S_1$  and  $S_2$  from 0.90 to 1.10.  $S_1$  is the ratio of the shape factor at W=2.2 to the shape factor at W=1.5.  $S_2$  is the ratio of the shape factor at W=2.728 to the shape factor at W=2.2.

yields a negative  $\omega(\omega = -0.11 \text{ for } \cos\theta = -1 \text{ and } \omega = -0.27 \text{ for } \cos\theta = 0).$ 

It is noted that for many values of  $\zeta_1$ , other than those considered above, there are additional parameter sets that produce satisfactory fits of the directional correlation and shape factor data. However, for a particular value of  $\zeta_1$  the suitable parameter sets are generally restricted to a single region of the x-u plane. In fact, parameter sets producing suitable solutions seem to exist for any value of  $|\zeta_1|$  greater than about 2. For  $\zeta_1$  near zero it is not possible to fit the spectral shape data. For  $\zeta_1$  near unity, some simultaneous fits to the directional correlation and spectral shape constraints may be found but the energy dependence is similar in nature to that for parameter set (b). Although no upper limit for  $|\zeta_1|$  was determined, it was established that satisfactory solutions exist for  $|\zeta_1| = 50$ . Also as  $|\zeta_1|$ increases, no trend toward poorer fitting of the experimental data is apparent.

In conclusion, parameter sets that will simultaneously fit the directional correlation and the shape factor for the 943-keV beta group of Re<sup>186</sup> exist for values of  $|\zeta_1|$  greater than 2. These parameter sets are also in rough agreement with the beta-circularly polarized gamma measurement but there is a discrepancy for the measurement at  $\cos\theta = -1$ .

# INTERPRETATION OF DATA FOR Tm170

Since available evidence indicated the 883-keV beta group of  $Tm^{170}$  has the allowed shape,<sup>4-6</sup> it was found convenient to take a somewhat different approach in interpreting the data for this transition than was used for Re<sup>186</sup>. It was found that in order to determine parameter sets producing an allowed shape, a somewhat more stringent constraining condition was required than simply setting the ratio of the shape correction factor at the extremities of the experimental energy range equal to 1. The experimental results for the (883-keV beta) (84-keV gamma) directional correlation in Tm<sup>170</sup> were characterized by the following two constraining conditions:

I. 
$$A_2^* = -0.070 \pm 0.010$$
 at  $W = 1.6$ ;  
II.  $A_2^* = -0.060 \pm 0.010$  at  $W = 2.5$ .

In Figs. 10 and 11, the conic sections produced by these constraining conditions are plotted for values of  $\zeta_1 = -4$  and  $\zeta_1 = +4$ , respectively. For both of these particular values of  $\zeta_1$ , there is a wide range of x and u values that satisfactorily satisfy the directional correlation constraining conditions. In general, it was found that for any value of  $\zeta_1$ , there is a range of x and u values that satisfy the directional correlation constraining conditions.

In order to predict the form of the shape correction factor for the parameter sets chosen on the basis of the directional correlation, the following procedure was used. The energy range over which shape factor data might be available was divided into two intervals. The first interval was from a beta energy W = 1.5 to W = 2.2; the second was from W=2.2 to the end-point energy,  $W_0 = 2.728$ . The ratio of the shape factor at W = 2.2 to the shape factor at W=1.5 and the ratio of the shape factor at W=2.728 to the shape factor at W=2.2 are denoted by  $S_1$  and  $S_2$ , respectively. By constraining the values of  $S_1$  and  $S_2$ , a variety of shapes including the allowed shape may be produced. The theoretical expression for  $S_1$  was equated to a series of values between 0.90 and 1.10. For a particular value of  $\zeta_1$ , the resulting equations produce a conic section in the x-u plane for each value of  $S_1$ . The conic sections produced by this process for  $\zeta_1 = -4$  and  $\zeta_1 = +4$  are shown in Figs. 12 and 13, respectively. The process was repeated for values of  $S_2$  between 0.90 and 1.10 producing a second series of conic sections, also shown in Figs. 12 and 13. It is expected that a parameter set that is represented by a point that is near both the  $S_1=1$  and  $S_2=1$  conic



FIG. 13. Plots of  $Tm^{170}$  shape factor constraining equations for  $\zeta_1 = 4$  for values of  $S_1$  and  $S_2$  from 0.90 to 1.10.



FIG. 14. Theoretical curves for modified beta-gamma directional correlation coefficient  $A_2^*$  as a function of the beta energy, W, for Tm<sup>170</sup> for the following sets of parameters: (a)  $\zeta_1 = -2.8$ , x=0, u=0; (b)  $\zeta_1 = -4.0$ , x = -0.20, u=0; (c)  $\zeta_1 = -8.0$ , x = -1.05, u=0.05; (d)  $\zeta_1 = -50.0$ , x = -11.5, u=0; (e)  $\zeta_1 = 50.0$ , x = 13.0, u=0. The experimental values for  $A_2^*$  which were calculated from the values for  $A_2$  in Table II are also shown.

sections will produce a near allowed shape. Furthermore, by choosing points that are near conic sections produced by particular values of  $S_1$  and  $S_2$ , it is possible to produce a variety of shape factors.

For the two values of  $\zeta_1$  chosen for this discussion, the possibility of simultaneously satisfying the directional correlation results and producing an allowed shape will be investigated. For  $\zeta_1 = -4$ , it is seen that the point x = -0.20, u = 0 satisfies the directional correlation constraining conditions and should also produce a near allowed shape since it is near both the  $S_1 = 1$  and  $S_2 = 1$  conic sections. However, for  $\zeta_1 = +4$  no set of parameters satisfying the directional correlation constraining conditions also produces a near allowed shape.

In general, parameter sets satisfying just the directional correlation results exist for all values of  $\zeta_1$  over a wide range of x and u values. However, if in addition the sets are required to produce a near allowed shape, the suitable sets are restricted to  $\zeta_1 < -2$  and  $\zeta_1 > 20$ . Also for a particular  $\zeta_1$ , the x and u values are restricted to a narrow range. Although no upper limit for  $\zeta_1$  was determined, it was established that satisfactory solutions exist for  $\zeta_1 = 50$  and  $\zeta_1 = -50$ . Also as  $\zeta_1$  increases no trend toward poorer fitting of the data is apparent.

From the wide range of suitable parameter sets, the following are chosen as typical:

(a)	$\zeta_1 = -2.8,$	x=0,	u=0;
(b)	$\zeta_1 = -4.0,$	x = -0.20,	u=0;
(c)	$\zeta_1 = -8.0,$	x = -1.05,	u = 0.05;
(d)	$\zeta_1 = -50.0,$	x = -11.5,	u=0;
(e)	$\zeta_1 = 50.0,$	x = 13.0,	u=0.

It is noted that set (a) is equivalent to the "modified  $B_{ij}$ " solution. Therefore, for this decay the modified  $B_{ij}$  approximation produces a suitable solution with  $\zeta_1 = -2.8$ . However, the "modified  $B_{ij}$ " solution is only one of a large number of suitable solutions.

In Figs. 14 and 15, the theoretical curves for the modified beta-gamma directional correlation,  $A_{2}^{*}$ , and the normalized beta spectrum shape correction factor,  $C_N(W)$ , are plotted as a function of energy for each of the above parameter sets. In Fig. 14 the experimental limits for the directional correlation are also shown. In Fig. 15 the shape factors have been normalized at a beta energy W=2.2. Also, in Fig. 15 the unique shape is shown for purposes of comparison. Thus, it is seen that each of the above sets represents a satisfactory solution of the beta-gamma directional correlation experiment and also produces an allowed shape for the beta spectrum.

### COMMENT ON SELECTION RULES

In view of the large range of values of the parameter  $\zeta_1$  that yield fits to the experimental data for both Re<sup>186</sup> and Tm<sup>170</sup>, the analysis considered here does not suffice to prove that the ordinary first forbidden beta decay matrix elements are attenuated relative to the  $B_{ij}$  matrix element. An indication that there is attenuation of the ordinary matrix elements (but not necessarily relative to the  $B_{ij}$  matrix element) lies in the large *ft* products for the decays. Attempts to find some general theoretical basis for attenuation of first forbidden beta decay matrix elements in terms of selection rules involving the concepts of "K forbiddenness" have been made.<sup>14</sup> In particular, the concept of K forbiddenness seems attractive when the beta decay follows the spin sequence  $3(\beta)^2$  and the spin 2 may be associated with a rotational state of zero intrinsic angular mo-



FIG. 15. Theoretical curves for the normalized beta spectrum shape correction factor,  $C_N(W)$ , as a function of the beta energy, W, for Tm<sup>170</sup> for the following sets of parameters: (a)  $\zeta_1 = -2.8$ ,  $x=0, \ u=0$ ; (b)  $\zeta_1 = -4.0, \ x=-0.20, \ u=0$ ; (c)  $\zeta_1 = -8.0, \ x=-1.05, \ u=0.05$ ; (d)  $\zeta_1 = -50.0, \ x=-11.5, \ u=0$ ; (e)  $\zeta_1 = 50.0, \ x=13, \ u=0$ . In order to illustrate how slightly the curves deviate from the allowed shape, the theoretical curve for the unique shape is also shown.

mentum. Then  $\Delta K = 3$ . In the present nuclides the spin sequence is  $1(\beta)2$ . The spin 2 may reasonably be interpreted as due to a collective excitation of the daughter<sup>20</sup> but the result is  $\Delta K = 1$ ; hence K forbidden-

<sup>20</sup> A. Bohr and B. R. Mottelson, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955).

ness does not seem to apply. The concept of "i forbiddenness" leads to fairly definite predictions only if the neutron undergoing decay and the proton into which it decays lie in the same major shell. For the present nuclides the neutron and proton lie in adjacent major shells, hence j forbiddenness does not yield a strong selection rule effect.

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# Perturbation of Alpha Particle-Gamma Ray Angular Correlations in $Am^{241}$ , $Am^{243}$ , and $Cm^{243}$ †

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By utilization of the recoil properties associated with the alpha decay process, perturbations of alphagamma angular correlations in Am<sup>241</sup>, Am<sup>243</sup>, and Cm<sup>243</sup> were observed in a variety of media including vacuum, metals, Mylar, oxides and liquids. In the vacuum and Mylar environments the Am<sup>243</sup> correlation is not only considerably more perturbed than in metallic environments but it exhibits an attenuation coefficient below the hard-core value for a static interaction. The perturbation in vacuum is attributed to a hyperfine structure interaction involving an electron shell excited in the alpha decay process. Several experiments designed to further explore the effect of alpha decay on the electron shell are suggested. It is pointed out that application of a strong magnetic field along the direction of one of the radiations may completely restore the correlation in vacuum. Improvement of the correlation in metals as compared to insulators may be related to the rapid recovery time of the electron shell in metallic media.

### INTRODUCTION

HE perturbation of alpha-gamma angular correlations has been reported in a number of even-even isotopes,<sup>1</sup> as well as in Åm<sup>241</sup>,<sup>2,3</sup> and Am<sup>243</sup>.<sup>4</sup> Interactions between the quadrupole moments of the intermediate nuclear states and electric field gradients of extraatomic origin appeared responsible for attenuating the correlations. The present experiments were originally designed to eliminate these extra-atomic effects by observing angular correlations between alpha particles and the gamma rays from daughter nuclei recoiling into vacuum. After unexpectedly large perturbations were found to occur in vacuum, the experiments were extended to a variety of media including several whose influence on the angular correlation had not been previously explored.

### EXPERIMENTAL METHODS

The methods used to prepare and study thin Am<sup>241</sup>. Am<sup>243</sup>, and Cm<sup>243</sup> sources have been discussed in detail elsewhere.<sup>5</sup> Approximately 0.05  $\mu$ g of activity was freed from macroscopic impurities by repeated cation exchange and vaporized from a hot Ta filament onto an area  $\frac{7}{8}$  cm in diameter. The backing material consisted of thin metal or plastic films which were transparent to alpha particles but not to heavy recoils. The percentage of Np<sup>239</sup> recoils escaping from the surface of the Am<sup>243</sup> sources was determined by counting the beta activity collected over the source in vacuum. At least 45% of the recoils were found to escape into the vacuum; an equal percentage presumably were absorbed in the backing film, while less than 10% remained in the radioactive deposit.

The sources were centered in a cubical vacuum chamber at a 45° angle to the walls of the chamber. Gamma rays were detected by means of a fixed sodium iodide crystal which paralleled one face of the chamber and subtended a half-angle ranging from 11 to 19 deg at the source. Two zinc sulfide screens centered on adjacent faces of the chamber detected alpha particles at 90° and 180° to the observed gamma rays. Pulses from the two alpha detectors were successively counted and fed into a slow coincidence unit together with pulses from the

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<sup>1</sup> J. Battey, L. Madansky, and F. Rasetti, Phys. Rev. 89, 182 (1953); J. K. Beling, B. T. Feld, and I. Halpern,</sup> *ibid.* 84, 155 (1951); J. C. D. Milton and J. S. Fraser, *ibid.* 95, 628 (1954); G. M. Temmer and J. M. Wyckoff, *ibid.* 92, 913 (1953); G. Valladas, J. Teillac, P. Falk-Vairant, and P. Benoist, J. Phys. Radium 16, 123 (1955).
<sup>2</sup> J. S. Fraser and J. C. D. Milton, Phys. Rev. 94, 795 (1954).
<sup>3</sup> V. E. Krohn, T. B. Novey, and S. Raboy, Phys. Rev. 105, 234 (1957).

<sup>234 (1957).</sup> 

<sup>&</sup>lt;sup>4</sup> F. Stephens, J. Hummel, F. Asaro, and I. Perlman, Phys. Rev. 98, 261 (1955).

<sup>&</sup>lt;sup>5</sup> E. Flamm, Ph.D. thesis, University of California Lawrence Radiation Laboratory Report UCRL-9325, 1960 (unpublished).