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The Forbidden Transition of Yttrium^{91} and Cesium^{137} *,**

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Y^{91} decays by the emission of a single beta-particle with a maximum energy of 1.54 Mev, while Cs^{137} decays in two ways, (1) beta-decay (maximum energy=0.518 Mev) to Ba^{137*} followed by a gamma-transition to the ground state, and (2) beta-decay (maximum energy=1.2 Mev) directly to the ground state. Probably not more than 5 percent of the Cs^{137} nuclei decay directly to the ground state. Conventional (allowed transition) Kurie plots of Y^{91} and the low energy group of beta-particles of Cs^{137} display curvature which is concave toward the energy axis at high energies and concave upward at low energies. If it is assumed that Gamow-Teller selection rules govern the beta-process, the Fermi function $F(Z, W)$ for allowed spectra must be multiplied by a factor $G=(W^2-1)+(W_0-W)^2$ in a beta-transition for which there is a change of parity and for which the spin change is two units. If these conventional Kurie plots of Y^{91} and Cs^{137} are modified by the factor G, the resulting plots are approximately straight lines. This indicates that the beta-decay of Y^{91} and the low energy beta-decay of Cs^{137} involve a spin change of two units and a parity change. These results contribute evidence for the validity of the Fermi theory of beta-decay and the Gamow-Teller selection rules.

The K internal conversion coefficient for the 0.663-Mev gamma from the decay of Cs^{137} was found to be 0.081, while the ratio of the K conversion electrons to the L conversion electrons was 5.0. The gamma-radiation seems to be either magnetic 2^4-pole or electric 2^5-pole.

INTRODUCTION

IN the beta-decay process the distributions of beta-particles as a function of energy are of two types, allowed and forbidden. Using the Kurie plot procedure to compare experimental results with the Fermi theory, allowed spectra should give straight line Kurie plots, while forbidden spectra are, in general, expected to yield Kurie plots which deviate from straightness. To date, experimental measurements of (presumably) allowed spectra have confirmed the theory fairly well, except perhaps in the very low energy region.[1] For many forbidden spectra, however, the type of deviation to be anticipated is not clearly understood because of a lack of uniqueness in the various possible formulations of the theory, and because the spins and parities of the initial and final nuclear states are frequently unknown. A third source of ambiguity is the possibility that more than one nuclear matrix element may appear in the distribution function, resulting in a spectrum which depends on the ratios and relative phases of the nuclear matrix elements.

Under certain circumstances, the shape of a forbidden spectrum can be predicted. If it is assumed that Gamow-Teller selection rules govern the beta-process, the distribution in a transition for which there is a change of parity and for which the spin change is two units will differ from that in an allowed transition by a simple function of the energy. Such a transition is classified as first-forbidden by the Gamow-Teller selection rules. In this case, the Fermi function $F(Z, W)$ for allowed spectra must be multiplied by a factor G, which, if $\alpha Z \ll 1$ and if the electron momentum is not too small, has the approximate form[2,3]

$$G=(W^2-1)+(W_0-W)^2.$$

Here W is the electron energy, and W_0 is the total energy available for the transition, both in units of mc^2. The omission of this correction tends to make the Kurie plot (computed as though the transition were allowed) concave toward the energy axis in the high energy region. If $W_0 \geqq 2$, the same omission also causes the Kurie plot to deviate upward at low energies.

In general, however, the link between theory and experiment is very weak for forbidden spectra. More often than not one or both spins of the parent and daughter nuclei in a beta-transition are not known. Initial and final parities of the nuclear states are even less well known. With so many unknown factors, theoretical predictions for the distribution of beta-particles in forbidden transitions, even on the assumption of a uniquely determined heavy-light particle interaction, is largely guesswork.

In some cases the shell model[4] of the nucleus may help to determine the spin and parity changes. For isotopes in certain regions of atomic number and atomic weight, this model predicts both the parities and the possible spin values for the nuclei involved. Ambiguities in the predictions of the shell model can be reduced by consideration of the energies and half-lives of beta- and gamma-transitions in a series of isobaric nuclei.

The isotope Y^{91}, which decays by β^--emission to Zr^{91} with a half-life of 57 days, is one for which both the spin and the parity change can be inferred from the shell model. For this transition, Feenberg and Hammack give a preferred spin change of 2 (from $\frac{1}{2}$ to 5/2) and a parity change from odd to even. Thus, if the shell model of the

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[1] C. S. Cook and L. M. Langer, Phys. Rev. 73, 601 (1948).

[2] Here α is the fine structure constant and Z is the charge on the nucleus.

[3] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. 60, 308 (1941); E. J. Konopinski, Rev. Mod. Phys. 15, 226 (1943).

[4] E. Feenberg and K. C. Hammack (to be published in Phys. Rev.)

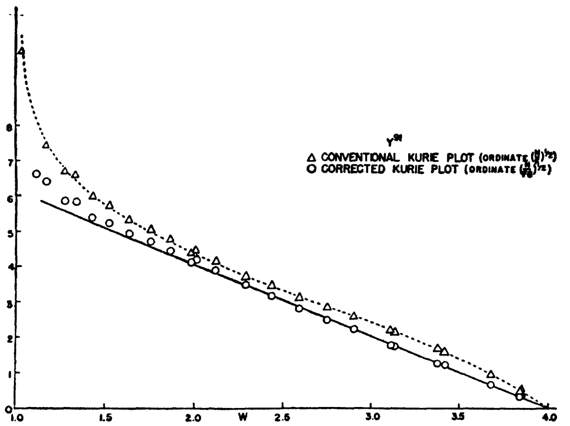

FIG. 1. Kurie plot of Y^{91} and a Kurie plot of the same element corrected by the factor $G=(W_0-W)^2+(W^2-1)$.

nucleus is at all reliable and if the Gamow-Teller selection rules apply, the decay of Y^{91} is an example of the particular kind of first-forbidden transition which was discussed above. Therefore, the distribution of the beta-particles as a function of energy should differ from the allowed distribution by the factor G. The recent work of Langer and Price[5] seems to confirm that this is the case. The Y^{91} spectrum thus provides a critical test of the Fermi theory for forbidden spectra and of the G-T selection rules. It also affords a minor test of the nuclear shell model. For these reasons it is felt that confirmation of the results published by Langer and Price is desirable.

The shell model again offers some help in understanding the decay of Cs^{137} since it predicts *even* parity for the ground states of both Cs^{137} and Ba^{137}. The transition is known to proceed by negatron emission to a mestastable state, Ba^{137*}, which then decays by gamma-emission (accompanied by internal conversion) to the ground state Ba^{137}. The known half-life of the metastable state requires an electric 2^5-pole transition with $\Delta I=5$ or a combination of electric 2^5-pole and magnetic 2^4-pole transitions with $\Delta I=4$. Either way the parity changes. A choice between these possibilities can be based on the internal conversion coefficients in the K and L shells. The spin of Ba^{137} is known to be $\frac{3}{2}$. Putting all this information together, one predicts $I=11/2$ or $13/2$ with odd parity for the metastable state Ba^{137*}. Mitchell and Peacock[6] have measured the K internal conversion coefficient (α_K) and the ratio N_K/N_L. Using the approximate theoretical equations of Dancoff and Morrison,[7] they conclude that $\Delta I=5$ and hence, $I=13/2$ in the isomeric state. The observations of Mitchell and Peacock[8] on the shape of the beta-spectrum lead them to conclude that the beta-transition $Cs^{137}\rightarrow Ba^{137*}$ is an example of the particular kind of first-forbidden transition discussed above with a distribution function modified by the presence of the factor G. Support for the latter conclusion may be derived from a comparison of ft values in the Y^{91} and Cs^{137} transitions. If f is derived from the theory of allowed transitions, the quantity W_0^2ft should be more nearly constant than ft in this particular type of decay. In fact, $W_0^2ft=9.7\times 10^9(Cs^{137}\rightarrow Ba^{137*})$ and $W_0^2ft=8.6\times 10^9(Y^{91}\rightarrow Zr^{91})$.

EXPERIMENTAL METHOD

The spectra of Cs^{137} and Y^{91} were studied in the double-focusing spectrometer described by Kurie, Osoba, and Slack.[9] The inherent resolution of this instrument is 0.29 percent as determined from a measurement of the thorium F-line, using a narrow source and a narrow counter window. For the present work, however, the high resolving power of the instrument was sacrificed to gain higher counting rates and to allow the use of thinner sources. With wide sources and a wide counter window, the resolving power was reduced to about one percent.

Radioactive sources were prepared by first defining the source area with insulin[10] on a thin Zapon film about 0.03 mg/cm² thick. The active material in solution was then applied to this area and fast-dried under an infrared lamp. The author feels it would be useless to give a figure for the source thickness since the active deposit seemed to be in small crystals. The source was prevented from charging[11] by painting two stripes of Aquadag on

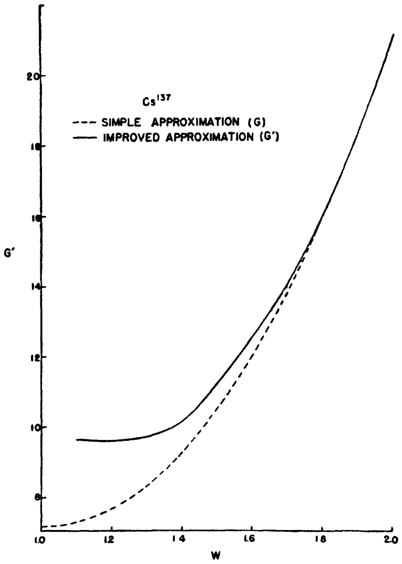

FIG. 2. A plot of $G=(W_0-W)^2+(W^2-1)$ and a plot of $G'=(W_0-W)^2+A(W^2-1)$ for Cs^{137}.

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[5] L. M. Langer and H. C. Price, Phys. Rev. **75**, 1109 (1949).

[6] A. C. G. Mitchell and C. L. Peacock, Phys. Rev. **75**, 197 (1949).

[7] S. M. Dancoff and P. Morrison, Phys. Rev. **55**, 122 (1939).

[8] A. C. G. Mitchell and C. L. Peacock, Phys. Rev. **75**, 1272 (1949).

[9] Kurie, Osoba, and Slack, Rev. Sci. Inst. **19**, 771 (1948).

[10] It is found that when insulin is applied to an area and then allowed to dry, subsequent application of a solution will be limited to an area defined by the insulin.

[11] C. H. Braden, G. E. Owen, J. Townsend, C. S. Cook, and F. B. Shull, Phys. Rev. **74**, 1539 (1948).

the back side of the source support to hold the source at ground potential.

Detection of particles was accomplished by means of a Geiger-Müller counter with a Zapon film window which transmitted negatrons above 3.5 kev. The window was 0.25 inch wide and 0.625 inch high, with a 5-mil wire centered lengthwise across the window to help support the Zapon film. A window of this size made from thin Zapon film is difficult to make gas-tight. To keep the pressure in the counter unchanged, a constant pressure device similar to that developed by Ter-Pogossian, Robinson, and Townsend[12] was connected to the counter. The counter pressure changed less than 0.2 percent even though gas was always leaking out through the window.

RESULTS

Yttrium^{91}

The data obtained for Y^{91} is shown in Fig. 1, plotted as a conventional (allowed transition) Kurie plot, and displays curvature which is concave toward the energy axis at high energies and concave upward at low energies. For this plot, the value of $F(Z, W)$ was obtained from the curves found in the *Theoretical Nuclear Physics Course* lecture notes obtained from Oak Ridge. The chosen value of Z is 40, corresponding to the daughter nucleus Zr^{91}. Also shown in Fig. 1 is the Kurie plot which results when $F(Z, W)$ is multiplied by the correction factor G. The near-straightness of the corrected curve is in satisfactory agreement with the work of Langer and Price, although a slight upward deviation still remains at low energies. The deviation at low energy can probably be attributed to scattering due to the fact that the source was not sufficiently thin. The observed value of W_0 is 4.0 mc^2 or 1.54 Mev.

Cesium^{137}

Previous work done in this laboratory[13] indicated that Cs^{137} decays by simple beta-emission to a metastable state, Ba^{137*}, which then decays (half-life = 158 sec.) to the ground state of Ba^{137} by the emission of a 0.663-Mev gamma-ray. The present investigation confirms a result obtained by Mitchell and Peacock[8] that the decay of Cs^{137} occurs in two ways, (1) beta-decay ($W_0 = 0.518$ Mev) to Ba^{137*} followed by the gamma-transition to the ground state, and (2) beta-decay ($W_0 \sim 1.18$ Mev.) directly to the ground state. As will be shown, probably not more than 5 percent of the Cs^{137} nuclei decay directly to the ground state. This confirms the findings of Mitchell and Peacock.

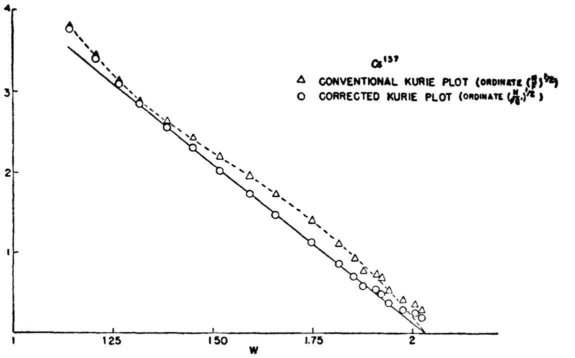

FIG. 3. Kurie plot of Cs^{137} and a Kurie plot of the same element corrected by the factor $G' = (W_0 - W)^2 + A(W^2 - 1)$.

A conventional Kurie plot of the more abundant negatron spectrum (Fig. 3, upper curve) reveals a curvature similar to that obtained for Y^{91}. When the function $F(Z, W)$ is multiplied by G, the re-computed Kurie plot yields a fairly good straight line (not shown in Fig. 3) at high energy. At low energy, however, the curve bends strongly upward. This latter behavior is caused in part by an imperfect approximation in evaluating the correction factor G. For $Z = 56$, the assumption that $\alpha Z \ll 1$ is not valid. Mr. J. Davidson of this laboratory has recomputed G, keeping terms in αZ and $(\alpha Z)^2$ which were previously neglected. His equation for the correction factor, now labeled G', and still subject to the assumption of reasonably large momentum, is

$$G' = (W_0 - W)^2 + A(W^2 - 1),$$

where

$$\begin{aligned} A = {} & \left[\frac{2}{1+(1-\alpha^2Z^2)^{\frac{1}{2}}}\right]\left[\frac{2+(4-\alpha^2Z^2)^{\frac{1}{2}}}{4}\right] \\ & \times \left\{\frac{12\Gamma[1+2(1-\alpha^2Z^2)^{\frac{1}{2}}]}{\Gamma[1+2(4-\alpha^2Z^2)^{\frac{1}{2}}]}\right\}^2 (2P\rho)^{\alpha^2Z^2/2} \\ & \times \left|1-\frac{\alpha^2Z^2}{4}+\frac{i\alpha ZW}{P}\right|^2 \left|\left(1-\frac{\alpha^2Z^2}{4}C\right) + \frac{\alpha^2Z^2}{4}\cdot\frac{\alpha^2Z^2W^2}{P^2}\sum_{\nu=1}^{\infty}\frac{1}{\nu\left(\nu^2+\dfrac{\alpha^2Z^2W^2}{P^2}\right)} \right. \\ & \left. + i\left\{\alpha^2Z^2\cdot\frac{\alpha ZW}{4P}\sum_{\nu=1}^{\infty}\frac{1}{\nu^2+\dfrac{\alpha^2Z^2W^2}{P^2}}\right\}\right|^2, \end{aligned}$$

where C = Gauss's number. Figure 2 compares G and G' for Cs^{137}. It should be noted that use of the simple form G would produce an upward deviation at low energies in the Kurie plot. Figure 3 shows how the Kurie plot for Cs^{137} is straightened out by the correction-factor G'. (It should be noted that G and G' would not differ appreciably from each other for Y^{91}, where $W_0 = 4.0$. This is because the term $(W_0 - W)^2$ is very large compared to $A(W^2 - 1)$ even at low W, where A is largest.)

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[12] Ter-Pogossian, Robinson, and Townsend, Rev. Sci. Inst. **20**, 289 (1949).

[13] Townsend, Owen, Cleland, and Hughes, Phys. Rev. **74**, 99 (1948). Townsend, Cleland, and Hughes, Phys. Rev. **74**, 499 (1948).

The less abundant group of negatrons was studied with a stronger source, but the counting rate was still so low that small reliance should be placed in the spectrum obtained. Also, since the source was thick, the resulting spectrum shape is probably distorted by inelastic scattering in the source. A Kurie plot of this spectrum for energies above 2.35 mc^2 is shown in Fig. 4.

Because of the uncertainty in the expected shape of the spectrum of the high energy group of beta-particles, an accurate determination of the relative abundance of this group is impossible. An attempt was made, however, to determine limits for the abundance of this high energy group. In order to simplify the calculations, straight line Kurie plots were used though the spectrum, in all probability, does not have the Fermi allowed shape. In order to set a lower limit for the abundance of this group, the straight line (*A* in Fig. 4) passing from the expected end point (1.18 Mev$\approx$0.663+0.518) through the point at 2.35 mc^2 was assumed to represent the spectrum; the relative abundance of the high energy group was accordingly found to be 2 percent. The author feels that this procedure gives a value for the abundance which is almost certainly too low. In order to set an upper limit for the abundance of the high energy group, the straight line (*B* in Fig. 4) passing from the expected end point through the point indicated by a triangle was assumed to represent the spectrum. The point represented by the triangle was obtained using a thin source and was normalized to conform to the thick source counting rate. This point is located between the end point of the low energy beta-group (0.518 Mev) and the *K* conversion peak (0.625 Mev). Counts in this region are due to the high energy beta-group and possible scattering of some of the internal conversion electrons. The relative abundance of the high energy group corresponding to line *B* in Fig. 4 was found to be 5 percent. Even though the scattering was minimized by use of a thin source, the point represented by the triangle is probably too high and the value of 5 percent can be considered an upper limit for the relative abundance of the high energy group.

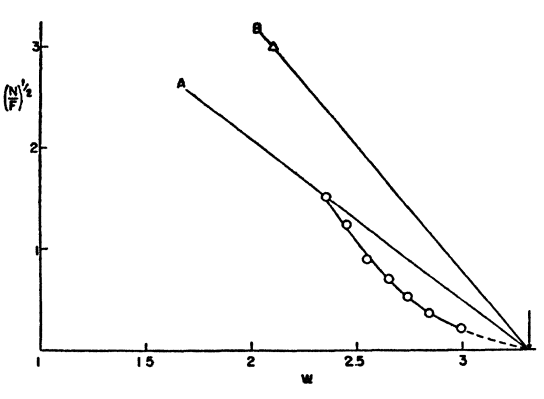

FIG. 4. Kurie plot of high energy group of beta-particles from Cs^{137} with two straight lines that were used to determine the abundance of this group.

The *K* internal conversion coefficient (α_K) for a transition is defined as the ratio of the number of *K* electrons ejected to the number of gammas which are emitted. This coefficient for the 0.663-Mev gamma was found to be $\alpha_K=0.081$, while the ratio N_K/N_L[14] was 5.0. The latter value agrees fairly well with the determination by Mitchell and Peacock who found $N_K/N_L=4.8$, but their value for α_K was 0.118. According to the Oak Ridge tables of internal conversion coefficients,[15] which are calculated from the relativistic formula, one finds that $\alpha_K=0.081$ for Ba^{137*} falls between the values for magnetic 2^4-pole ($\beta_K{}^4=0.094$) and electric 2^5-pole ($\alpha_K{}^5=0.053$) radiation, which in both cases requires a change of parity. (Since the tables of Rose *et al.*, were not specifically computed for $Z=56$ and energy 0.663 Mev, a careful graphical interpolation was carried out to obtain the values for $\beta_K{}^4$ and $\alpha_K{}^5$ which are quoted above.)

The value of 0.081 for the *K* internal conversion coefficient may be too high, because the window of the counter cuts out an undetermined part of the beta-spectrum. Even though the window of the counter has a cut-off of three kev, it probably attenuates the number of negatrons that reach the counter with energies below fifty kev. If there is inelastic scattering in the source, a portion of the beta-spectrum will be degraded to a region where there is attenuation in the counter window.

CONCLUSIONS

Yttrium^{91}

The agreement observed by the present author and by Langer and Price between experimental results for Y^{91} and the theoretical predictions indicate that its decay involves a spin change of two units together with a change of parity. The results show the usefulness of the Feenberg-Hammack nuclear shell model in predicting spin and parity changes and also provide evidence for the validity of the Gamow-Teller selection rules and the Fermi theory for forbidden spectra.

Cesium^{137}

The results for Cs^{137} are not so conclusive. Examination of the data leads to the following conclusions:

(1) There is branching in the decay of Cs^{137}, with the higher energy group accounting for between two and five percent of the total disintegrations. The end point of the low energy beta-group is 0.518 Mev, the energy of the gamma is 0.663 Mev, and the end point of the high energy betas is about 1.18-Mev.

(2) The shape of the low energy beta-spectrum indicates that this transition may involve a spin change of two together with a change of parity.

(3) A spin change of 4 or 5 and a change of parity appear to be most probable for the transition from metastable Ba^{137*} to the ground state.

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[14] No use was made of this ratio because no theoretical curves which are calculated from the relativistic formula were available.

[15] Rose, Goertzel, Spinrad, Harr, Strong (prepublication copy).

Using the ground state of Ba^{137} ($I=\frac{3}{2}$, even) as a starting point, Fig. 5 shows two possible spin assignments which can be made for levels in the disintegration scheme of Cs^{137}.

In diagram A, Fig. 5, the spin change in the high energy transition is $\Delta I=2$ with no change in parity, placing it in the second-forbidden class, according to the G-T selection rules. The figure of 0.02 to 0.05 for the branching ratio is perhaps too small for this interpretation in view of the fact that the larger energy change partially compensates for the higher degree of forbiddenness. Unfortunately, there are no well established examples of similar transitions with which to make comparison.

If diagram B, Fig. 5, is accepted as correct, the high energy group will still be second forbidden according to the Gamow-Teller selection rules, but with $\Delta I=3$ and no change of parity. The probability for this transition is considerably smaller than for second-forbidden with $\Delta I=2$ (no), so that the number of beta-particles found in the high energy group may be now somewhat too high.

Even though the decay scheme for Cs^{137} is not definite, there is no pronounced disagreement with theoretical expectations. The shape of the low energy beta-spectrum seems to conform with the shell theory of the nucleus, the Gamow-Teller selection rules, and the Fermi theory for forbidden spectra.

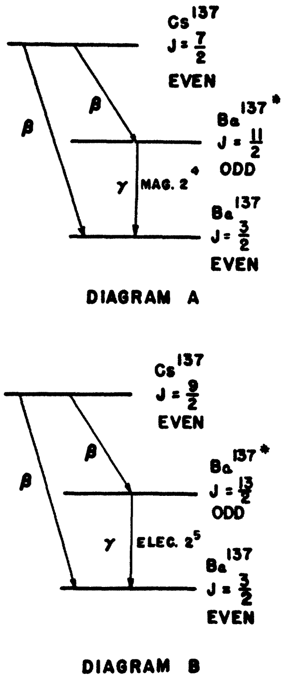

FIG. 5. Spin and parity assignments to the levels in the decay scheme of Cs^{137}.

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