Precise Shape Measurements of Beta Spectra of Y^{90} and $Y^{91\dagger}$

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Precise measurements of the detailed shapes of the beta spectra of Y^{91} and Y^{90} were made in order to determine better to what extent the theory of beta decay in its present approximation is adequate to describe the distributions which are observed for these *unique*, $\Delta I = 2$, party changing transitions. The use of an integrally biased semiconductor as the detector in a high resolution magnetic spectrometer provided an extremely low background which facilitated the accurate determination of the end points with a minimum amount of extrapolation. A proper end-point determination is necessary for a meaningful shape-factor analysis. In both cases, it was found that the experimental shape factor shows a small but definite deviation from the theoretical expectation. For a theoretical shape factor, $L_0 q^2 + 9L_1$, with L_0 and L_1 based on exact electron radial wave functions including the effects of a finite nuclear radius, the necessary correction can be supplied by an empirical factor of the form $1+b/W$ with $0.3 \le b \le 0.4$. The end points of the Y⁹¹ and Y⁹⁰ spectra were determined to be 1.545 ± 0.005 MeV and 2.273 ± 0.005 MeV, respectively.

I. INTRODUCTION

 \mathbf{B}^Y making use of improved techniques of measure-
ment and of analysis of the data, a comparison was ment and of analysis of the data, a comparison was made of the beta spectrum shapes of the once-forbidden transitions in the decays of Y⁹¹ and Y⁹⁰ with the *unique* distribution predicted by the present approximation of the theory. Recent advances in technology have made it possible to obtain more definitive measurements of subtle energy-dependent variations of the shape factors. Furthermore the availability of tables of exact electron radial wave functions (ERWF), including corrections for a finite nuclear radius, now permit a better comparison between theory and the experimental measurements.

For beta transitions which involve a spin change of two units and a parity change, the theory of forbidden beta decay predicts that the transition can proceed only via the G-T part of the interaction. In the "normal" approximation only the B_{ij} matrix element governs the decay, and the shape of the beta spectrum is predicted to be *unique* and of the form $S \sim p^2 + q^2$, where *p* is the electron momentum, $q = W_0 - W$ is the neutrino momentum, and W_0 is the end-point energy, all in relativistic units.

When the *unique* once-forbidden shape was first identified¹ in the decay of Y^{91} , the accuracy of the measurements was such that the substitution of $S = p^2 + q^2$ rather than $S = 1$ in the beta distribution equation led to a reasonable linearization of the Fermi-Kurie (F-K) plot of the data. At that time, the peculiar beta spectrum of RaE was the only clear example of a distribution that could not be fitted by the straight line F-K plot which was expected for the spectra of allowed transitions, but not, in general, for those of forbidden transitions. The observation of the *unique* shape for Y 91 , therefore, gave considerable support to the general formalization of the theory of forbidden beta decay as well as experimental evidence for the validity of the

G-T selection rules. It also confirmed the expectations of the then recently proposed nuclear shell model, which predicted a $\Delta I = 2$, parity changing transition between the ground states of Y⁹¹ and Zr⁹¹. A similar *unique* shape was found^{2,3} soon thereafter for the spectrum of Y^{90} , and many other cases of *unique* shapes were reported subsequently.

It was soon realized that a better fit to the experimental data could be obtained with a modified shape factor⁴ of the form $S = q^2 L_0 + 9L_1$. L_0 and L_1 are functions of *Z* and *W* which can be obtained from tables that include finite de Broglie wavelength corrections but which are not corrected for the finite nuclear radius. Essentially equivalent formulations were offered by Davidson⁵ and by Kotani and Ross,⁶ who presented a table for the parameter $\lambda_1 \approx 9L_1/p^2L_0$ in the expression $S = q^2 + \lambda_1 p^2$.

Detailed beta-spectrum measurements have suggested⁷ the necessity for an additional empirical correction factor in order to describe shapes obtained for many beta spectra under very ideal experimental conditions. One of the original observations of this deviation from the theoretical prediction was for the spectrum of Y⁹⁰. Because of the interest at that time in possible Fierz interference, a factor of the form $1+b/W$ was used to fit the experimental data. Although the possibility of Fierz interference has since been ruled out on other grounds, this form has been retained for the empirical correction factor. Other forms such as $1+aW+b/W$ $-cW²$ might give even closer fits to the data and might even have some justification on the basis of a particular theoretical model. However, it has so far been sufficient

f This work was supported by the U. S. Office of Naval Re-

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³ L. M. Langer and H. C. Price, Jr., Phys. Rev. 76, 641 (1949).
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^{1955),} Chap. 10, p. 304. 5 J. P. Davidson, Phys. Rev. 82, 48 (1951).

⁶ T. Kotani and M. Ross, Phys. Rev. 113, 662 (1959).
⁷ O. E. Johnson, R. G. Johnson, and L. M. Langer, Phys. Rev.
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Feuvrais, J. Phys. Radium 18, 559 (1957).

FIG. 1. Relative sensitivity of the solid-state detector. The ratio of the counting rate obtained with the solidstate detector to that obtained with the proportional counter is plotted against the energy. The circles represent data obtained with the semi-conductor detector cooled with liquid nitrogen. The triangles represent data obtained at room temperature.

to fit the data with only the one parameter *b,* with $0.2 \le b \le 0.4$.

Whereas the earlier results showed a deviation at low energies from the expected F-K plot, the data were not sufficiently reliable to be definitive about the details of the shape factor over an extended energy range. In order to make a shape-factor plot of the experimental data, $N/p^2F(W_0-W)^2$ versus \tilde{W} , it is necessary to first determine a value for W_0 . Now, in a case where the shape of the spectrum is known, such as the statistical shape for that of an allowed transition, one has only to extrapolate a straight line to obtain the intercept, W_0 . This, of course, was the original intention for introducing the F-K plot. However, if the exact shape of the spectrum is unknown, uncertainties arise in making an extrapolation to the maximum energy. The best procedure is, therefore, to obtain significant data as close as possible to the end point.

This ideal technique is limited by the relatively low counting rate near the end of the spectrum. This limitation is particularly serious if the background counting rate is significant, in which case an accurate knowledge of the variation of background with the energy setting of the spectrometer becomes necessary. This quantity is difficult to determine because the contribution to the background from bremsstrahlung arising from electrons striking slit edges is a function of the spectrum itself. It is much more satisfying to have an experimental arrangement in which the background and scattering are so small that uncertainties in the energy dependence are of no consequence. Such a situation was achieved in the present experiment by taking advantage of the inherently low sensitivity to gamma rays of a surface barrier semiconductor used as an integrally biased detector in the 40-cm radiusof-curvature, shaped magnetic-field, 180° focusing spectrometer.

A further restriction placed on the final evaluation of the end point is that the resulting shape-factor plot remain finite as both *N* and *Wo— W* approach zero. The execution of this criterion was facilitated by means of the Indiana University IBM-709 computer.

The results of the present measurements yield shapefactor plots for the spectra of Y⁹¹ and Y⁹⁰ which are not described completely by the theoretical *unique* shape factor in any of the usual forms. The theoretical shape factor using the exact radial wave functions also is inadequate. For both Y^{90} and Y^{91} , the deviation can be fitted by an empirical factor of the form $1+b/W$, with $0.3 \le b \le 0.4$.

II. EXPERIMENTAL PROCEDURES

Recent advances in technology have enabled us to make more precise measurements on the spectral shapes of $Y⁹¹$ and $Y⁹⁰$ than have been possible in the past. Earlier investigations7,8 were limited by some or all of the following: relatively high backgrounds, scattering of electrons in the measuring instrument, poor resolution. These factors contributed distortion effects which were not negligible in the region near the end point of the beta spectrum where the counting rates are low.

In this study, as in the recent work concerning low intensity spectra,⁹ the introduction of a semiconductor detector used in conjunction with the magnetic spectrometer reduces the background to a noncritical level. Because of the semiconductor's small size and its insensitivity to bremsstrahlung, the background counting rate is only 2-3 counts/min. Furthermore, control experiments indicate that the counting rate arising from scattering and bremsstrahlung from the detector slit is less than 1 count/1000 real counts at any point in the spectrum.

The high resolution of the magnetic spectrometer is another important feature which makes it possible to obtain a spectrum with a minimum of distortion. Other investigators⁸ were limited by the fact that the spectrum had to be corrected for the resolution distortion caused by the measuring instrument. This distortion is especially noticeable at the high-energy end of the distribution. Our data were obtained with an instrument having a resolution of $\sim 0.6\%$. The correction for finite resolution is negligible over the main body of the spectrum

⁸ R. T. Nichols, R. E. McAdams, and E. N. Jensen, Phys. Rev.

^{122, 172 (1961).&}lt;br>- D. E. Wortman and L. M. Langer, Phys. Rev. 131, 325 (1963);
L. M. Langer and D. E. Wortman, ibid. 132, 324 (1963); L. M.
Langer, E. H. Spejewski, and D. E. Wortman, ibid. 132, 2616 (1963).

FIG. 2. F-K plot of Y^{91}
 β^- spectrum. In the upper plot, the circles represent data taken with the intense, thicker source. The crosses show that the same general distribution is obtained with a much thinner, less intense source. The lower plot shows how well the theoretical shape factor linearizes the F-K plot.

and is insignificant even at the end point for spectra with such maximum energies as those of Y^{90} and Y^{91} .

Improved statistics and reproducibility of the data obtained with the semiconductor detector give greater confidence in the results. Other improvements include a more stable magnet current supply and a new rotating coil field-measuring device, which permit reproducible magnetic-field measurements and electron-momenta determinations of better than 0.1% at each point.

A. Magnetic Spectrometer

The Y⁹¹ and Y⁹⁰ beta spectra were studied in detail in the high resolution, 40-cm radius of curvature, shaped magnetic-field spectrometer.10,11 It was operated at a resolution of $\sim 0.6\%$, with a source width and a detector slit width of 4 mm.

An integrally discriminator-biased-semiconductor radiation detector, of the silicon surface-barrier type, was used throughout the measurements. The detector was operated at a temperature of -196° C. This low temperature considerably reduces the intrinsic noise of the detector, and hence allows measurements to be extended into the lower energy region. As a check to see if the sensitivity of the cooled semiconductor detector is energy-independent, the spectrum of the same source under identical conditions was measured in the spectrometer by both the semiconductor detector and a proportional counter. The proportional counter was of the end-window type with a loop anode. The counterwindow cutoff was for 9-keV electrons. Previous studies have shown that there is no inherent energy dependence in the sensitivity of the proportional counter over the region of interest.¹²

The ratio of the counting rate per momentum interval, *N(Hp),* measured with the semiconductor detector to that measured with the proportional counter is shown in Fig. 1. For comparison, the same ratio is plotted for data taken with the semiconductor detector at room temperature. Because of the increased noise level at the elevated temperature, the discriminator bias must be set at a higher voltage; thus the efficiency for detections begins to fall off at a higher energy. It is apparent that there is no need for a correction for any energy dependence of the sensitivity of the semiconductor for energies above ~ 90 keV when it is operated at liquidnitrogen temperature.

B. Sources

Two Y⁹¹ sources were prepared. The thickness of source 1 was $\langle 0.1 \text{ mg/cm}^2$ and that of source 2 was <0.7 mg/cm² . The thin source served to check whether the spectrum obtained with the thicker source was being distorted by scattering of the negatrons within the source material. The agreement between the thick- and thin-source data gave assurance that the spectrum was not being influenced by source thickness for all energies above \sim 100 keV.

The Y⁹¹ was received in the form of carrier-free YCl₃ from Oak Ridge National Laboratory. Each source was prepared by liquid-depositing the activity onto an \sim 20 μ g/cm² Zapon film, which was supported by a 0.9 mg/cm² aluminized Mylar backing. Insulin was

¹⁰ L. M. Langer and C. S. Cook, Rev. Sci. Instr. **19,** 257 (1948). 11 D. C. Camp and L. M. Langer, Phys. Rev. **129,** 1782 (1963).

¹² J. H. Hamilton, L. M. Langer, and W. G. Smith, Phys. Rev. **112,** 2010 (1958).

FIG. 3. Shape-factor plot of the beta spectrum of Y 91 . Neither the theoretical once-forbidden *unique* shape, $S_B \sim 9L_1 + L_0 q^2$, nor Kotani's approximation,
 $S_K \sim \lambda_1(p) p^2 + q^2$, fits the experimental data. The influence of an empirical correction factor, $1+b/W$, is also shown.

used¹³ to define the source area. Each source was dried and covered with a thin $(\sim 20 \ \mu g/cm^2)$ Zapon film.

The Y⁹⁰ was received in the form of carrier-free YCl₃ from ORNL. A source of $\langle 0.7 \text{ mg/cm}^2$ was prepared in the same manner. It was not felt necessary to prepare a thinner Y⁹⁰ source since the Y⁹¹ experiments showed no distortion above \sim 200 keV for the same source thickness.

Earlier tests¹² have indicated that such source thickness and backing do not measurably distort the distribution for energies above ~ 200 keV. Since in the present experiments, it was the intention to work with the better statistics obtainable from somewhat thicker sources, the additional complication of evaporating sources on fragile, thin Zapon backings was avoided. The analysis of the data is confined to the energy region above 200 keV.

III. RESULTS

A. Y⁹¹

The data obtained with the more intense source were used in the analysis of the beta decay of Y⁹¹ . Several runs were made through the spectrum over a period of 2 days, and the data were corrected for a half-life of 58 days. Each datum point has a statistical accuracy of better than 1% . Figure 2 shows an F-K plot of the data. The upper curve represents the data uncorrected for the theoretical $\Delta I=2$, parity changing shape. The lower curve shows the apparent linearization obtained by applying the *unique* shape correction factor. In the upper plot, the circles represent data taken with the thicker, more intense source, and the crosses represent the data taken with the thinner source. Since the more intense source is approximately ten times thicker than the thin source, any distortion effects at the low-energy end of the spectrum arising from scattering of negatrons in the source would be expected to be proportionately greater. The data, however, follow the same distribution for both sources above \sim 100 keV. It was thus possible to have confidence in the reliability of the data which could be obtained with better statistics by using the thicker source.

The experimental shape-factor plot is shown in Fig. 3. By requiring that the shape factor remain finite near the end point, an end-point energy of 1.545 ± 0.005 MeV is obtained.

The lower curve in Fig. 3 represents the theoretical once-forbidden, *unique* shape factor calculated by using the exact ERWF values tabulated¹⁴ by Bhalla and Rose. For comparison, Kotani's approximation to the theoretical shape factor,⁶ $\lambda_1(p)p^2+q^2$, is shown as the next higher curve. It is obvious that, under the closer scrutiny afforded by the shape-factor plot, the good fit indicated by the linearization of the F-K plot is limited and does not exploit the full significance of the data. Neither of the theoretical shape factors offers a completely satisfactory fit to the data. Since an empirical correction factor of the form $1+b/W$ has been found necessary to fit the data in other well-measured spectra, it was also applied in this case. The upper curve represents the theoretical shape factor multiplied by $1+0.4/W$, and the next lower curve the theoretical shape factor multiplied by *1+0.3/W.* A value of *b* between 0.3 and 0.4 seems to give the best fit to the data. Using Kotani's approximation, the best fit is obtained for a value of *b* between 0.2 and 0.3.

14 C. P. Bhalla and M. E. Rose, ORNL Report 3207, 1962 (unpublished).

¹³ L. M. Langer, Rev. Sci. Instr. 20, 216 (1949).

FIG. 4. F-K plot of $Y^{90} \beta^-$ spectrum. The upper set of points represents the usual F-K analysis. The lower set represents the data corrected with the theoretical once-forbidden *unique* shape factor.

TABLE I. Experimental values of shape factor, $S_E \sim N/p^2q^2F$, for β^- spectrum of Y⁹¹.

W	S_{E}	W	S_{E}	W	$S_{\pmb E}$
1.246	4044	2.279	2874	3.200	3703
1.319	3779	2.282	2861	3.357	3945
1.388	3578	2.454	2911	3.382	3937
1.390	3570	2.476	2891	3.497	4251
1.551	3325	2.626	3053	3.538	4396
1.562	3310	2.662	3045	3.560	4408
1.722	3146	2.742	3128	3.590	4418
1.901	2975	2.810	3226	3.667	4733
1.919	2928	2.833	3185	3.726	4744
2.086	2893	3.014	3438	3.739	4843
2.107	2848	3.019	3452	3.841	5116
2.194	2877	3.189	3743	3.906	5371
				3.914	5375

B. Y⁹⁰

Several runs were made through the spectrum over a period of 2 days, and the data were corrected for a halflife of 64.5 days. Figure 4 shows the F-K plot of the experimental data. The upper curve represents the usual F-K plot of the data, and the lower curve shows the data corrected for the theoretical once-forbidden, *unique* shape. Each datum point shown has a statistical accuracy of better than 1% . The values for the Coulomb function, F, were obtained from Feister's tables.¹⁵

The experimental shape-factor plot is shown in Fig. 5. The end-point energy is determined to be 2.273 ± 0.005 MeV. Again, the lower curve is the once-forbidden, *unique* shape determined by using the exact ERWF compiled by Bhalla and Rose; the next higher curve is Kotani's approximation. As in the case of \overline{Y}^{90} , neither of

15 Natl. Bur. Std. (U.S.) Applied Mathematics Series, No. 13 (1952), Tables for the Analysis of Beta Spectra.

the theoretical curves fits the data completely. Applying the additional correction factor of the form $1+b/W$ to the theoretical $\Delta I=2$, parity changing shape results in a much better fit. Here again, the value of *b* which best fits the data is between 0.4 (upper curve), and 0.3 (next lower curve). Using Kotani's approximation, the value of *b* needed to fit the data is between 0.2 and 0.3, in good agreement with the earlier measurements and analysis.⁷ Recent measurements¹⁶ on the *unique* once-

TABLE II. Experimental values of shape factor, $S_E \sim N/p^2q^2F$, for β^- spectrum of Y⁹⁰.

W	S_{E}	W	$S_{\it{E}}$	W	$S_{\it{E}}$
1.398	2459	3.215	1631	4.268	2016
1.559	2228	3.222	1617	4.350	2072
1.737	2032	3.257	1625	4.433	2150
1.914	1880	3.295	1624	4.441	2144
1.926	1887	3.326	1642	4.518	2197
2.104	1775	3.375	1643	4.604	2259
2.107	1799	3.483	1672	4.610	2280
2.285	1705	3.562	1697	4.673	2286
2.291	1699	3.565	1701	4.762	2379
2.472	1662	3.663	1726	4.784	2433
2.476	1665	3.741	1753	4.824	2431
2.477	1650	3.743	1758	4.866	2517
2.582	1627	3.826	1790	4.940	2540
2.664	1608	3.909	1821	4.946	2518
2.684	1614	3.917	1824	4.954	2568
2.761	1612	4.002	1900	5.003	2596
2.835	1590	4.092	1924	5.024	2587
2.841	1594	4.095	1933	5.029	2593
2.930	1600	4.103	1954	5.097	2720
3.024	1610	4.155	1947	5.103	2726
3.025	1576	4.156	1957	5.186	2755
3.118	1595	4.169	1943	5.187	2766
3.202	1605	4.262	2028	5.239	2841
				5.272	2926

¹⁶ L. M. Langer, E. H. Spejewski, and D. E. Wortman, Phys. Rev. **133**, B1145 (1964).

FIG. 5. Shape-factor plot of the beta spectrum of Y⁹⁰. Neither the theoretical once-forbidden *unique* shape, $S_B \sim 9L_1 + L_0q^2$, nor Kotani's approximation, $S_K \sim \lambda_1(\rho) \rho^2 + q^2$, fits the experimental data. The influence of an empirical correction factor, $1+b/W$ *,* is also shown.

forbidden spectrum in the decay of Rb⁸⁴ also indicated a best fit to the data could be obtained with a *1+0.2/W* correction applied to the Kotani approximation. Tables I and II list the unnormalized values for the experimental shape-factor plots of Y⁹¹ and Y⁹⁰ shown in Figs. 3 and 5.

IV. DISCUSSION

The purpose of this investigation was to obtain more precise experimental determinations of the shape factors of the once-forbidden, *unique* β ⁻ decays in Y⁹¹ and Y⁹⁰ than have previously been experimentally possible. It was the intention to determine in better detail the energy dependence of any possible deviation from the shape expected from the predictions of the theory in its present form. Such a deviation has been reported for the Y⁹⁰ spectrum.⁷

A much more precise determination of the shapes of the Y⁹¹ and Y⁹⁰ spectra has been made in the present case. This was possible because of the noncritical background of the semiconductor detector, the low internal scattering of the spectrometer, and the high resolution of the instrument. Hence, the region near the end point of the beta spectrum could be examined definitively. This allows a better determination of the end-point energy, which in turn is critical to the analysis.

The results of these measurements show that the data exhibit a definite deviation from the theoretical once-forbidden *unique* shape throughout each spectrum. These deviations may be fitted with an empirical correction factor of the form $1+b/W$, with $0.3 \leq b \leq 0.4$.

Deviations of this same form have been observed for many other beta spectra.¹⁷ Several attempts have been

" J. I. Rhode and O. E. Johnson, Phys. Rev. **131,** 1227 (1963); R. P. Sharma and H. G. Devare, *ibid.* **131,** 384 (1963); W. H. made to provide a theoretical explanation for the observed deviations, particularly in the case of allowed transitions. Some of these are based on intriguing and exotic assumptions such as additional types of neutrinos,¹⁸ modifications of the $V-A$ law of beta decay,¹⁹ and contributions to the interaction Hamiltonian proportional to the derivatives of the wave functions.²⁰ A simpler and perhaps more promising explanation has been offered by Bhalla.²¹ Using exact ERWF, he has shown that for allowed decays, the contribution from matrix elements which are usually relegated to the twice-forbidden approximation, may contribute measurably and in a way that results in the observed deviations. It is possible that a detailed calculation²² may indicate that third-forbidden matrix elements contribute to once-forbidden beta decay in a non-negligible way and so account for the deviations in the *unique* spectra.

ACKNOWLEDGMENTS

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22 Dr. Bhalla has indicated his intention to make such a calculation.

Brantley, W. B. Newbolt, and J. H. Hamilton, Bull. Am. Phys. Soc. 9, 348 (1964); see references given in Ref. 11 for other examples.

¹⁸ C. L. Hammer and R. H. Good, Jr., Phys. Rev. **117,** 889 (1960); M. Nakagawa, H. Okonogi, S. Sakata, and A. Toyoda (private communication).

¹⁹ B. Eman and D. Tadic, Glansik Mat. Fiz. Astron. Ser. II 16, 89 (1961); J. M. Pearson, Phys. Rev. 126, 1100 (1962).