# Evidence for Small Deviations in the Allowed Positron Spectrum of $Zr^{89}$†

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The decay of $Zr^{89}$ has been carefully studied with magnetic and scintillation spectrometers with special emphasis on the detailed shape of the positron spectrum. The decay scheme has been verified. The positron decay is by a single, allowed group followed by a single 915-kev gamma ray. The $Zr^{89}$ spectrum has a nonstatistical shape corresponding to an excess of low-energy beta particles. Theoretical refinements for screening and finite deBroglie wavelength were applied but were found to be much too small to explain the observed deviation from a statistical spectrum. The same shape factor that was found to fit the $In^{114}$, $Y^{90}$, $P^{32}$, and $Na^{22}$ data (in addition to the once forbidden, unique shape factor for $Y^{90}$) also fits the $Zr^{89}$ data, i.e., $(1+b/W)$ with $0.2\leq b\leq 0.4$. It is significant that the deviation has the same direction and approximate magnitude as was found for the electron spectra.

## INTRODUCTION

SMALL deviations from the predictions of the present theory of beta decay have been reported[1,2] in the electron spectra of $In^{114}$, $Y^{90}$, and $P^{32}$ and the positron spectrum of $Na^{22}$. The measured shape factors of these four spectra were fitted with an empirical equation of the form $(1+b/W)$ with $0.2\leq b\leq 0.4$. $W$ is the total electron energy. The sign of $b$ is the same for both positrons and electrons. The dependence of the sign of $b$ on the type of the beta particle is significant. Any theoretical explanation of this effect must account for this similarity in the spectra of positrons and electrons.

The above correction term has until now been observed in only one allowed positron spectrum, that of $Na^{22}$. To interpret the observed $Na^{22}$ shape factor as arising from the same origin as that responsible for the other three spectra may be questioned somewhat because of the high log $ft$ value of 7.4 for this allowed transition. It was pointed out, however, that it appears extremely unlikely that even cross terms of the allowed with the second forbidden matrix elements could contribute significantly even with the high $ft$ value.[2]

The allowed positrons, $E_0=543$ kev, emitted in the decay of $Na^{22}$ populate a level at 1.28 Mev in $Ne^{22}$; this level is de-excited by the emission of a prompt 1.28-Mev gamma ray. Recently, Steffen found a small anisotropy in the beta-gamma directional correlation.[3] Steffen suggested that the directional correlation may be explained by cross terms of the allowed matrix elements with the twice forbidden ones and that this might be related to the nonstatistical shape.

These two effects may well be related but their origin is uncertain. It is possible that the deviation from the present predictions observed in $Na^{22}$ may arise from an origin other than that responsible for the effect observed in $In^{114}$, $Y^{90}$, and $P^{32}$.

Because of the importance of establishing whether a shape correction factor such as $(1+b/W)$ is required for allowed positron spectra with a $b$ of the same sign as observed for the above electron spectra, other decay schemes were examined as to their suitability for providing definitive measurements of a positron spectrum. One stringent requirement is that the source be uniform and thin. $Zr^{89}$, which has been reported[4] to have a single allowed positron spectrum of reasonably high energy, meets the requirements of source preparation, since it can be prepared carrier-free.

The decay of $Zr^{89}$ has been carefully investigated. The accepted decay scheme[4,5] has been verified. The positron spectrum was measured with precision and a nonstatistical shape was observed. The experimental shape factor may be fitted with an equation of the form $(1+b/W)$ with $0.2\leq b\leq 0.45$. Fits were also obtained with an equation of the form $(1+c_1W+c_2W^2)$. The direction and magnitude of the deviation from the statistical shape is the same as observed in the other spectra.[1,2]

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[1] O. E. Johnson, R. Johnson, and L. M. Langer, Phys. Rev. **112**, 2004 (1958).

[2] J. H. Hamilton, L. M. Langer, and W. G. Smith, Phys. Rev. **112**, 2010 (1958). (Other references on these effects listed here.) [Also see H. Daniel, Nuclear Phys. **8**, 191 (1958).]

[3] R. M. Steffen, Phys. Rev. Letters **3**, 277 (1959).

[4] F. Shore, W. Bendel, and R. Becker, Phys. Rev. **83**, 688 (1951); F. Shore, W. Bendel, H. Brown, and R. Becker, Phys. Rev. **91**, 1203 (1953).

[5] D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958); B. S. Dzelepow and L. K. Peker, *Decay Schemes of Radioactive Nuclei* (Academy of Sciences of the U. S. S. R. Press, Moscow, 1958).

This result not only gives support to the previous work[1,2] that there are deviations from theory in allowed beta spectra but offers more definitive proof that these deviations are independent of the sign of the emitted beta particles.

## EXPERIMENTAL PROCEDURES

### Magnetic Spectrometer

The positron spectrum of $Zr^{89}$ was measured in a 40-cm radius of curvature, 180-degree focusing, shaped magnetic field spectrometer.[6] Modifications of the equipment and the procedures of operation have been discussed in detail previously.[1,2,7] The question of possible instrumental distortions of a spectrum has been carefully considered and no evidence for such has been found.[1,2,7]

The loop anode proportional counter with a thin, unsupported, aluminum-coated Zapon window (cutoff$\simeq$6 kev) was used as the detector. The normal beam defining baffle was used with a source width and counter acceptance slit each of 5 mm.

### Scintillation Spectrometer

The gamma spectrum was examined with a 100-channel analyzer. A $1\frac{3}{4}$ in.$\times$1 in. NaI crystal mounted on a Dumont 6292 photomultiplier was used as the detector. $Na^{22}$ and $Ce^{144}$ were used as calibration sources. The gamma spectrum was also measured on a single-channel analyzer.

### Sources

$Zr^{89}$ was made by the reaction $Y^{89}(d,2n)Zr^{89}$ with 11-Mev deuterons in the Indiana University cyclotron. After bombardment, the $Y_2O_3$ target material was dissolved in concentrated HCl. The target solution was passed through a Dowex 1 anion exchange resin column. The $Zr^{89}$ was retained on the column at this high HCl concentration; the yttrium was not retained under these conditions. The $Zr^{89}$ was then eluted with 4*M* HCl. The separation of zirconium and yttrium was checked by eluting with concentrated HCl the 16 sec isomer of $Y^{89}$, the daughter of $Zr^{89}$, from the $Zr^{89}$ parent which was retained on the column.

The sources were prepared from different cyclotron bombardments. The same chemical procedure was followed in each case. Source number one was prepared by depositing the separated $Zr^{89}$ in liquid form on an aluminum-coated laminate of Zapon and LC600 ($\simeq$150 $\mu$g/cm$^2$ thick). The source was carrier-free with no visible mass. The source thickness was estimated as $<10$ $\mu$g/cm$^2$. It was covered with a $<10$ $\mu$g/cm$^2$ Zapon film.

After the initial measurements, procedures were developed for the thermal evaporation of the $Zr^{89}$. Source number two was prepared by evaporating the activity onto a 180 $\mu$g/cm$^2$ aluminum foil. The chemistry gave carrier-free $Zr^{89}$ which yielded an invisible source after thermal evaporation in vacuum. The source thickness was $<10$ $\mu$g/cm$^2$. It was also covered with a $<10$ $\mu$g/cm$^2$ film.

Previous studies[2] have shown that source backings of 180 $\mu$g/cm$^2$ will produce distortions in beta spectra below $\simeq$100 kev. Also these studies indicate no other measurable distortions should be expected from sources prepared in the above manner. Repeated attempts to evaporate $Zr^{89}$ onto thinner backings ($\simeq$20 $\mu$g/cm$^2$) failed because of the very high evaporation temperature required.

## TREATMENT OF DATA

Five measurements of the positron spectrum were made from 100 to approximately 840 kev (two with source one and three with source two). Each measurement consisted of two sets of data which covered the above energy range and which were obtained on different cycles of the magnetic field. All of these measurements were made under similar conditions. Almost every experimental point has a one percent statistical accuracy in the counting rate.

The average reported half-life of $Zr^{89}$ is 78.5 hours.[4,8] A fraction of source one was followed for over 9 half-lives and decayed with a half-life of 79.0$\pm$0.5 hours. Source two was followed for over 2 half-lives with the same result. The data were corrected for a half-life of 79.0 hours.

Fermi-Kurie (F-K) plots of the data were made with the aid of tables.[9] The data were corrected for outer screening which arises from the modification of the nuclear electrostatic potential by the orbital electrons. These corrections were made for $Z=40$ and an "outer screening potential" of $V_0=3.9$ as well as for $V_0=6.5$, the upper limit (see reference 9). The effect of finite deBroglie wavelength was considered but is negligible over the entire range covered by the measurements.

## RESULTS

The Fermi-Kurie plots of the data from source one (runs one and two normalized) and from source two (runs one and two normalized) are seen in Fig. 1. The straight lines are least squares fits for $W\geq 1.95$ $m_0c^2$. The screening correction has been applied for $V_0=3.9$. The F-K plots exhibit an excess of low-energy electrons. However, the high-energy region is only slightly nonlinear. From the individual least squares fits of the high-energy data ($W\geq 1.95$), a $W_0$ value of 2.755$\pm$0.005 $m_0c^2$ was obtained. This corresponds to an endpoint

[6] L. M. Langer and C. S. Cook, Rev. Sci. Instr. **19**, 257 (1948).
[7] J. H. Hamilton, L. M. Langer, R. L. Robinson, and W. G. Smith, Phys. Rev. **112**, 945 (1958).
[8] L. Katz, R. G. Baker, and R. Montalbetti, Can. J. Phys. **31**, 250 (1953).
[9] *Tables for the Analysis of Beta Spectra*, National Bureau of Standards Applied Mathematics Series No. 13 (U. S. Government Printing Office, Washington, D. C., 1952).

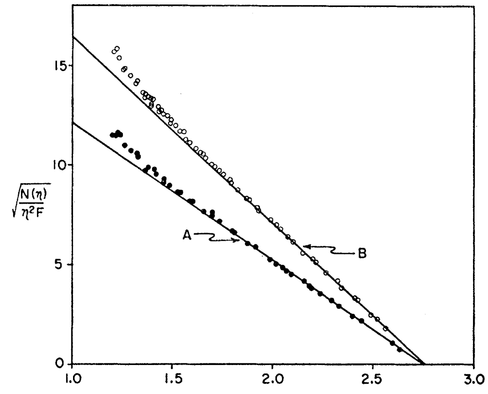


FIG. 1. Fermi-Kurie plots of the $Zr^{89}$ data, $A$ from source 1 and $B$ from source 2.

energy of 897±7 kev.This value includes possible uncertainties in the absolute calibration of the spectrometer.

Measurement three with source two was made approximately one half-life after measurement two. This was done to check that all parts of the spectrum were decaying with the same half-life. The statistical deviations in the counting rates were slightly more than 1% on many of the points of run three so these data were not included in the further analysis.

The outer screening correction is in the right direction to explain the excess of low-energy beta particles in positron decay (but in the wrong direction for the electron spectra reported in reference 1). However, the correction is much too small to account for the observed results. Even when the maximum possible correction ($V_0=6.5$) is made, the low-energy data are changed only slightly.

The nonlinear F-K plot indicates an energy dependent shape factor. The energy dependence is seen more clearly in Fig. 2 where $C(W) \propto N(\eta)/\eta^2 F(W_0-W)^2$ is plotted against $W$ for $W_0=2.755$ $m_0c^2$. The error bars represent the 1% standard deviation in the counts per unit momentum interval, and are the same for almost every point.

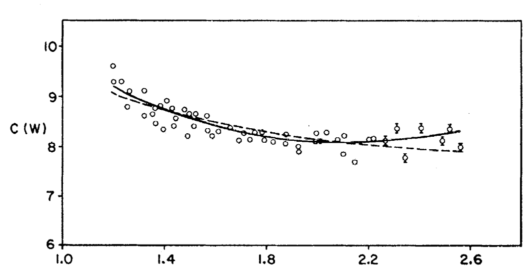


FIG. 2. Shape factor, $C(W)=N/\eta^2F(W_0-W)^2$, plot for $Zr^{89}$ (source 2 data). $W_0=2.755$ $m_0c^2$. The dashed curve is $k(1+0.37/W)$ and the solid curve is $k(1-0.39W+0.09W^2)$. The coefficients were determined by least-squares fits to the data.

The deviation in the F-K plot was considered as if it were a possible second beta group. A straight line fit to the high-energy data was subtracted from the total. The endpoint energy of such a second group would be between 1.7 and 2.1 $m_0c^2$ with an intensity of 3 to 5% relative to the intensity of the total positron spectrum. If this assumption were correct, one should expect to find gamma radiation or internal conversion electrons which correspond to a transition energy of 0.34–0.55 Mev.

The gamma spectrum of $Zr^{89}$, as measured on the 100-channel analyzer, is seen in Fig. 3. A similar spectrum was also obtained with a single-channel analyzer. A single gamma ray at 915 kev and annihilation radiation are clearly seen. The annihilation radiation of $Na^{22}$, with a very small flat Compton distribution from the 1.28-Mev gamma ray subtracted, is also given for comparison. The gamma spectrum below 700 kev was studied in greater detail with the 100-channel analyzer (Fig. 4). Here the $Na^{22}$ gamma spectrum does not have the Compton distribution from its 1.28-Mev gamma ray subtracted. No evidence for a gamma ray with energy different from 511 kev was observed. An upper limit of 1% was set for the intensity of such a gamma ray relative to that of the annihilation radiation.

The electron spectrum from 300 to 550 kev was carefully examined in search of possible conversion lines from transitions in the above energy range. No evidence was found for any conversion lines in this region.

## DISCUSSION

The definitive establishment of the decay scheme is essential to an analysis of the data. Figure 5 gives the reported decay scheme.[4,5] Previous evidence for the correctness of this scheme is well determined. The ground-state spin of $Y^{89}$ has been measured[10] as $\frac{1}{2}$ and the shell model predicts odd parity. Shore, Bendel, Brown, and Becker have carried out probably the most thorough investigation of the decays of $Zr^{89}$ and $Zr^{89m}$ and have discussed the decay scheme in detail.[4] From

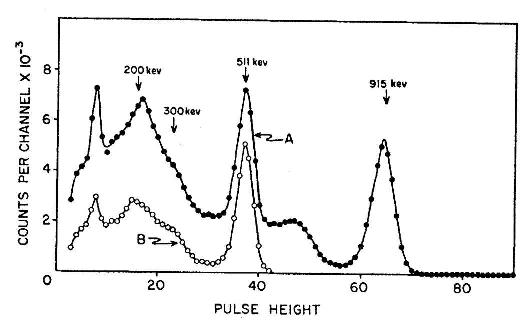


FIG. 3. Gamma spectrum of $Zr^{89}(A)$ and $Na^{22}(B)$ for energies less than 1.15 Mev as measured on a 100-channel analyzer. The Compton distribution from the 1.28-Mev gamma ray of $Na^{22}$ is subtracted from the $Na^{22}$ data.

[10] J. E. Mack, Revs. Modern Phys. 22, 64 (1950).

measurements of the $K$ to $L$ ratios of the 915-kev transition in $Y^{89}$ and the 588-kev transition in $Zr^{89m}$ as well as the conversion coefficient of the 915-kev transition, the branching ratios of the positron spectra from $Zr^{89}$ and $Zr^{89m}$, the spins and parities of the 9.5-kev level in $Y^{89}$, the ground state of $Zr^{89}$ and the 588-kev level, $Zr^{89m}$, are uniquely obtained[4] as $9/2+$, $9/2+$, and $1/2-$, respectively (see Fig. 5). The analysis which leads to these assignments is given in reference 4 and a review here is unnecessary.

The log $ft$ value for the allowed positron decay from $Zr^{89}$ to the 915-kev level is 6.1. This is only slightly above the range of log $ft$ values for normal allowed transitions. It would be extremely unlikely for cross terms between allowed and twice forbidden matrix elements to contribute to this decay.

Another important question is whether any levels higher than 915 kev are populated, since the observed low-energy deviation in the F-K plot might then correspond to a second group. If such were the case, the approximate endpoint energies and intensities of possible positron transitions to such levels would be 350–560 kev and 3–5%, respectively, as determined from an analysis of the F-K plot as two groups.

To populate levels above 915 kev with any observable intensity, the beta transition should be allowed or possibly once forbidden, nonunique. The unique transition which is several orders of magnitude slower than allowed transitions would not compete favorably here even if the decay energy were the same, but with the decay energy lowered by a factor of approximately two, this possibility can be ruled out. This would indicate that only high spin states would be populated from the $9/2+$ ground state of $Zr^{89}$. Transitions from these high spin states would be expected to go through the $9/2+$ level to the ground state rather than to the $1/2-$ ground state directly. It is worth noting that if a low spin state such as $5/2+$ were populated this should decay directly to the ground state by $M2$ radiation but with some competition from $E2$ radiation to the $9/2+$ level. An upper limit of 1% has been set for any gamma ray with energy between 0.95 and 2.0 Mev in the $Zr^{89}$ decay.[4] Such high-

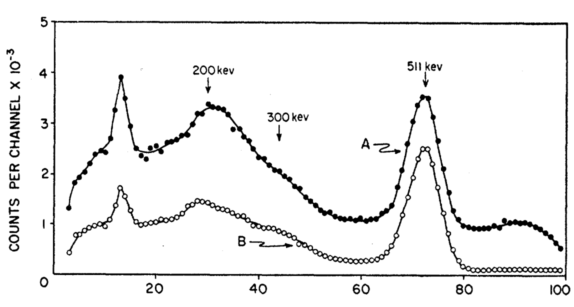


FIG. 4. Gamma spectrum of $Zr^{89}(A)$ and $Na^{22}(B)$ for energies less than 0.7 Mev as measured on a 100-channel analyzer. The Compton distribution from the 1.28-Mev gamma ray of $Na^{22}$ is not subtracted from the data.

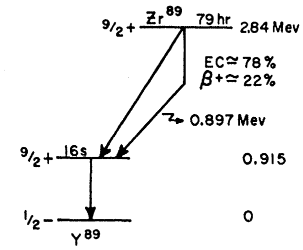


FIG. 5. Decay scheme of $Zr^{89}$.

energy transitions would be weakly converted so 1% is a good upper limit for the total intensity of depopulation of such levels to the ground state. The possible lower energy $E2$ transition is ruled out below.

The measurements of the gamma spectrum indicated no gamma rays with energy between 100–430 kev with an intensity $\geq 1\%$. The conversion electron spectrum offers more conclusive evidence that, indeed, no levels above 915 kev are populated. Even in the complicated decay of $Nb^{90}$, $K$ and $L$ conversion electrons from a 90% $E2$ transition with gamma-ray intensity $\simeq 1\%$ and energy 372 kev were easily observed in this spectrometer.[11] Hence, the limit of 1% for the intensity of population of levels above 915 kev is lowered still further.

Moreover, the positron decay from $Zr^{89}$ to such levels in $Y^{89}$ would be accompanied by large amounts of $K$ capture. (There is 75–80% $K$ capture to the 915-kev level and higher percentages would be expected for lower energy transitions.) Thus, the 3–5% population from the positron intensity should be increased by 8–15% or more for the $K$ capture contribution in order to estimate the total population of such a level. This makes the evidence quite conclusive that there are no levels populated above 915 kev which could make any contribution to the present measurements, and that there is therefore only one positron group and it is allowed.

These data were analyzed in the same manner as were the earlier results.[1,2] There, a correction of the form $(1+b/W)$ with $0.2\leq b\leq 0.4$ for both positrons and electrons was found to fit the experimental shape factor. The original choice of this form for the shape factor was motivated by the earlier test for Fierz interference. However, these results[1,2] cannot be explained by the Fierz effect which predicts a different sign for $b$ for positrons and electrons because the experimental $b$ has the same sign for both positrons and electrons.

Least-squares fits of $(1+b/W)$ were made to the $Zr^{89}$ data by the IBM 650 electronic computer at Vanderbilt University. The result of the fit to the four individual sets of data was $b=0.37$. When the maximum possible screening correction was used, the least-squares fit to the data gave $b=0.30$. This is not a significant change when one considers the limits on $b$ which correspond to reasonable fits to the $Zr^{89}$ data, $0.25\leq b\leq 0.45$. Figure 2 shows the least-squares fit of the curve $k(1+0.37/W)$ to

[11] N. H. Lazar, G. D. O'Kelley, J. H. Hamilton, L. M. Langer, and W. G. Smith, Phys. Rev. **110**, 513 (1958).

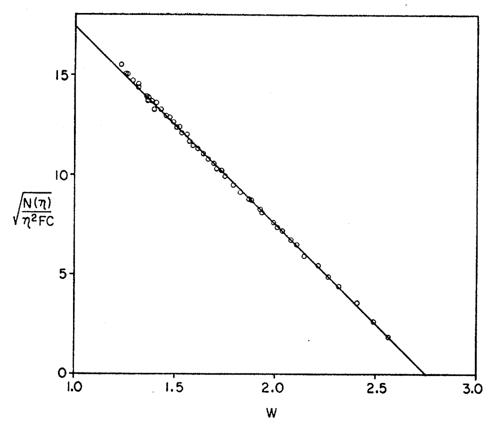


FIG. 6. Fermi-Kurie plot of the $Zr^{89}$ data corrected with $C=k(1+0.37/W)$.

the data. The $Zr^{89}$ positron spectrum with this shape factor included is given in Fig. 6.

Another possible fit to the data was considered. The equation $k(1+c_1W+c_2W^3)$ was least-squares fitted to the data by the computer. This gave possibly an even better fit as is seen in Fig. 2. The best values for $c_1$ and $c_2$ are $-0.39$ and 0.09, respectively. The equation $(1+c_1W+c_2W^2)$ will also fit the earlier results on $Na^{22}$, $In^{114}$, $Y^{90}$, and $P^{32}$.

The $Zr^{89}$ results are further evidence that there are deviations from the predictions of the present theory in beta spectra. More important, it establishes more definitely that the deviations have the same form and direction for both positron and electron decay. The four spectra studied previously[1,2] are pure Gamow-Teller transitions. In the positron decay of $Zr^{89}$ there are both Fermi and Gamow-Teller radiations. The $Zr^{89}$ results may suggest but do not constitute proof that the observed deviations are present in Fermi transitions. The measured shape factor of all five transitions may be fitted by some combination of $(1+c_1W+b/W+c_2W^2)$. It is stressed that this shape factor is not to be confused with that expected for forbidden transitions although the form is the same as that suggested[12] for once forbidden, nonunique decays.

There is still no satisfactory explanation for these observed deviations from the present predictions of the theory of beta decay. The formulation of the theory undoubtedly provides room for suitable refinements. So far, attempts[13,14] at an explanation have either been based on unattractive assumptions or lead to unattractive consequences.

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[12] T. Kotani and M. Ross, Progr. Theoret. Phys. (Kyoto) **20**, 643 (1958).

[13] B. Kuchowicz, Polish Acad. Sci. **7**, No. 94 (1959).

[14] C. L. Hammer and R. H. Good, Jr., Phys. Rev. **117**, 889 (1960).