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Beta Decay of Fe^{59}†

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The decay of Fe^{59} was studied with particular interest in the shape of the beta spectrum and in the comparative half-life of the very weak, highest energy beta group. This group was found to result from 0.30% of the decays and to have an end-point energy of 1.573±0.003 MeV. The experimental data could be fitted with a shape factor $p^2+3.3\ q^2$, consistent in form with that expected for a twice-forbidden $\Delta I=2$ (no) transition. The comparative half-life ($\log ft=10.96$) is relatively short. The two more intense beta transitions are 0.475±0.003 MeV, 51.2%, $\log ft=6.74$ and 0.273±0.005 MeV, 48.5%, $\log ft=5.92$. The energies of the two high-energy gamma transitions, determined from their K internal conversion lines, are 1.101±0.003 and 1.300±0.003 MeV.

I. INTRODUCTION

A STUDY of the beta disintegrations of Fe^{59} is of particular interest because of the fact that the low-intensity, highest energy beta-ray group appears to have a comparative half-life which is much smaller than that usually associated with other $\Delta I=2$, (no) transitions in the twice-forbidden category. Furthermore, in the light of more recent developments, some of the original arguments[1] used to classify the Fe^{59} transition as twice forbidden are no longer valid or forcing. For example, Metzger found that the shape of the beta-ray spectrum of the highest energy group was not that expected for an allowed transition, and that it could not be fitted as either a once-forbidden or a twice-forbidden unique transition. Since it had what was considered to be a high comparative half-life ($\log ft=10.9$), Metzger concluded that the transition must be twice-forbidden nonunique.

It is now known that there exist certain "allowed" transitions, which involve no parity change with a spin change of one or zero, and also many nonunique once-forbidden transitions with high comparative half-lives, comparable to that of Fe^{59}. In many cases, such transitions have been found to give rise to beta spectra with nonstatistical, nonunique shapes.[2,3] Furthermore, the spectrum reported by Metzger for Fe^{59} does not satisfy the requirement that a twice-forbidden nonunique transition should give rise to a shape factor of the form $p^2+\lambda q^2$, where p and q are, respectively, the momenta of the electron and the neutrino and λ is a constant determined by the values of the matrix elements.

In an attempt to understand the nature of the highest energy beta transition of Fe^{59}, a detailed study of the beta spectrum was undertaken. A special effort was made to obtain definitive information on the shape of the beta spectrum, the end-point energy, and the relative intensity of the weak high-energy group. As a result of our measurements, it was found that the shape of the high-energy spectrum is consistent with the theoretical expectation for a nonunique twice-forbidden transition but that the comparative half-life is, indeed, still somewhat low ($\log ft=10.96$).

Earlier investigations[1,4,5] have established the general features of the decay scheme. Fe^{59} decays with a half-life of 45 days, mainly by two inner beta groups of 0.47 and 0.27 MeV to excited states of Co^{59}. In less than 0.05% of the time the decay goes directly to the ground state of Co^{59} with an energy release of 1.57 MeV. It is this weak transition which is the particular interest of the present investigation.

II. EXPERIMENTAL PROCEDURE

The precise determination of the spectrum shape for a transition with the low relative intensity of the high-energy group of Fe^{59} requires a re-examination of the various experimental factors that might lead to the distortion of such a measurement. Effects arising from scattering and background, which may be negligible for routine beta-spectrum measurements, may become of concern in establishing the subtle energy dependence associated with the forbidden nature of such a weak transition. Several control experiments were performed to ascertain the extent of any distortions that might be introduced into the high-energy region of interest by scattering of the electrons from the intense low-energy groups or from the presence of the strong gamma radiation. The analysis was confined to data for which it was clearly established that such effects had negligible influence. The relatively high resolution of the magnetic spectrometer and its inherently low internal scattering were important factors in making the final measurements significant. The use of a semiconductor detector in the magnetic spectrometer was effective in reducing the background to a noncritical level.

A. Spectrometer

The Fe^{59} beta spectrum was studied in a high resolution, 40-cm radius of curvature, shaped magnetic-field

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[1] F. R. Metzger, Phys. Rev. **88**, 1360 (1952).

[2] L. M. Langer and D. R. Smith, Phys. Rev. **119**, 1308 (1960).

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[4] R. L. Heath, C. W. Reich, and D. G. Proctor, Phys. Rev. **118**, 1082 (1960).

[5] *Nuclear Data Sheets*, compiled by K. Way, *et al.* (Printing and Publishing Office, National Academy of Sciences–National Research Council, Washington 25, D. C.).

spectrometer. Certain modifications to improve the original instrument have been incorporated and these have been discussed in previous papers.[3,6–10]

The current for the magnet was supplied by a transistorized dc supply and was monitored by a digital voltmeter. The current was stable to better than 1 part in 10 000 during the time required for any datum point. A precision rotating coil gaussmeter[3] was used to measure the magnetic field.

Independent calibrations of the spectrometer in terms of the K internal conversion lines of Bi^{207}, Cs^{137}, and the F line of ThB agree to better than 1 part in 10 000 and, thus, are indicative of the absence of any energy dependence in the gaussmeter measurements.

The spectrometer was operated at approximately 0.7% resolution with source and detector widths of approximately 4 mm.

B. Detectors

A solid-state radiation detector, of the silicon surface barrier type, was used to measure the high-energy, low-intensity Fe^{59} group. The detector, 5 mm×25 mm, was located behind a 4-mm-wide slit opening in a $\frac{1}{16}$-in.-thick sheet of Ta. The signal from the detector was amplified by a commercial low-noise amplifier system to operate the scaler counters.

The solid-state detector is more useful for the measurement of the outer group than the proportional counter because its small size makes it possible to obtain a much lower background counting rate. In addition, the use of the semiconductor detector obviates the use of high voltages and of the bulky gas flow system which the proportional counter requires. The use of a solid-state detector in a spectrometer offers several other advantages over other types of detectors. There are no problems of thin windows, of counter gas filling, or of counter gas composition. Also, the sensitivity of the solid-state detector is not affected by the magnetic field.

However, the sensitivity of the solid-state detector is not independent of energy over the lower energy region. Thus, for measurements extending to the lower energies, necessary to determine the intensity of the total beta spectrum, an end-window proportional counter with a loop anode was used. The counter window was an unsupported aluminized Zapon film with a thickness of approximately 75 $\mu g/cm^2$.

Previous measurements[6–8] have shown that there is no inherent energy dependence in the sensitivity of the proportional counter over the region of interest. Control experiments described in Subsection D have also shown that the sensitivity of the solid-state detector is independent of energy over the range of the high-energy spectrum.

C. Sources

The Fe^{59} experiments reported in this paper were performed with several shipments of high specific activity Fe^{59} from Oak Ridge. Many thin sources of the Fe^{59} were used in the study of the low-energy end of the beta spectrum; several somewhat thicker and more intense sources were used to study the highest energy group.

The thin sources were prepared by liquid depositing approximately 2 mC of $FeCl_3$ on a thin Zapon film ($\leq 10\ \mu g/cm^2$) which was then covered with a similar film. The source was spread over 4 mm by 25 mm and was defined by insulin[11,12]. The source thickness, which was estimated to be ~ 0.04 mg/cm², had negligible distorting effect on the interpretation of the data in the low-energy region.

Since more intense sources are necessarily thicker, some difficulty was encountered when a thicker deliquescent $FeCl_3$ source was tried in order to study the high-energy region. For this reason, a nondeliquescent iron-hydroxide source was used.

This source of approximately 20 mC was prepared, *in situ*, on a 0.00005-in. Au backing by the addition of a drop of NH_4OH. The Au backing was used in order to withstand the heat required to drive off the excess ammonium salt. The source was then covered with a Zapon film of 20 $\mu g/cm^2$. Since the measurements with this source were restricted to the high-energy region, the 0.6 mg/cm² source thickness had no distorting effect on the interpretation of the data. Control experiments were performed to determine the effect of the 0.00005-in. Au backing.

The decay of each source was followed over a period of at least 50 days and the half-life was found to be 45±3 days. It is significant to note that the measurements are in agreement with most of the other half-life determinations and definitely do not agree with the half-life value (63.1±0.8 days) recently reported by E. Fuschini *et al.*[13] In addition, no evidence for any short half-life contaminants was found in the spectrometer data or in gross decay curves checked against a uranium standard.

D. Control Experiments

As a further check on the sensitivity of the solid-state detector, the spectra of Ga^{66} and Y^{91} were measured in the spectrometer by both the semiconductor detector and a proportional counter. The ratio of the counting rate per momentum interval, $N(H\rho)$, measured with

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

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the solid-state detector to that measured with the proportional counter for the same source, plotted in Fig. 1, shows that the sensitivity of the solid-state detector is independent of energy over the region of interest. It is interesting to note that no correction is needed for any variation of the solid-state detector's sensitivity becasue of the backscattering from its surface.

The extreme criteria for the source thickness and backing which were demanded for the investigation of possible small deviations in the shape of beta spectra[6,7] may be somewhat relaxed in the measurement of the highest energy group. The source thickness used for Fe^{59} has been found to have no effect on the spectral shape in this region. Also, at such energies, backscattered electrons from the 0.00005-in. Au backing should have no influence on the interpretation of the data. To verify this, two 1 mCi sources of Y^{91} were prepared. The first was liquid deposited on a 0.9-mg/cm^2 Mylar backing and covered with a (20 μg/cm^2) Zapon film. The second was liquid deposited on a 0.00005-in. Au backing and covered with a similar film. The ratio of the counting rate with the Au backing to that with the Mylar backing is plotted as a function of energy in Fig. 2. There is no measurable difference between the distribution obtained with the Au backing and that obtained with the Mylar backing for energies above ~650 keV. Previous experiments comparing Mylar backed sources with sources on extremely thin Zapon backing have shown that the Mylar has no effect on the energy distribution above 100 keV. We can, thus, conclude that there is no distortion of the high-energy spectrum of Fe^{59} by any backscattering from the Au backing.

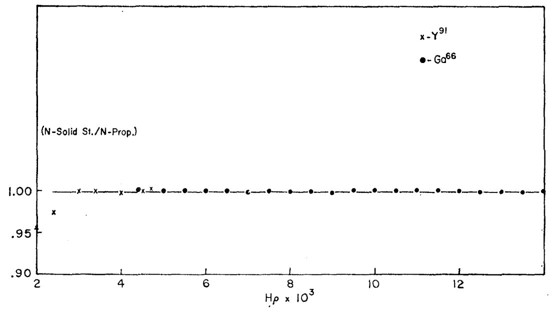

FIG. 1. Relative sensitivity of the solid-state detector. The ratio of the counting rate obtained with the solid-state detector to that obtained with the proportional counter is plotted against $H\rho$. The crosses represent data obtained with an intense Y^{91} source which yielded good statistical accuracy in the low-energy region. The dots represent data obtained with a Ga^{66} source.

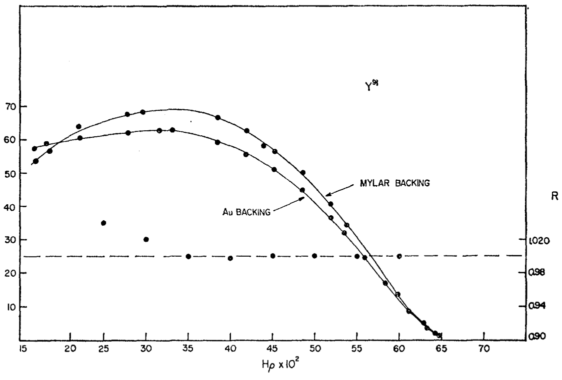

FIG. 2. Comparison of the beta spectrum of Y^{91} from a source mounted on a 0.00005-in. Au backing with the spectrum from a source of comparable thickness mounted on a 0.00025-in. Mylar backing. The right-hand ordinate represents the point by point ratio of intensity of Au-backed to Mylar-backed sources normalized to unity.

Since a 20-mCi source was used in investigating the high-energy region of the Fe^{59} spectrum, additional checks were made to assure that low-energy electrons from the more intense inner groups were not scattered in a way that would distort the measurements in the high-energy region. Hence, a 20-mCi Y^{91} source on a 0.00005-in. Au backing and a 20-mCi Cu^{64} source on a Mylar backing were prepared. Both sources were of a thickness comparable to that of the thickest Fe^{59} source. Figures 3 and 4 are plots of the ratio of $N(H\rho)$ to the maximum value of $N(H\rho)$ vs the ratio of $H\rho$ to the the value of $H\rho$ evaluated at the end point of the beta spectrum for Y^{91} and Cu^{64}, respectively.

Cu^{64} has an allowed spectrum with an end point not too different from that of the inner group of Fe^{59}. Y^{99} has a once-forbidden unique spectrum with an end point very similar to that of the highest energy group of Fe^{59}. Both of these plots indicate that the scattering of electrons beyond the end point falls rapidly to a negligible level.

III. DATA AND RESULTS

Several runs through the spectrum were made with the appropriate sources. These were normalized to the intensity of the first source in the regions where the

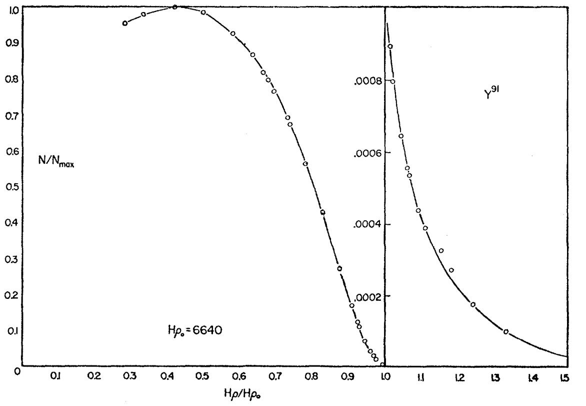

FIG. 3. Measure of scattering beyond end point of beta spectrum of Y^{91}. The counting rate per unit momentum interval is normalized to unity at the maximum of the spectrum. The abscissa is normalized to unity at a value of $H\rho = 6640$, which corresponds to the end-point energy.

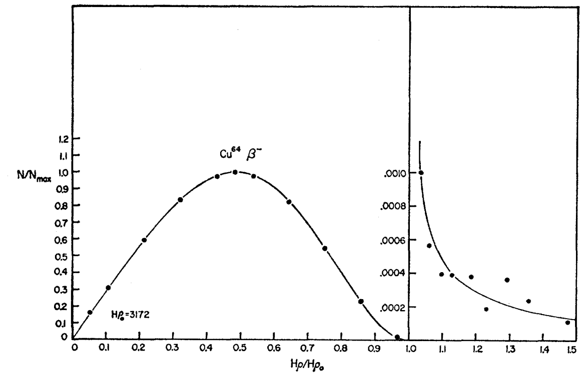

FIG. 4. Measure of scattering beyond the end point of beta spectrum of Cu^{64}. The counting rate per unit momentum interval is normalized to unity at the maximum of the spectrum. The abscissa is normalized to unity at a value of $H\rho=3172$, which corresponds to the end-point energy.

data overlapped. The total beta spectrum of Fe^{59} is shown in Fig. 5. The statistical accuracy of most of the points is 1%.

A Fermi-Kurie (F-K) plot of the data above $W=2.7$ is shown in Fig. 6. The lower plot shows that a nonunique twice-forbidden shape factor linearizes the F-K plot from which an extrapolated end point is obtained which is the same as that used for W_0 in the shape factor. The data below $W=3.18$ are complicated by the K internal conversion line, the photoelectron lines from the Au backing, and Compton electrons ejected out of the source material by the high-energy gamma rays; hence, these data are not used to determine the shape of the high-energy group. The spectrum from $W=3.18$ to 3.34 and from $W=3.55$ to the end point is free from special corrections because of the high resolution of the spectrometer and the lack of scattering.

Figure 7 is an F-K plot of the individual groups obtained by successive subtractions. The shape of the highest energy group was extrapolated to lower energies on the basis of the twice-forbidden nonunique shape. The other groups were assumed to have the statistical shape. It is felt that the effect of the source thickness, and other possible small corrections, renders meaningless consideration of a possible 130-keV group of approximately 1% intensity, which has been reported[4] on the basis of $\gamma-\gamma$ coincidence measurements.

A shape factor plot is shown in Fig. 8 for the highest energy group with an end-point energy of 1.573±0.003 MeV. The points below $W=3.1$ are high, as might be expected, because the gamma transition has an upper Compton electron edge which extends to about that energy. Hence, these points are not used in the fitting of a shape factor. The limits set on the value of the end point are determined by requiring that the shape factor does not "take off" to zero or infinity as one approaches the end point.

From Figs. 5 through 8, one finds that the electron spectrum can be resolved into three groups. The highest energy group at 1.573±0.003 MeV has an intensity of 0.30% (log $ft=10.96$). The intensity of the next group at 0.475±0.003 MeV is 51.2% (log $ft=6.74$). The third group at 0.273±0.005 MeV consists of 48.5% (log $ft=5.92$) of the decays. Self-consistency of the results is checked by comparing the energy of the outer group to the sum of the energy of the first inner group and that of the cascading gamma ray. From the K internal conversion line, the energy of the gamma transition was determined to be 1.101±0.003 MeV. The resultant sum of 1.576 MeV is in excellent agreement with the end

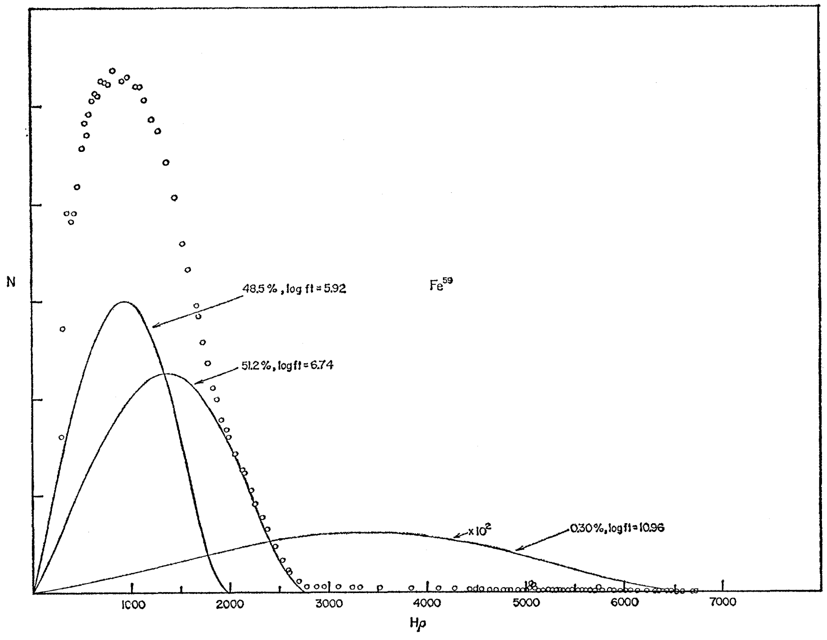

FIG. 5. Beta spectrum of Fe^{59}. The analysis into groups is based on the Fermi-Kurie plots shown in Figs. 6 and 7.

point of the highest energy group. The higher energy internal conversion line was found to correspond to a gamma transition of 1.300±0.003 MeV which also agrees extremely well.

Using the thin window proportional counter, a careful search was made for any intense low-energy internal conversion electrons which might arise from the possible feeding of a low-lying level in Co^{59}. Such a level could alter the interpretation of the decay scheme and result in a change in the comparative half-lives. No internal conversion electrons were found in the energy range from 5 to 50 keV.

IV. DISCUSSION

One of the reasons that prompted this investigation is the fact that a twice-forbidden nonunique theroetical shape factor of the form $p^2+\lambda q^2$ and the experimental shape previously reported are incompatible. Hence, the theoretical predictions, as well as the experimental results, should be reviewed.

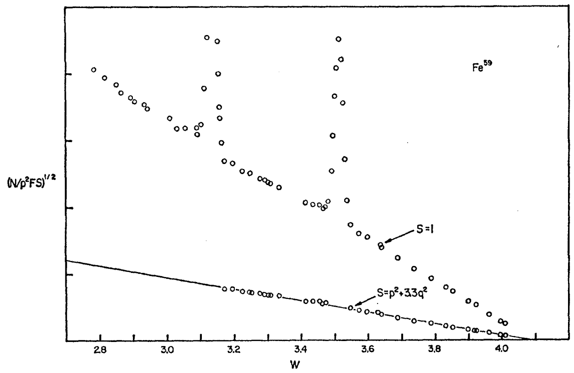

FIG. 6. F-K plot for the highest energy beta group in the decay of Fe^{59}. The lower curve shows the linear plot of the highest energy spectrum using the theoretical shape factor, S, that best fits the experimental data.

The spin of the ground state of Co^{59} has been measured and interpreted as $\frac{7}{2}-$. Using this assignment, the shell model seems to be conclusive in predicting that there is no parity change in the Fe^{59} transition to the ground state of Co^{59}. The thirty-third neutron falls at a place where there is some uncertainty as to whether the level in Fe^{59} should be $\frac{5}{2}-$ or $\frac{3}{2}-$. Since the $2p_{3/2}$ shell is filled by the thirty-second neutron, one might expect the $1f_{5/2}$ state to be occupied by the thirty-third neutron. However, As^{75} with 33 protons and Ni^{61} with 33 neutrons have spins of $\frac{3}{2}$. It appears that the additional pairing energy results in some preference for the formation of a $(2p_{3/2})^3\,(f_{5/2})^2$ state with a resultant spin of $\frac{3}{2}-$ rather than the $(2p_{3/2})^4\,f_{5/2}$ configuration.

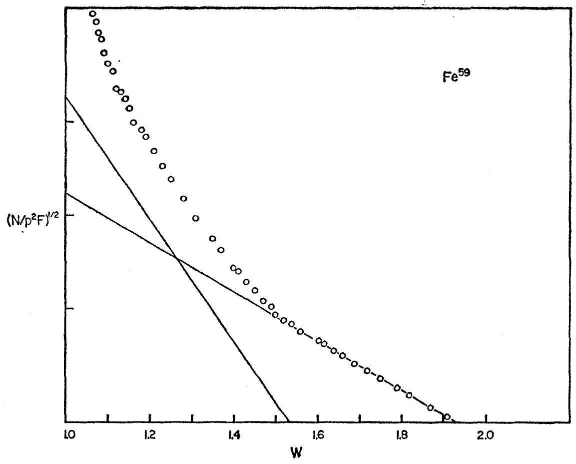

FIG. 7. F-K plot for the lower energy inner groups in the decay of Fe^{59} obtained by successive subtractions.

If the Fe^{59} level is $\frac{3}{2}-$, then the high-energy transition is of the $\Delta I=2$ (no) class. However, if the level were $\frac{5}{2}-$, a spin change of one unit and no parity change would designate that the transition is in the allowed category. In that case, the comparative half-life would have to be interpreted as being high because of some unusual lack of overlap between the wave functions of the initial and final states. There exist, of course, many examples of such behavior. If the transition is sufficiently depressed, then one may expect to observe the influence upon the shape factor of higher order matrix elements, which are usually neglected in the allowed approximation.

If the transition is, indeed, twice forbidden with a spin change of 2, one expects[14] to find a shape factor which can be fitted by the theoretical expression $p^2+\lambda q^2$. It is not very likely that an allowed transition with a spin change of one unit and with an abnormally high comparative half-life will give rise to this same shape factor. The reason is that one expects the presence

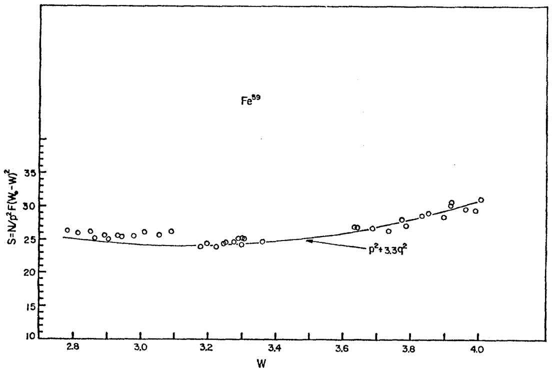

FIG. 8. Shape-factor plot of the high-energy group of Fe^{59}. The solid curve represents the best fit to the experimental data of a theoretical shape factor of the form to be expected for a $\Delta I=2$, no transition. The data at values of W below 3.1 were not used in the analysis. They appear somewhat high because of the Compton electrons.

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[14] See, for example, C. S. Wu, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 11, p. 347.

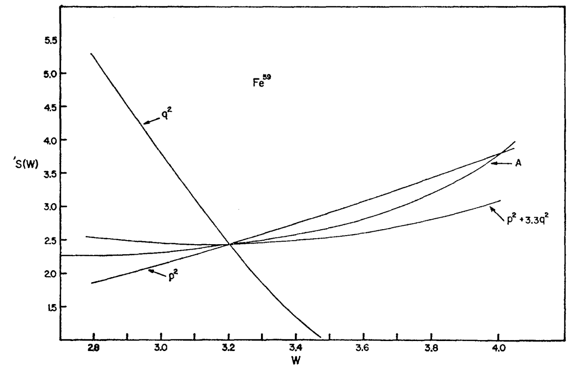

FIG. 9. Shape factor plot for Fe^{59}. Curve A represents the experimental data observed by Metzger. The theoretical shape factor which provides a good fit to the data obtained in the present investigation is shown. It is not possible to fit curve A with a shape factor of the form $p^2+\lambda q^2$ for any values of λ between 0 and ∞.

of the constant term in the "allowed" shape factor still to be quite evident. Therefore, if the Fe^{59} spectrum can be fitted with a shape factor of the form $p^2+\lambda q^2$, this would be very good evidence that the transition is, indeed, twice-forbidden nonunique. However, the experimental curve reported by Metzger is not compatible with a shape factor of the form $p^2+\lambda q^2$ as is illustrated in Fig. 9.

A sufficient number of values were chosen for λ to demonstrate that the experimental and theoretical results are not in agreement. The curves labeled by p^2 and by q^2 in Fig. 9 are the limiting cases for $\lambda=0$ and $\lambda=\infty$. The curve labeled by $p^2+3.3q^2$ in the diagram shows that our experimental curve and Metzger's curve are likewise not in agreement. As Fig. 8 demonstrates, our experimental curve is easily fit by the $\Delta I=2$, (no) theoretical shape factor of the form $p^2+3.3q^2$. Thus, on the basis of this experiment, one can conclude that the shape of the 1.573 MeV transition is consistent with a $\Delta I=2$, (no) shape factor.

The comparative half-life of the Fe^{59} transition is relatively short compared to that of other $\Delta I=2$, (no) transitions. For Na^{24}, Tc^{99}, I^{129}, Cs^{135}, Cs^{137}, and Cl^{36}, such transitions have values in the range $12.5\leq\log ft\leq 13.5$. The value for Fe^{59} is $\log ft=10.96$. Nevertheless, it has been noted[15] that, in comparison with the values obtained for once forbidden transitions, the moments associated with the $\Delta I=2$, (no) decays appear to be smaller by about an order of magnitude. Perhaps it is, therefore, not unreasonable to find one out of seven transitions to have a comparative half-life closer to the theoretical expectation.

One expects to obtain a better constant than ft by considering the energy dependence of the shape factor in integrating over the spectrum. Thus, the value of $\log[(\alpha Z/\rho)^2\, p_0^2\, ft]$ is found to lie between 15.7 and 15.9 for all transitions except Cl^{36} and Fe^{59}. For Cl^{36} the value is 16.2, while for Fe^{59} it is 14.4. The value for Cl^{36} can perhaps be justified on the basis that it is an even nucleus. This argument, of course, does not hold for Fe^{59}.

In conclusion, the shape of the highest energy beta transition of Fe^{59} has been measured. The theoretical shape factor $p^2+3.3q^2$ is consistent with the data; hence, the possibility that this is a $\Delta I=2$, (no) transition cannot be excluded. Furthermore, the beta spectrum consists of the following three groups:

(1) 1.573±0.003 MeV, 0.30%, $\log ft=10.96$
(2) 0.475±0.003 MeV, 51.2%, $\log ft=6.74$
(3) 0.273±0.005 MeV, 48.5%, $\log ft=5.92$

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[15] E. Konopinski, *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. 10, p. 310.